SESSION 4: REACTORS, FUELS AND TARGETS

CHAIRMAN: T. MUKAIYAMA (JAPAN)

MOTIVATION FOR TRANSMUTING LONG-LIVED RADIOACTIVE PRODUCTS

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Abstract
In the Netherlands the efforts on waste transmutation are coordinated in a research programme called RAS. One of the aims of this RAS program is to inform the public and advise the authorities on methods for transmutation/conditioning of nuclear waste, and on techniques which are being developed. Any new way to treat waste should of course not lead to significant risks for the present population. Small risks might be accepted, but these should sufficiently be compensated for. Benefits for the present generation are related to the better exploitation of the full energy content of the actinides, which will reduce fuel costs and waste streams from mining as well as from spent fuel. Future generations might profit from the fact that the waste has been cleaned from actinides and that proliferation risks are eliminated. Another benefit could be that transmutation also could lead to a reduction of dose-risks by leakage of mobile elements such as Rn-222 and the metalloid fission products like technetium and iodine. It is shown in this paper that the balance of benefits and risks is quite different for long-lived fission products than for actinides.

1. GENERAL CRITERIA TO JUDGE TRANSMUTATION SCENARIOS

Spent fuel from light water reactors contains about 100 kg of long-lived fission products, about 300 kg of toxic actinides and 900 kg of short-lived fission products for each GWe year generated. For the actinides the mass balance depends strongly on the fuel cycle history, for the fission products this is less so. About one third of the long-lived fraction of fission products consists of geochemically mobile fission products Tc-99 and I-129. Partitioning and transmutation (P & T) of fission products will have different benefits and risks than P&T of actinides, as may be judged by means of the following three criteria:

1) *Exploitation of the full energy content of the actinides will reduce fuel costs and streams of waste from mining as well as from spent fuel.* A higher long-term toxicity in the fuel and a change in reactor parameters (delayed neutrons, temperature- and void- dependent effects) could be the price one has to pay for this benefit. If the corresponding risks would be low enough, one hopes to reach acceptable waste-storage strategies, in which the life-time of long-lived radioactive components is shortened and in which actinides would be harmless in any waste-disposal scenario even if this scenario would be falsified by human intrusion.

2) Some of the untreated actinide waste transforms itself into a manageable form of partly fissile matter by decay of short-lived products, and disposal sites might become attractive actinide mines. *Future proliferation risks could be eliminated by transmutation.* Actual proliferation risks

during the P & T process itself could be the price for this benefit, and P & T processes could only generate broad public acceptance if risks and costs are controllable.

3) *Dose-risks by leakage of mobile elements like Rn-222 and the metalloid fission products technetium and iodine could be reduced.* The price which should be paid relates to the avoidance of ecological risks of the P & T process itself [1], as one should not spread radiotoxicity by procedures such as machining or spilling of solvents. It seems a sensible strategy to distribute the efforts in such a way that the total dose-risk is minimised for a given amount of spent fuel.

Above mentioned criteria should be related to a cost-risk analysis. For oil and coal the loss of human lives to the present generation lies between 1 and 10 per year for a production of one GW(e). Casualties are lower for nuclear energy [2], and the long term risks ought to be also lower than long-term risks from the use of fossil fuel. As each scenario will have a price, it could be an approach to see this price expressed in an amount of dollars needed to save a life. To protect contemporary individual radiological workers from harmful overdoses, one might offer a price of for example about 100 000 US $ for each man Sv avoided [3]. If however risks for any individual are low enough, collective risks will only be handled according to a strategy called ALARA (As Low As Reasonably Achievable), after all it is considered to be "more important to avoid one man to be hit by a bag of barley than to avoid every one to be hit by some grains".

2. TRANSMUTATION of ACTINIDES in REACTORS and SUB-CRITICAL SYSTEMS

In thermal incinerators some actinides could be used as fuel. Commercial thermal reactors are however energy and plutonium producing entities, which are not dedicated to waste transmutation. These LWRs may of course transmute Pu-239, and application of plutonium in MOX fuel for electricity generating LWRs will clearly diminish its growth (see fig.1). However the amount of transuranium elements will increase anyway due to continuous capture of neutrons in U-238. It is difficult to leave out this U-238 because its capture process is essential to the economy and safety of LWRs: it increases the reactivity swing of the fuel by breeding fissile plutonium, and the Doppler broadening of capture resonances will keep temperature coefficients negative even for large systems. As long as U-238 is the main component in the fuel, the accumulated plutonium mass will increase in any recycling scenario. Due to the build-up of even plutonium isotopes the multi-recycled fuel will become much less fissile in thermal spectra, whereas it remains fissile in fast spectra. If no external neutron source would be applied, an LWR would require an increasingly higher fissile enrichment, and this would create a less economical situation. High concentrations of even-N transuranium isotopes in LWR fuel would also give safety problems due to the fact that the number of formed fission neutrons in such multi-recycled fuel increases with a hardening of the neutron spectrum (see table 1). This problem should be solved, either by integrating the fuel with the moderator or by applying an external neutron source to a sub-critical system, otherwise the reactor could become a prompt critical fast reactor after an accidental loss of its moderator! Thermal systems will have as a further disadvantage that the toxicity in the U/Pu cycle increases continuously due to the growing in of heavier actinides. One would end up with a toxic mixture, in which the long term radio-toxicity is almost entirely determined by americium (thousands of years) and neptunium (millions of years). It is shown by J.L. Kloosterman and W.J.M. de Kruijf in a paper for this meeting that an extremely high neutron fluence (of the order of 10^{22} cm^{-2}) is needed to reduce the long term toxicity in an americium sample.

Fast incinerators have been proposed for the transmutation of actinides. Possibly the breeding mantles of fast incinerators could be replaced by moderated sub-assemblies, which could then also be used for the transmutation of fission products. From several studies it has been concluded that critical fast reactors could only have a minor-actinide fraction in the fuel up to 5%. If the transuranium concentrations in the fuel would be higher, some reactivity problems could appear, and the number of delayed neutrons per fission could become too small, especially for curium isotopes [5]. Reactivity problems could either be enhanced by positive voiding-feedback or by insufficient Doppler-damping of temperature excursions. If critical reactors are not considered safe enough one could consider systems with $k_{eff} < 0.95$. External neutrons could then be supplied by another reactor (possibly a fusion reactor), or by a spallation source with a GeV proton beam. Such external

sources could even provide enough excess neutrons for transmutation and generation of energy at the same time, and advanced reactor systems could be developed to reduce not only the amount of plutonium, but also to reduce the very long-term toxicity due to americium and neptunium [6].

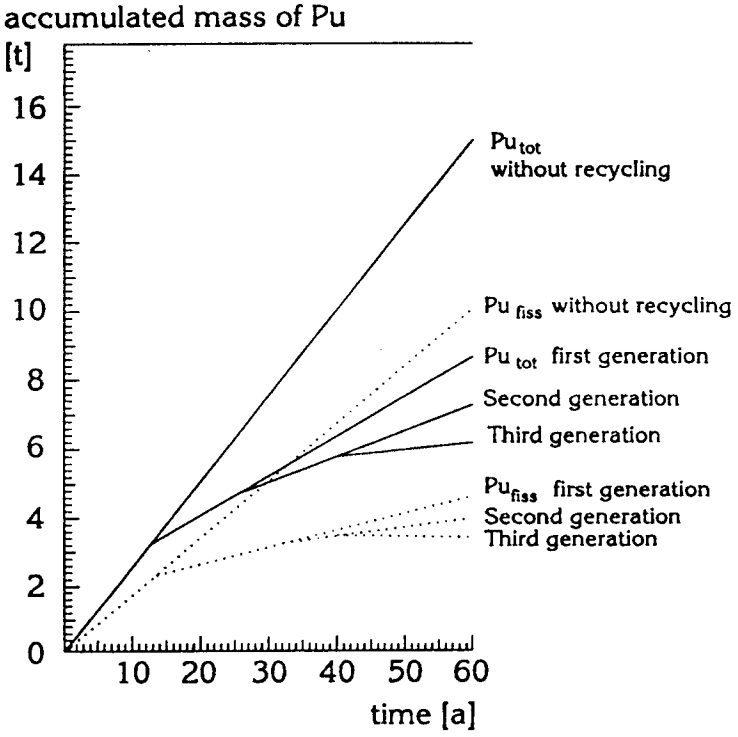

Fig.1: Evolution of the total mass Pu_{tot} and of the total fissile mass of plutonium Pu_{fiss} in a multi-recycling PWR scenario, 1300 MWe, 45 MWd/kg, and a recycling time of 12.5 years [4].

Table 1: Average amount of fission neutrons for a single neutron, which has been absorbed in fuel pins of Light Water Reactors or Liquid Metal Burner Reactors. This amount is given for several actinides.

Nuclide	LWR	LMFBR
U -235	1.98	1.92
U -236	0.11	0.43
U -238	0.29	0.39
Np-237	0.05	0.49
Pu-238	0.25	2.02
Pu-239	1.84	2.29
Pu-240	0.02	1.24
Pu-241	2.20	2.45
Pu-242	0.05	1.10
Am-241	0.04	0.46
Cm-242	0.64	1.33
Cm-243	2.94	2.99
Cm-244	0.20	1.58

Each proton of high energy (one GeV or more) will liberate dozens of spallation neutrons from a heavy-metal target [7]. Either directly or after moderation these neutrons can be used to transmute actinides or fission products. In a slightly subcritical booster setup the number of neutrons might be multiplied with a factor $k_{eff}/(1-k_{eff})$. Cost and safety aspects of neutron generation and the amount of excess neutrons both depend on the value of k_{eff}. For small values the safety of the system might be best; on the other hand costs for electricity and capital investment in the accelerator would be less if k_{eff} would be close to one. In comparing the different proposed systems, the safety-cost balance for actinides is quite different than for fission products. First of all actinides are a potential source of neutrons [8] and secondly for actinide-transmutation by fission fast neutrons are most suitable, whereas fission-products rather transmute by capture of moderated neutrons [9].

3. TRANSMUTATION of FISSION PRODUCTS

In contrast to the situation for the actinides any thermal reactor could be used to diminish the toxicity of fission products. In principle even commercial LWRs could be used to get rid of the technetium and perhaps also of the iodine-129. An efficient transmutation at low flux would however require huge loadings of waste, additional fuel enrichment and extra recanning and reprocessing efforts. Reactors with a somewhat higher flux (HWRs like CANDU) and with possibilities for refuelling on-line, are therefore being studied. High-flux thermal reactors could be entirely dedicated to transmutation, and moderated sub-assemblies of high flux fast reactors might also have potentials, as is shown in a contribution of Kloosterman and Li to this meeting. For a large scale transmutation of long-lived fission products a feasible and economic technology is however not yet available, and there still are several limitations related to safety and cost aspects.

Fission products with low cross sections would require extra neutrons [7,8], and one considers application of high-energy proton accelerators as neutron boosters. Any accelerator-based system will however be less efficient than a corresponding reactor system without a booster, which can be seen as follows: One 1.5 GeV neutron produces 30 neutrons in a lead/tungsten target. Suppose the accelerator efficiency is about 50% (a very optimistic statement). In this case the price of one thermal neutron in terms of electrical energy is 1500/(30*0.5) = 100 MeV. As this electrical energy had to be derived from thermal energy one would have required about three times more energy. It is unrealistic to assume that each neutron will transmute a nucleus, but even then one would need an equivalent of at least 300 MeV thermal energy to transmute one nucleus. If this 300 MeV would have been generated by means of a nuclear reactor, this would mean that more than one fission in the reactor is needed to transmute one nucleus at the accelerator. A direct transmutation in the reactor seems more economic because then also one free neutron might become available for each fission in the reactor itself. Reactor-based transmutation of a technetium nucleus is clearly more direct because the 200 MeV, which is generated by fission, will still be useful to generate electricity, which is no longer needed for the accelerator. In hybrid accelerator-reactor combinations each neutron from the accelerator-target system might produce again up to ten new fission neutrons in a sub-critical assembly, and transmutation costs of such systems could be in between that of pure accelerators and that of critical reactors [9,10]. An advantage for accelerator scenarios could be the good neutron economy [8], which would allow for a better loading and lower handling costs.

4. REDUCTION OF DOSE-RISKS FROM LEAKAGE

As long as the integrity of a disposal site is guaranteed, *long-lived fission products will determine the leakage-risks* (products of toxicity and mobility). For an unperturbed granite repository [11] table 2 shows the risk due to spent LWR fuel in a once through scenario [9]. It is seen from this table that the unspent uranium would give the main residual actinide contribution, and that Tc and I-129 dominate dose-risks. Diffusion of anions of iodine and per-technate in ground water is more rapid than flow of the ground water itself, as is now again being realized.

Any collective dose rate could be compared with the natural rate from radon, and even for the most relevant long-lived fission product Tc-99 the collective dose-risk is only marginal. If it can be assumed that in due time the Tc will distribute itself evenly in time and space over the world, a value of 10^{-12} Sv/year would be the order of magnitude for the personal average dose-rate due to electro-nuclear production of one GW(e) year. This value corresponds to about one part per billion in terms of the natural radon dose-rate.

TABLE 2: TIME-INTEGRATED LEAKAGE-DOSE DUE TO SPENT LWR FUEL
(direct storage of spent fuel, due to nuclear generation of one GW(e) year)

Period :	One million years	Hundred million years
Nuclides :		
Tc-99	98 %	46 %
I -129	2 %	1 %
Cs-135		24 %
U -235		6 %
U -238		14 %
Np-237		5 %
Pu-239		4 %
man Sv	9000 *)	20 000 *)

*) Collective dose for the global population. The average yearly individual dose may be obtained by dividing by the affected number of people and the indicated period.

Arguments on the collective leakage-dose should be treated with some caution. First of all it is questionable whether the risk should be ranked as in table 1 (a collective integrated dose from leakage out of a repository). If one would for example rank according to the highest possible individual dose, the I-129 risk could dominate for repositories of clay [12] or rock salt [9]. Local dose-risks are almost entirely due to uptake of iodine in the thyroid. This gland usually contains about 10 mg of iodine, and risks could be made marginal by diluting the I-129 isotopically with natural iodine. In most transmutation scenarios collective leakage doses are mostly due to U-234.

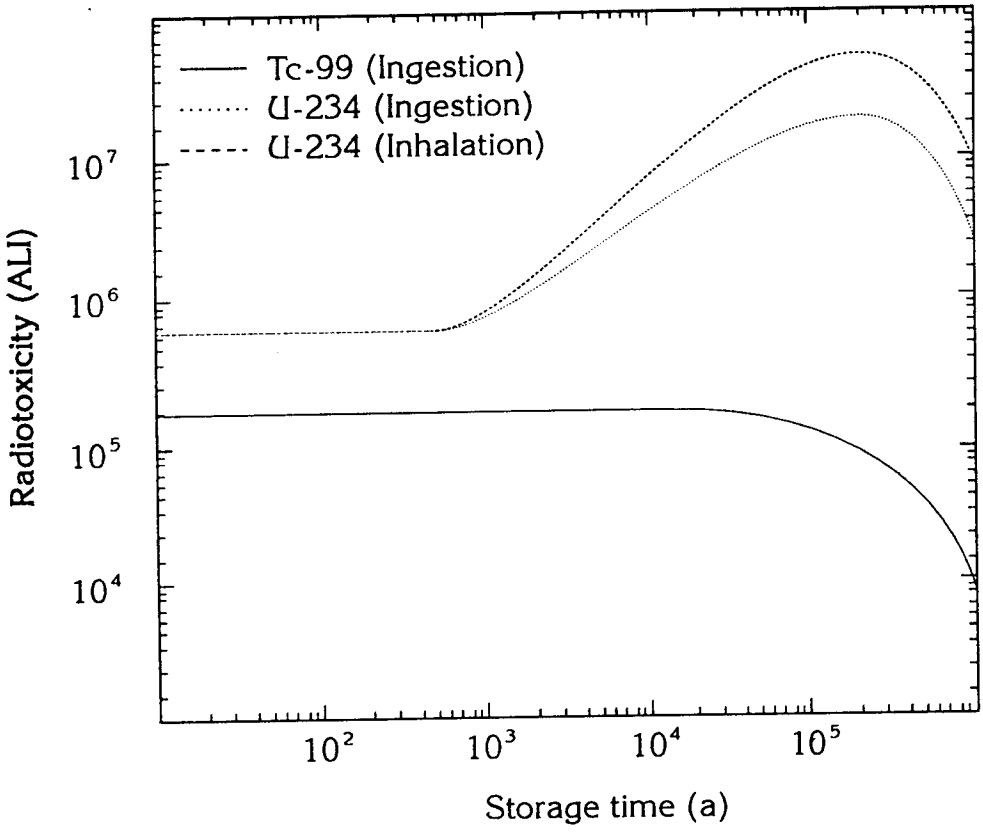

Fig.2: The radio-toxicity of Tc compared to that of U-234 and its daughters. The amount of material (9.6 kg of Tc-99 and 2 kg of U-234) is related to the yearly release of the Dutch reactor at Borssele (0.5 GW(e)).

Figure 2 shows for comparison the radio-toxicity of the Tc and the U-234, as these are released yearly by the Dutch reactor at Borssele (0.5 GW(e)). It should be noted that the dose-risk follows the radio-toxicity as given in the figure 2 if it is multiplied by the fraction of the material, which will reach the human biosphere. This might amplify the relative hazard of the U-234 considerably, as the geo-chemical mobility of radon is higher than that of technetium, and the reprocessed uranium (REPU) will only be subjected to shallow land burial in present scenarios. Therefore it is clear that the U-234 and its radon emanation leads to a much higher dose-risk than the corresponding amount of technetium, especially after a few thousand years when the extremely mobile radon-gas will be liberated by the radium, which has been formed in the mean time.

Due to the fact that U-234 will in the long run lead to mobile Rn-222 one could for example double the local dose due to emanations for a few hundred thousand years by spilling only a few hundred milligrams of U-234 or one of its precursors into the soil over a surface of only a few square kilometres. This isotope of uranium, which also occurs naturally (with an isotopic abundance of 0.0055 %) is responsible for most of the present radiation dose to mankind. Its most dangerous daughter Ra-226 has a half life of 1600 year, and about 60 t of radium in the

soil emanates almost 6 litre of radon each day, which builds up an equilibrium value of about 20 litre of radon in the total biosphere of our planet. This tiny amount nevertheless contributes for about 60 % to the total radiation dose, which averages to about 10^{-3} Sv per person yearly and this leads to an estimate of the collective dose of the order of $5 * 10^6$ man Sv/year. It would therefore be environmentally very unwanted if the present amount of U-234 would increase, and it might very well be that build-up of U-234 in the soil gives a much higher dose than the so much feared build-up of heavier actinides.

5. RISKS IN DIFFERENT TRANSMUTATION SCENARIOS

There are long-term dose-risks in regular transmutation scenarios, but also in end-scenarios, in which production of nuclear energy would be ended and large quantities of fissile waste are to be disposed of at once. Similar situations as in end-scenarios would occur if large quantities of weapon plutonium have to be transmuted [13]. Procedures would depend on the criteria to be adopted. In case of ultimate safeguarding requirements one could pollute the Pu-239 with less fissile material by irradiating it with thermal neutrons. This renders the plutonium less useful as a weapons material, and makes it hard to divert. This procedure would however increase the toxicity and the Pu-238 content considerably, and one would need to store the irradiated plutonium for a long time, and possibly in an irreversible and geologically secured way. As this storage procedure would hardly comply with present public demands for waste disposal, any acceptable end-scenario should be terminated by the fission of its actinide waste. This could only be achieved by means of an extended use of fast reactors, which however also yield increasing amounts of potential radon-emitters in the waste such as U-234, Pu-238, or Cm-242. It seems therefore very likely that non of the present scenarios for reduction of the actinides fulfils the third criterium mentioned upfront of this paper. If this situation cannot be improved, there might be very little motivation left to transmute Tc-99 and I-129 in end-scenarios. Release of potential radon emitters in the waste should be guarded against in any scenario, and it might be conceivable to recycle the spent uranium (REPU) together with the other actinides. The scenario in which it is most likely that all the uranium will be recycled, is the Th/U breeding cycle, which therefore will be treated below:

Relative dose risks and proliferation risks for the Th/U Cycle

Competing demands of safety, economy and non-proliferation also apply to the thorium cycle, in which capture breeds Pa-233 and the fissile U-233 is formed by the β-decay thereof. By parasitic capture in Pa-233 and in U-233 some U-234 will be formed, and after many cycles the isotopic dilution of the U-233 would reduce the risk for proliferation [14]. In this respect the Th/U cycle would be safer than the U/Pu cycle, in which the recycled plutonium is always considered to be weapons grade as soon as it can be purified chemically and traces of for example americium can be removed. On the other hand the U-233 as produced in the first few Th/U cycles is also weapons grade. Further it is possible to obtain isotopically pure U-233 by decay of Pa-233 in an external cycle. In the equilibrium fuel the rate of formation of Pa-233 by neutron capture is equal to its rate of decay into U-233 ($T_{1/2}$ = 27 d). Therefore the Pa-233 concentration will be an increasing function of the neutron flux in the fuel, and especially for high flux systems parasitic capture will remove many neutrons. Therefore it has been proposed to apply either an external neutron source or an extra initial enrichment of the fuel. Especially high-flux systems would need these fissile additives and/or a powerful accelerator to provide for the extra neutrons [13], and even then these systems are to be refuelled frequently and on-line partitioning of the Pa-233 is needed in order to reduce parasitic absorbtion. As was discussed above partitioning of Pa-233 leads to pure weapons grade U-233, and this procedure would require special controls at the facility. Apart from the proliferation problems one has the problem of the high radiation field at the reprocessing factory. Unless regular clean-ups can be performed, high radiation levels at reprocessing facilities result from the hard (2.6 MeV) gamma radiation from Tl-208, as produced at the end of a chain of α and β-decay according to: U-232 -> Th-228 -> ····· -> Tl-208.

In a low-flux system only a small external neutron source [14] or the addition of little extra plutonium to the fuel could compensate for capture losses in Pa-233, and the above mentioned on-line reprocessing would no longer be required. This is important, because as was discussed above, spilling of U-234 should be minimized. Application of an extremely long burn-up, possibly with a regular recanning of the fuel and an increased accelerator power, could limit the U-234 spilling. Nevertheless it should be realized that the isotopic contents of U-233,234 mixtures could range from 30-50 % in the equilibrium fuel. Because dose-risks due to Rn-222 might dominate over dose-risks from any actinide in the long term, *the Th/U cycle might give a very high long-time dose-risk due to radon emanation especially if one ever decides to stop the cycle. Any scenario in which the waste from the back-end of the Th/U cycle would come to the surface either by intrusion or by accident, would for example dwarf the technetium risk, unless gas tight disposal of the remains of U-234 is considered to prevent the radon to reach the atmosphere.*

6. CONCLUDING REMARKS and RECOMMENDATIONS

Transmutation of the existing plutonium is priority number one. After all proliferation risks are most clear for plutonium, and possibilities for future mining for this element should be eliminated to prevent very long lasting proliferation risks. No exclusive LWR-scenario has yet been found, which entirely solves the plutonium problem by transmutation.

Reduction of minor-actinides is priority number two. After all these actinides contribute for about 10 % to the total long-term toxicity of once used spent nuclear fuel. Recycling in PWRs will only increase the minor actinides. Fast reactors will probably be most suited to reach a high fission rate in a high neutron flux. Safety aspects of fast reactors, which are loaded with minor actinides could possibly be controlled by means of external neutron sources.

Criteria on the reduction of dose risks disfavour transmutation scenarios in which the waste will be contaminated with U-234 or with one of its precursors, unless a disposal method is applied that prevents radon emanation from the waste to reach the atmosphere.

Reductions of collective dose-risks, which are far below the natural dose-risks seem at first sight to be of less relevance. In proper disposal scenarios the world- and time-integrated collective dose from fission products is less than 10 000 man Sv for each GWe year, and thereby less than a fraction 10^{-9} of the natural dose risk. Resistance against the dumping of low-level nuclear waste into the ocean has shown that there are incentives to reduce the dose even below such marginally small values. These incentives seem hardly motivated by the wish to reduce dose risks to human beings, and in this respect it is illuminating to recall the argument on the dose risk from the fission product I-129, for which other people argue that "isotopic dilution with natural iodine should reduce the highest individual doses". As long as no agreement exists on existence of safe thresholds for dose-risks, priorities are a matter of taste, and are therefore subject to changes.

ACKNOWLEDGMENT

This project has largely been executed as a participation in a CEC strategy study on nuclear waste transmutation, and the CEC is acknowledged for its support.

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IMPACT OF PLUTONIUM AND AMERICIUM RECYCLING IN PWR ON MOX FUEL FABRICATION

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Abstract

In a study with European partners for the Commission of the European Community, BELGONUCLEAIRE examines the implications of recovering americium at fuel reprocessing and recycling both plutonium and americium as MOX fuel in light water reactors.

The effect on the neutron physics of a 900-MWe PWR, operated by quarter-core reloadings up to a burnup of 45 GWd/t, with a quarter of the assemblies fuelled with MOX, is checked with respect to the reference strategy of recycling Pu only.

At the first recycling step with Pu and Am, both supposed recovered at reprocessing, a 10 % Pu content is already needed to guarantee the burnup : with Pu alone, this was 8 % ; the 10 % content level was necessary at the second recycling only. The raise in Pu content due to the addition of Am will severely limit the number of recycling steps allowable.

In the refabrication plant, the mixing of Pu with Am, as an extrapolation of the present MOX fabrication practice, will need some addition of shielding to the blending glove box ; this extra-shielding, while costly, seems nevertheless feasible and acceptable.

INTRODUCTION

In the frame of a joint study for the Commission of the European Community (CEC, DGXII), relative to strategic and technical aspects of nuclear waste transmutation [1], BELGONUCLEAIRE examines the implications of recycling plutonium and americium in the form of MOX fuel in light water reactors. First, the effects on reactor physics of such a strategy with multiple recycling steps are calculated, then the problems of MOX fuel refabrication are identified with reference to the present operation conditions of the MOX fuel fabrication plant of BELGONUCLEAIRE in Dessel, Belgium [2].

The hypotheses which have been assumed for a multiple recycling of homogeneous mixed-oxide fuels containing americium in addition to Pu will be given ; the multiple recycling of Pu alone is taken as a reference case in this study.

The physics aspects are first dealt with ; these aspects are deserving further verification in a close cooperation with ECN, Petten [3].

The paper is then centred on the MOX fuel fabrication aspects related to radiation dose protection in the MOX fuel plant. This part of the study offers some similarity with a preceding paper published in cooperation with EDF, which had considered that Pu and minor actinides could be recycled in fast reactors [4]. A major difference is that, while the former study had considered also the recycling of neptunium and curium, the present one is concentrated on the effect of adding americium only to plutonium, in a more realistic or shorter term approach, provided that chemistry research to recover americium proves successful.

HYPOTHESES FOR RECYCLING IN LWR

The reactor assumed for recycling is a 900-MWe PWR, characterized by a quarter-core fuel management with an average discharge burnup of 45 GWd/t. The MOX fuel is designed to be equivalent in energy to UO_2 fuel enriched to 3.7 % U235. At discharge all assemblies, loaded with UO_2 or with MOX, have been irradiated 4 calendar years at a load factor of 75 % in the average.

The lead times in the fuel cycle are four years for core irradiation, three years for cooling up to reprocessing, and two years for refabrication and transport before core reloading.

The multiple recycling strategy in the reference Pu core is as follows :

- in the first stage, MOX 1, 141 kg of MOX fuel are burnt in parallel to 859 kg of UO_2 fuel ; these 141 kg correspond to the mass of Pu which can be recovered from 1 tonne of UO_2 spent fuel from the initial PWR operation ; these 141 kg are a result from the calculations done, and they correspond well to the frequently quoted ratios of one MOX assembly for 7 initial UO_2 assemblies ;

- it is assumed that at the following reprocessing operation, the Pu quantities out of this one MOX assembly and of six UO_2 assemblies are mixed together : insofar, the Pu composition available for recycling is not the Pu from first MOX generation, but a blend ;

- on these premises, our calculations indicate that in the second stage, MOX 2, 167 kg of MOX fuel can be burnt together with 833 kg of UO_2 fuel : there is now one MOX assembly, with degraded Pu, to be blended with 5 spent UO_2 assemblies, and so on.

This is a simplified simulation of what could occur in reality in a whole reactor park, without considering the influence of lag times. A detailed representation of lag times, reactor availabilities and fuel cycle plant peculiarities has been made in the parallel CEA study [5].

SOME RESULTS OF PWR PHYSICS CALCULATIONS (Pu CORE)

From the core physics calculations, some salient results only will be presented, so as to make clear the approach followed. These results have been obtained using the WIMS code package with the data library WIMS-86, with cross-section improvements for the Am and Cm isotopes.

These results are to be considered as preliminary, because a second series of calculations is underway, in cooperation with ECN, Petten [3], using the most recent JEF2 cross-section data.

Three types of results are given below, concerning, respectively, the Pu enrichments needed, the quantities of Pu reprocessed and blended, and the isotopic composition of Pu available for refabrication. In fact, only the latter ones are directly used in the second part of the study. The latest time allowed for refabrication, 2 years after reprocessing, is assumed : this corresponds to a large build-up of Am^{241} in Pu.

TABLE I

MULTIPLE RECYCLING OF Pu IN LWR

MAIN PHYSICS RESULTS

Successive MOX Recycling Steps	(UO_2)	MOX 1	MOX 2	MOX 3
(Pu+Am) content in MOX fuel (%)	-	7.8	10.4	11.8
Quantities of Pu recovered for 1 tonne of heavy metal at latest refabrication time (kg)	11.0	17.4	22.2	25.9
Isotopic composition of Pu+Am for the next refabrication (%):				
Pu 238	2.3	3.0	3.6	
Pu 239	53.2	47.0	44.0	
Pu 240	24.7	28.2	30.2	
Pu 241	11.8	12.4	12.4	
Pu 242	6.8	7.9	8.5	
Am 241(*)	1.3	1.4	1.4	

(*) from the decay of Pu241 during refabrication time.

It can be seen that the required Pu enrichments grow significantly with the successive recycling steps. They lead to increasing difficulties to keep the MOX loadings compatible to the usual design and safety objectives, so that the third step, MOX 3, probably represents in practice an upper limit for this multiple recycling scheme.

COMBINED Pu+Am RECYCLING : CORE PHYSICS RESULTS

In this case, it is assumed that not only Pu, but also Am can be recovered at spent fuel reprocessing ; recycling in the form of MOX fuel (Pu + Am) is tempted in the same PWR under the same conditions. A recovery yield of 99.5 % for Pu and 98 % for Am has been assumed ; for such orientation studies, such high recovery yield values are representative enough.

The reasoning developed above for recycling Pu alone, is applied again but for the sum of Pu and Am, with unchanged burnup targets. The major results of core physics calculations follow in Table II.

TABLE II

MULTIPLE RECYCLING OF Pu + Am in LWR

MAIN PHYSICS RESULTS

Successive MOX Recycling Steps	(UO_2)	MOX 1	MOX 2	MOX 3
(Pu+Am) content in MOX fuel (%)				
Pu	-	9.8	13.6	16.6
Am	-	0.5	1.1	1.6
Quantities of Pu and Am (kg) for 1 tonne of heavy metal at latest refabrication time				
Pu	10.9	17.9	23.9	29.2
Am	0.6	1.4	2.3	3.0
Isotopic composition of Pu and of Am for the next refabrication (%)				
Pu : 238	2.3	3.5	4.7	
239	53.8	48.2	45.7	
240	25.1	28.4	30.2	
241	11.9	12.2	11.5	
242	6.8	7.7	8.0	
Am : 241	70.6	64.6	63.7	
242m	0.1	0.6	1.0	
243	29.3	34.8	35.3	

With respect to the enrichments needed to recycle Pu only, the enrichments required in this Pu+Am case are still higher. It is doubtful that the second step, MOX 2, will still be acceptable : an additional study, centred on the safety aspects, would be needed to resolve this question of limitation. The first step only, MOX 1 (PuAm) will further be considered.

CRITICAL STAGES IN THE MOX FABRICATION PLANT

In the MOX fuel fabrication plant of BELGONUCLEAIRE at Dessel [2], the front-end stages of the manufacturing process are the most critical ones in terms of radiation protection, since they correspond to handling operations on pure oxide powders.

Currently, PuO_2 powders are received from the reprocessors and first stored ; then they are introduced in a glove box to be milled and blended together with UO_2 powders to produce the so-called primary blend. These dose intensive operations are followed by secondary blending, pressing and sintering, before the sintered pellets are put into fuel pin claddings : dilution first, and canning afterwards, lower the dose rates.

The case of the powder storage had particularly been considered in the previous similar study [4]. Here, this is the case of the primary blending which is retained as the most typical. A sketch of the blending device, schematized for the sake of the calculation is given on Fig. 1.

Neutron and gamma dose rates at a distance of 30 cm from the external glove box wall have been calculated for a given configuration of blending devices (the UO_2 silo is not represented), glove box and shielding, so that the "reference" successive thicknesses surrounding the powders are, in cylindrical geometry :

a) 2 mm steel of the silo bottle ;

b) 100 mm of polythene acting as neutron shield, between two thin layers of 0.5 mm steel ;

c) 10 mm plexiglass, glove box wall ;

d) 36 mm of "KIOWAGLASS", a composite material including 30 % of lead.

Note that b) and d) are additional shields which are not present in the standard fabrication line.

For the primary blending operation, a maximum of 60 kg oxide can be handled, of which 30 % can be PuO_2 (containing a small amount of Am241). All calculations have therefore been run for this maximum quantity of 18 kg PuO_2, kept constant, but with a variable composition as given in Table I (Pu case) or Table II (Pu+Am case).

In the Pu+Am case, it is assumed that AmO_2 is added to these 18 kg of PuO_2.

The total AmO_2 content is for example 0.97 kg for the MOX 1 (PuAm) case. In the MOX 1 (Pu) case, it was 0.22 kg.

DOSE RATE CALCULATION RESULTS

The gamma source at fabrication time is calculated by simulating the successive steps (irradiation, cooling, reprocessing, and time shift up to refabrication), using the code ORIGEN 2 [6] with ad-hoc libraries. The losses at reprocessing are assumed to be 0.5 % for Pu and 2 % for Am.

The dose rates due to gamma rays are obtained using the standard gamma shielding computer code QAD-CG [7] for the 18 energy group spectrum coming from ORIGEN. Build up factors are fitted for the glove box materials.

For the neutron doses, the one dimensional transport programme ANISN-ORNL [8] has been employed with the EURLIB 15/5 cross-section library (15 neutron group and 5 secondary gamma ray ones).

Gamma and neutron fluxes are converted into equivalent dose rates using standard conversion factors.

The computing processes have been validated via OECD benchmarks for spent fuel transport and also by comparison with measurements on MOX sources.

Table III gives the dose rates at 30 cm from the glove box calculated as emanating from the reference PuO_2 powder (MOX 1) and from the variant powders corresponding either to a second Pu recycling step MOX 2 or to a first recycling of Pu and Am.

TABLE III

Impact on MOX Fuel Fabrication
at 30 cm from glove box of primary blending

Case	Dose rates (μSv/h)		
	Gamma	Neutrons	Total
MOX 1	9.7	7.3	17
MOX 2	9.7	9.1	18.8
PuAm	67.4	8.0	75.4
PuAm + 20 mm additional steel	17.5	5.8	23.3

Dose rates are expressed in μSv/h. In the reference MOX 1 case, the total dose rate of 17 μSv/h can be split into 7.2 (primary gamma-rays) + 7.3 (neutrons) + 2.5 (secondary gamma-rays). A value of 20 μSv/h (= 2 mrem/h) is taken as a guiding value for these comparisons, although it is not a real limit in the plant. Indeed, the staff will not stay longer than needed near the glove box of primary blending, according to the ALARA principle.

The results show that the conditions for the MOX 2 fuel fabrication are very similar to those of the MOX 1 fuel. The main neutron source via (α, n) reactions from Pu238, increased indeed marginally only. This is a favourable result of blending the Pu from MOX fuel with the Pu from UO_2 fuel.

On the other hand, the addition of americium at reprocessing, assuming a 2 years delay between PWR fuel reprocessing and refabrication, induces a strong increase of the gamma dose, so that the total dose becomes 4,5 times larger than for the reference MOX case. This would require an addition of steel shielding ; a layer of 25 mm steel is needed to reduce the total dose to a level similar to the reference case. (Table III also gives the calculation results for an addition of 20 mm).

DISCUSSION AND CONCLUSIONS

On the viewpoint of core physics, the effect of recovering americium in addition to plutonium at reprocessing, and of introducing it in the MOX fuel, is that at the first recycling step already a 10 % (Pu+Am) enrichment is needed. In the usual Pu recycling case, this is 7.8 % ; the 10 % enrichment level is needed at the second recycling step only.

Recycling Am in addition to Pu in the MOX fuel limits the number of recycling steps to one instead of two or three. Actually, more refined core calculations would be needed to identify clearly the limits for a safe core operation.

It is assumed that reprocessing takes place for spent MOX assemblies diluted into the bulk of spent UO_2 assemblies. In this way, the quality of the recovered plutonium, while progressively degraded, does not deteriorate too quickly.

This dilution explains why the dose rates at refabrication do not increase markedly with the recycling steps when Pu only is recycled : from the first to the second step (MOX 1 to MOX 2), the dose rates at the considered critical stage of fabrication increase slightly only.

In contrast, changing to a combined Pu + Am recycling strategy multiplies the dose rates by a factor 4.5. An additional steel shielding thickness (25 mm) is needed to bring the dose rates back to the same level.

This extra-shield hinders the fabrication and increases somewhat the cost of operations. Nevertheless, it seems feasible as an extrapolation of standard MOX fabrication conditions.

The justification of such extra-costs is to be found in the attainable reduction of radiotoxicity of the nuclear waste for long-term storage times (over 300 years) [9].

Penalties on refabrication are similar, whether Pu and Am have been recycled once in PWR, like assumed here, or recycled several times in fast reactors, like assumed in [4].

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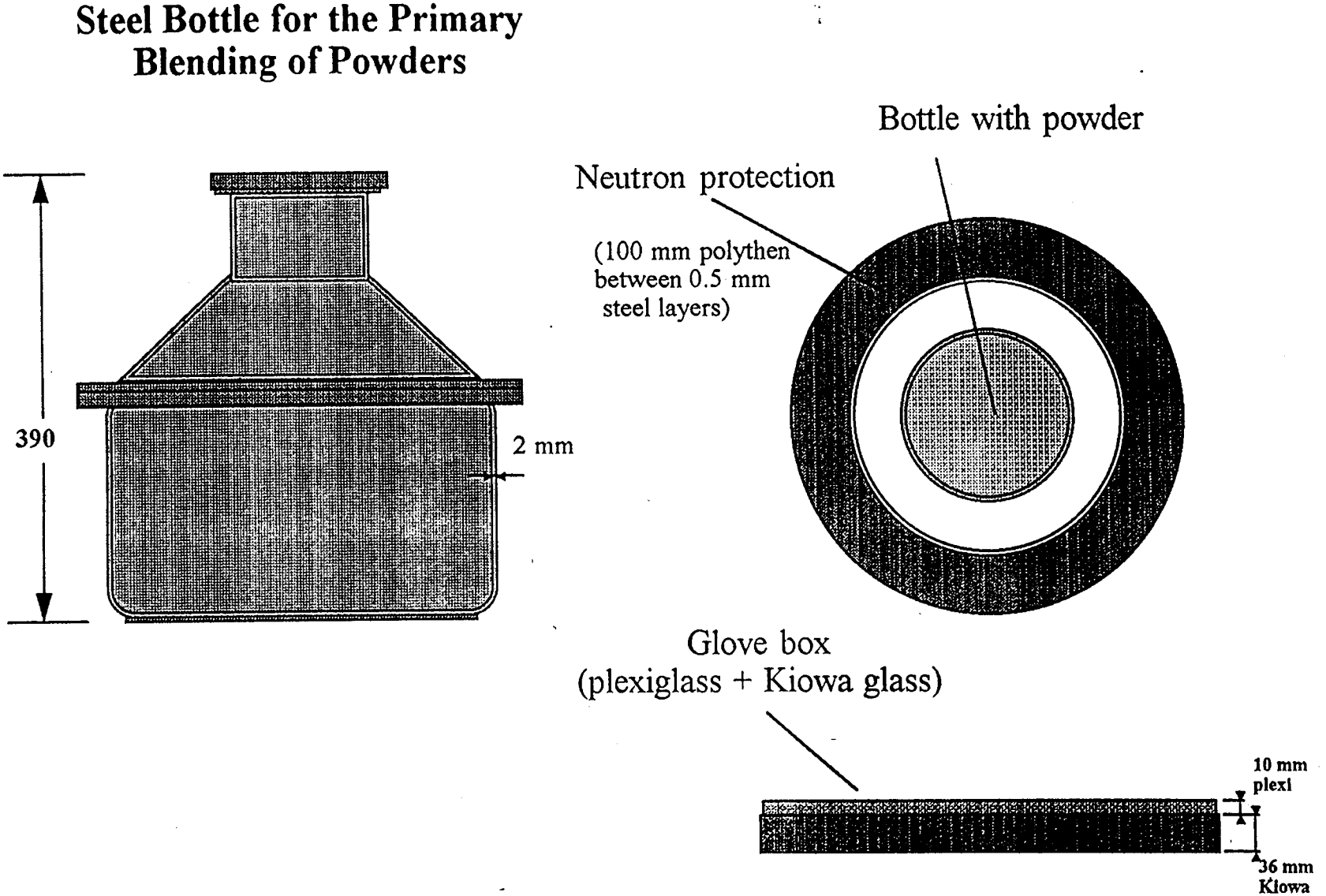

Fig. 1. Geometrical model for dose rate calculations with primary blending bottle

CONCEPT AND EXPERIMENTAL STUDIES ON FUEL AND TARGET FOR MINOR ACTINIDES AND FISSION PRODUCTS TRANSMUTATION

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ABSTRACT

High activity long-lived radionuclides in nuclear wastes, namely minor actinides (Americium and Neptunium) are in large amount generated by current nuclear reactive.

The destruction of these radionuclides is a part of the French SPIN (Partitioning and Burning) programme consistent with the determination to send a minimum amount of harmful products for final storage.

Transmutation concepts are defined for Neptunium and Americium tacking into account fuel cycle strategies.

Neptunium destruction does not pose any major problems. It's a by product of uranium consumption, as plutonium and in despite of a slight gamma activity due to the protactinium 233 it's quite easy to handle.

Diluting Neptunium in the MOX should not be an obstacle for fabrication, in-pile behaviour and reprocessing either. Consequently we make the proposal of homogeneous mode of Neptunium in MOX PWR's fuel which should be soon explored in the experimental OSIRIS reactor and in FR's Phenix and Superphenix.

The analysis is more complex for multi isotope Americium. Its destruction is difficult because of gamma radioactivity which complicates fabrication.

Experiments in Phenix and calculation showed that FR offer a good potential for Americium incineration, but similar datas do not exist for PWR.

It will remain a well know difficulty for fabrication and reprocessing. In this case we have to put a real new face to the fabrication flow-sheet of Americium compounds and we propose to develop the heterogeneous mode.

Targets choice are defined in term of :
- safety, considering fuel reaction with cladding and water sodium,
- transmutation rate, limited by target behaviour, in FR's (Phenix), PWR's (OSIRIS) and HFR (Petten),
- reprocessing, checking the solubility of such targets by Purex process.

So, at the beginning of our programme the account has been on improving fuel and targets properties related to safety and fuel cycle.

In order respond to the public concern about wastes and in particular the long-lived high level ones, a French law issued on December 30, 1991 identified the major objectives of research for the next fifteen years, before a new debate and possibly a decision on final wastes disposal in Parliament (2006).

To comply with the requirements of the law, CEA has launched an important and long term R and D programme. A part of this programme called SPIN [1] is devoted to partitioning and transmutation of these wastes. Once separated, elements have to be mixed with fuel or manufactured into target elements for transmutation.

Present transmutation studies of Minor Actinides (M.A) and long-lived fission products (LLFP) are conducted on a wide range of options :
- fission reactors (FR and PWR),
- M.A. homogeneous or heterogeneous recycling,
- LLFP transmutation in fission reactors.

Fuel studies cover the following areas :
- Fuel fabrication and irradiation for homogeneous recycling Np and Am both in thermal and fast reactors.
- Inert matrices basic research, with fabrication and irradiation for targets in the heterogeneous recycling mode, in particular for Americium.

HOMOGENEOUS RECYCLING

The Superfact experiment has shown on the one hand the good metallurgic performance of M.A fuels under irradiation in the Phenix F.R and on the other hand that the transmutation affected the whole of the fuel without "rim effect" leading to the disappearance of Np and Am actinides of between 25 and 30 % for those burn-up limited to 6 atoms % [2].

Neptunium destruction does not pose any major problems : Neptunium is a by product of uranium consumption, as plutonium and in despite of a slight gamma activity due to the protactinium -233 it's quite easy to handle.

Diluting Neptunium in the MOX fuel should not be an obstacle for fabrication, in-pile behaviour and reprocessing [3] consequently we make the proposal of homogeneous mode with priority to Neptunium in MOX fuel, which will be soon explored.

Figures 1 and 2 show respectively :
- the irradiation capsule for a new SUPERFACT-2 experiment in Phenix FR aiming to reach higher burn-ups (> 10 at %) than the previous SUPERFACT-1 experiment,
- the ACTINEAU experiment related to transmutation studies in PWRs.

Demonstration NACRE experiments are foreseen in SUPERPHENIX which will involve 1 and later 4 full subassemblies, with fuel pins containing ~ 2 % Np homogeneously mixed to the standard MOX FR's fuel. Aim is to reach with M.A homogeneous fuel the same burn-up (at % or equivalent fuel power days) than for the standard MOX fuels.

HETEROGENEOUS RECYCLING

Recycling is more complex for multi-isotope Americium [3]. Its destruction is difficult because of gamma radioactivity which complicates fabrication. It will remain a well known difficulty for reprocessing. In this case we have to put a real new face to the fabrication flow-sheet of Americium compounds and we propose to develop the heterogeneous recycling.

In the SUPERFACT-1 experiment we have used a content of 45 % weight of M.A which represents the heterogeneous recycling.

But in M.A-UO_2 fuels the disappearance of M.A is counter-balanced by the appearance of Pu isotopes throughout the fuel pellets.

Working from this base, we have expanded research on the transmutation of M.A using inert matrices as support in place of UO_2 in order to reduce the production of plutonium.

Targets choice are defined in term of :
- safety, considering fuel reaction with claddings, water (PWR) and sodium (FR),
- transmutation rate, limited by in-pile target behaviour studied in Phenix-FR, HFR Petten and OSIRIS-PWR [4],
- reprocessing, checking the solubility of such targets by Purex process.

Figure 3 shows the irradiation device for inert matrices test : MATINA experiment in Phenix FR launched in 1994.

LLFP TRANSMUTATION

An experiment of Tc-99 irradiation called ANTICORP 1 will start in a subassembly of PHENIX (core periphery) using a moderator to increase the epithermal captures. Figure 4 shows the irradiation device for the ANTICORP experiment.

The irradiation of 3 Tc pins, identical to the samples of the HFR-Petten irradiation [4], will be undergone in 1995 [5]. The fabrication by ITU of the bars should be completed by the beginning of 1995 after a grain structure study of as-casting Tc. For a long irradiation a refined grain structure is researched by increasing the cooling rate during the metal solidification.

On other hand a Tc alloy metallurgy study is launched to stabilize a cubic crystallographic phase by light addition of a cubic metal (Nb, Ti, Mo, V, Cr) in place of the hexagonal Tc system.

In the both case the aim is to avoid an anisotropic swelling during irradiation, well knowed phenomena occurring for coarse grain structure and anisotropic system either.

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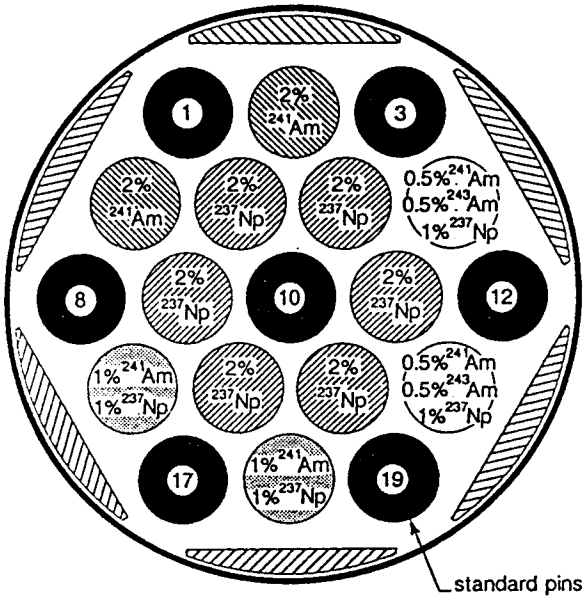

Fig. 1 Neptunium and Americium Homogeneous Concept. SUPERFACT 2 Experiment in PHENIX

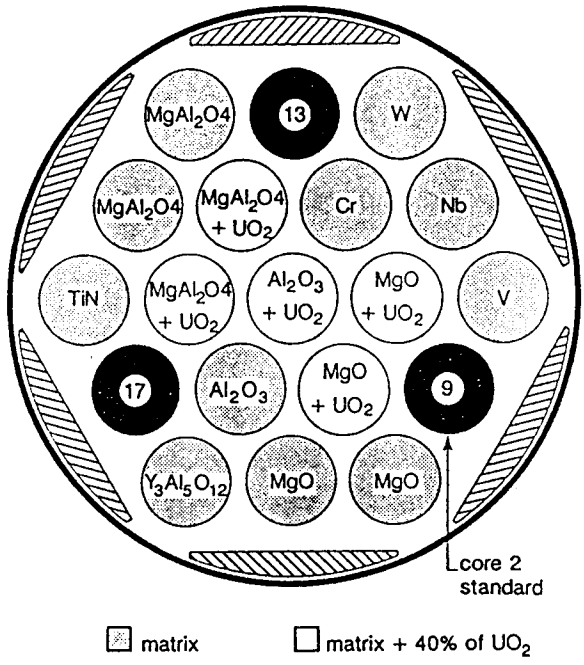

Fig. 3 Heterogeneous Concept. Behaviour of the inert matrix. MATINA Experiment in PHENIX

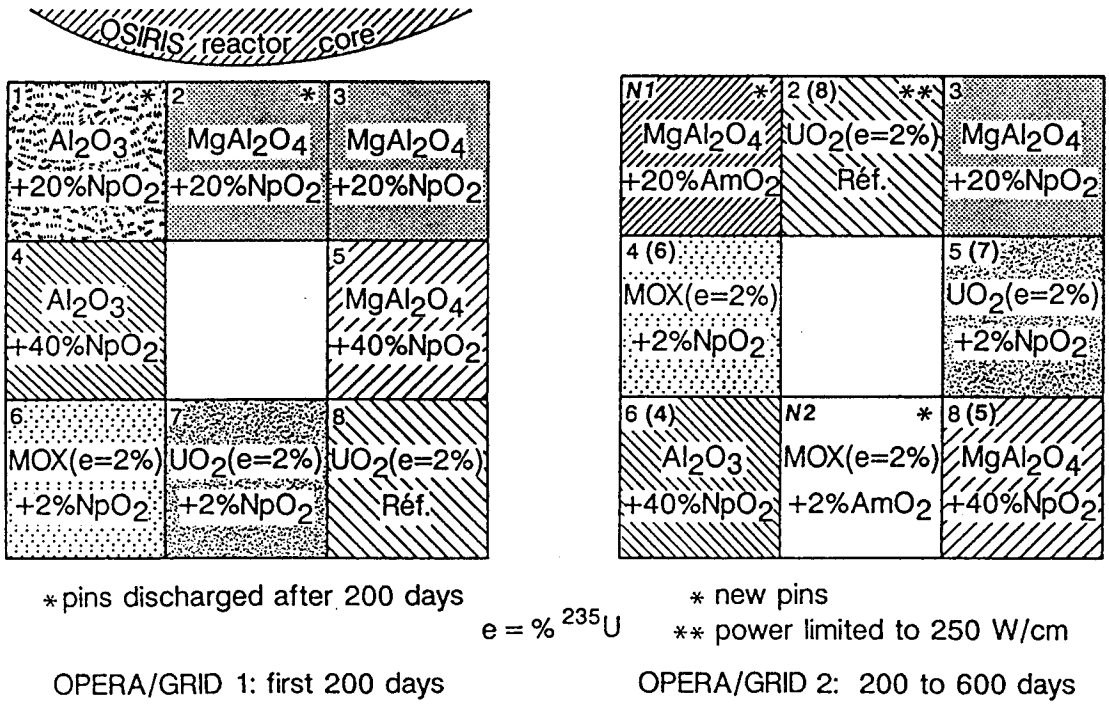

Fig. 2 Neptunium and Americium Heterogeneous and Homogeneous Concepts : ACTINEAU Experiment in the OPERA loop of OSIRIS Reactor. Pins positions in grid 1 (Np) and grid 2 (Am)

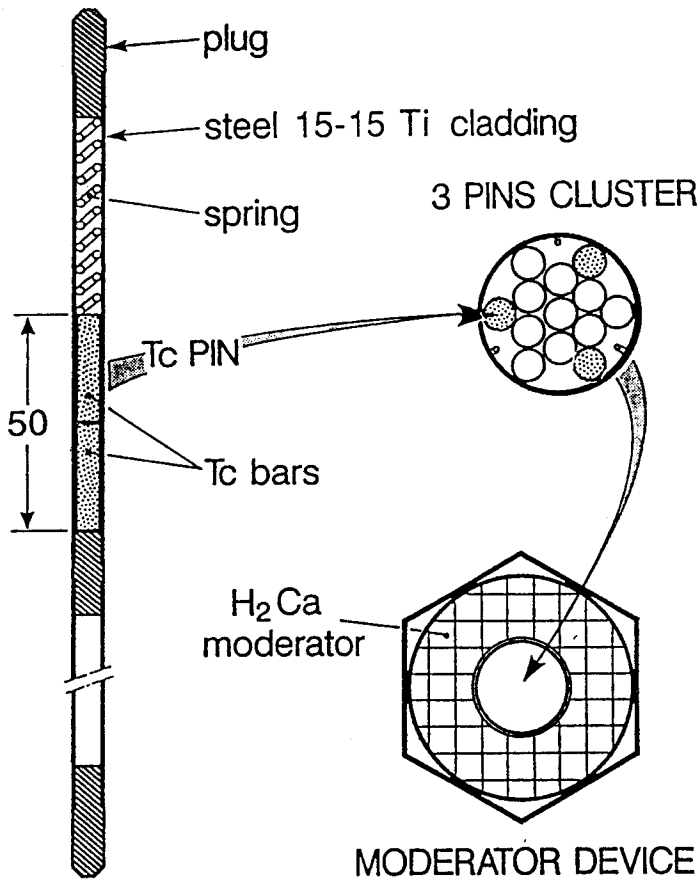

Fig. 4 ANTICORP 1 Experiment in PHENIX

Potential of Fast Reactors for Transmutation of Actinides and Long-Lived Fission Products

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The paper will discuss some results of the work which has been performed within the frame of a contract between the Commission of the European Communities and the Siemens AG on "Transmutation of long-lived radionuclides by advanced converters".

The main items of the paper are:

- Impact of Minor Actinides (MA) recycling on the properties of MOX fueled fast reactor cores with different size.
- Comparison of homogeneous and heterogeneous recycling of MA in fast reactors.
- Comparison of oxide and metal fueled fast reactors with respect to their MA transmutation capabilities.
- Potential of fast reactors for long-lived fission product transmutation.

1 Introduction

The recycling of Minor Actinides (MA) and long-lived fission products in fast reactors with the final goal to reduce their contribution to the long term radiotoxicity of the waste has been studied within a contract between the Siemens AG and the Commission of the European Communities.

Since it is well known that introducing MA into the fuel of a fast reactor results in an increase of the positive sodium void effect (SVE) and a reduction of the Doppler constant, design measures were investigated to limit the deterioration of these important safety parameters to an acceptable amount. These design measures were core size reduction and heterogeneous instead of homogeneous recycling. Additionally, oxide and metal fuel cores were compared with respect to the influence of MA recycling on their core properties. Finally, the transmutation of the long-lived fission products Tc-99, I-129 and Cs-135 has been studied in positions at the core outer boundary and in in-core positions.

2 Boundary conditions and methods of calculations

The main boundary conditions for the definition of the different reactor cores under investigation were taken from the European Fast Reactor EFR. These were mainly

- the maximum fuel burnup of 20 % which is achieved after 6 years residence time with a load-factor of 80 % and 6-batch loading scheme,

- limits for the maximum linear rating of 500 W/cm at beginning of life (BOL) and 400 W/cm at end of life (EOL),

- a plutonium composition (in weight %) of

  Pu-238 : Pu-239 : Pu-240 : Pu-241 : Pu-242 : Am-241 = 2 : 54 : 26 : 10 : 8 : 2

  corresponding to a light water reactor (LWR) discharge burnup of 40 MWd/kg HM,

- a MA composition of Np-237 : Am-241 : Am-243 : Cm-244 = 49 : 37 : 11 : 3

consistent to the same LWR burnup with Uranium fuel.

The main methods of calculations are indicated in View Graph (VG) 1.

3 Investigation of fast reactor cores of different size with homogeneous and heterogeneous MA recycling

Starting from the European Fast Reactor EFR as a basis, three reduced core sizes were considered which were realized by, firstly, reducing the core height from 1.0 m to 0.7 m and 0.5 m with constant core diameter and, secondly, by reducing the core diameter from 4 m to 1.8 m for a constant core height of 0.7 m. The main design data of these cores are listed in the upper part of VG 2 and in VG 3.

The heterogeneous recycling was simulated by implementing 16 target subassemblies (S/A) into the large core no. 1 in VG 3 and 6 or 12 target S/A into the small core no.4. The smear density of the MA-oxide was varied from 100 % down to 66 % and 33 % in order to adapt the total MA content in the core.

In VG 4 the results for the mass balances of the small core and the large one with 0.5 % MA (just the Am-241 linked with the plutonium), 5.4 % homogeneous and 5.4 % heterogeneous MA content are gathered. The following trends can be stated:

- The MA transmutation efficiency in terms of % per cycle and of kg per TWh is very similar in both recycling strategies if for the heterogeneous version the MA production in the standard fuel S/A is also taken into account. This is also illustrated in VG 5 where all calculated values in kg per TWh are given as a function of the MA content. All points fit to a linear dependence without separate influence of core size or recycling strategy. For 5 % MA content a transmutation efficiency of 12 kg per TWh can be deduced which corresponds to the production of about 5.5 LWRs.

- For smaller MA contents the transmutation efficiency is reduced more than proportionally because of the MA production of the standard fuel of about 3 kg per TWh. For 2.5 % MA content only 4 kg per TWh or the production of 1.8 LWRs are transmuted.

- The efficiency in relative terms is larger in the larger cores because of their higher flux level. Again, the values are small for small MA contents and they reach 8 % per year in the large core versus 6 % in the small one. During the S/A life of 6 years total reductions of 36 % in the large cores and 30 % in the small cores can be reached. In the heterogeneous MA S/A the reductions are certainly larger: for both core types values of 10 % per year and 50 % per S/A lifetime can be stated. In any case multiple recycling of MA is necessary for a considerable reduction of the MA.

Some remarks have to be made concerning the build-up of Pu-238 in the MA containing fuel. Even in very hard neutron spectra the production by capture processes in Np-237 and Am-241 with their subsequent decays cannot be completely avoided. Therefore the amount of Pu-238 produced in standard fast reactor spectra can be significant. As an illustration VG 6 shows the Pu-238 fraction in the total Pu as a function of MA content at BOL for different Pu enrichments. The Pu-238 fraction goes up to 15 - 20 % for 15 % MA content with the lower value occuring in case of high Pu-enrichments, which is caused by the harder neutron spectra in the higher enriched cores. In the MA containing target S/As the Pu-238 content even reaches values of 80 %.

Presently a limit of 5 % is set for the Pu-238 fraction in the total Pu. This is caused by the fact that Pu-238 is emitting neutrons by spontaneous fission and α-particles, so that efficient measures for shielding and cooling (Pu-238 produces 560 W/kg) during refabrication and transport are necessary.

If it turns out that recycling strategies with larger EOL contents of Pu-238 are beneficial, a more accurate evaluation of the limiting value would be required. This would not only have to include the Pu-238 content itself, but generally the special isotopic composition of the fuel under consideration, the mass throughput of the plant, special aspects like the influence of subritical multiplication and possibilities to enhance the shielding and cooling efficiency.

Taking the 5 % limit as given for the time being, a BOL MA-content of 3 % should not be exceeded.

In the lower part of VG 4 the results for the sodium void effect and the Doppler constant are given. The fissile sodium void effect (SVE) is strongly increased in the

homogeneous cases (30 % in the small core and 20 % in the large core for 5.4 % MA content) and the Doppler constant is reduced by 30 % in both cores. These disadvantages can be avoided to a large extent in case of heterogeneous recycling of MA: the fissile void effect is almost unchanged or slightly reduced compared to the reference case, the total core SVE (standard plus MA S/A) is increasing to a smaller amount (about 10 %) and the Doppler constant is less decreasing. This is an interesting advantage of the heterogeneous strategy.

4 Oxide and metal fueled cores with MA transmutation

For this comparison the EFR-like core no. 2 in chap 3 and VG 3 with 0.7 m core height and a thermal power of 2600 MW was taken. The main core and S/A data were identical for both fuel types with the exception of the fuel data itself with the related smear density (9.2 g/cm^3 for oxide, 11.9 g/cm^3 for metal) and the Pu-contents to adapt the reactivity to the right level (20.0/25.0 % for oxide, 16.4/19.4 % for metal for the inner /outer core zone). The cycle length has been increased from 292 to 352 efpd to keep the maximum burnup about constant.

The efficiency of the MA transmutation has been calculated for a MA content of 5 % in the total heavy metal of both fuel types and for both cases a reference case without additional MA was established. The results concerning the MA transmutation efficiency are gathered in the upper part of VG 7. They do not indicate a clear advantage for either fuel type:

- Due to the larger MA mass at BOL resulting from the higher metal fuel density, the transmuted mass per year is about 24 % higher for metal fuel than for oxide fuel.

- The relative changes per year are however slightly larger in the oxide fuel since differences with respect to microscopic cross-sections and flux level are largely compensating each other. Consequently the transmutation half-time is a little bit smaller in oxide cores.

Some important safety and operational parameters are gathered in the lower part of VG 7. Here the following aspects are important:

- Compared to the oxide fuel the metal fuel shows some specific advantages, which are based on the higher fuel density, i. e. lower Pu-enrichments, lower loss of reactivity per year due to fuel burnup and a longer fuel residence time. Assuming the same number of cycles per residence time for both cores, the absorber system requirements are reduced by about 45 % so that the control rod worth reduction by 13 % is more than compensated.

- The sodium void effect (SVE) deteriorates for metal fuel compared to oxide fuel by about 50 - 60 %. The Doppler constant is reduced by about 50 %.

A valuation of the changes of these important safety parameters is not easy because of their complex influence on the core safety behaviour.

For the sodium void effect the situation is quite clear since its increase is a disadvantage in all types of accidents. The role of the Doppler coefficient, however, depends upon the detailed accident conditions. In cases of unprotected loss of heat sink or loss of flow the smaller Doppler coefficient is an advantage since it reduces the reactivity to be provided by other negative reactivity effects (i. e. fuel expansion, core expansion, control rod drive line expansion) for reactor shutdown. In transient overpower conditions and transients with sodium voiding, however, smaller Doppler coefficients cause more severe accident conditions. Therefore, no clear tendency can be given for the influence of the MA on the core safety behaviour without detailed safety studies.

5 Transmutation of fission products in fast reactors

The fission products Tc-99, I-129 and Cs-135 with half-lifes of $2.1 \cdot 10^5$ years, $1.6 \cdot 10^7$ years and $2.3 \cdot 10^6$ years, respectively, belong to the long-lived nuclides in the high-level waste. Their risk contribution during storage is relatively high because of their pronounced mobility in geochemical environments around the waste repositories. Their transmutation by neutron capture into stable isotopes (Tc-99 → Ru-100, I-129 → Xe-130, Cs-135 → Ba-136) is therefore of interest.

The one-group capture cross-sections of these isotopes are rather small in a standard fast reactor neutron spectrum. They are below 0.5 barn so that in an average neutron flux of $3 \cdot 10^{15}$ cm^{-2} s^{-1} less than 25 % of the fission products are burnt during a fuel subassembly life of 6 years.

One possibility to increase the capture cross-section of the fission products is to place them in a moderated environment in order to soften the neutron spectrum. This effect was studied by introducing $ZrH_{1.7}$ in the target subassemblies containing Tc-99, I-129 and Cs-135 and by varying the $ZrH_{1.7}$ content. VG 8 shows the one-group cross-sections of these isotopes as a function of the $ZrH_{1.7}$ content. The strong influence of the moderator content can be seen, but the fission product content itself has a similar impact. The larger fission product content leads to a spectrum hardening in the target subassemblies so that the capture cross-section is decreasing.

VG 8 is also showing that the cross-section of Cs-135 is small even in case of a large moderator content. Therefore, there is no chance for a transmutation of this isotope in a fast reactor.

The following results are restricted to Tc-99 as the most frequent long-lived fission product. Its transmutation in a $ZrH_{1.7}$ moderated surrounding was studied first in periphal positions and then in in-core positions.

Basis for this study was the EFR core (core 1 in chap. 3, VG 3), in which the radial blanket row was completely replaced by 78 target subassemblies (S/A). For the design of these S/A it was assumed that 20 % of the volume consist of structural material (steel), 40 % are filled with sodium and that the remaining 40 % are left for fission products plus moderator. The fission product content was varied between 2 and 20 % and the moderator content between 5 and 20 %.

For a judgement about the transmutation efficiency the transmutation half-time, which should be as small as possible, and the transmutation rate in kg/a, which should be as large as possible, are two indicators. They are plotted in VG 9 as a function of the Tc-99 content with the $ZrH_{1.7}$ content as additional parameter. It is obvious that the Tc-99 content has a much stronger influence on both indicators than the $ZrH_{1.7}$ content.

If one considers the fact that during burnup of the reactor about 20 kg of Tc-99 are produced per year and that the corresponding value in a LWR is about 30 kg/a, it becomes evident that the transmutation rate should be beyond a value of about 70 kg/a so that the own production and that of two LWRs could be

transmuted. This could be possible with about 20 % Tc-99 content and 10 - 20 % $ZrH_{1.7}$ content, but the corresponding transmutation half-time would be 40 - 50 years. This figure, however, is much too large since it would require the irradiation during some hundreds of years to reach a quantitative reduction of the Tc-99 waste. Consequently the use of in-core positions was investigated in the second step of this study.

The basis for this study was core 1 in chap. 3 and VG 3. In this core up to 84 fuel S/A have been replaced by Tc-99 containing S/As. Their position has been varied in order to optimize the radial power distribution.The final configuration with 84 Tc S/As is shown in VG 10. Since the number of fuel S/As is reduced by 84 out of originally 376, the total thermal power was reduced from 3600 MW to 2600 MW. In order to get some margins with respect to the maximum linear rating limits, the core height was increased in some cases from 1 m to 1.2 m. The $ZrH_{1.7}$ content was varied from 0 % to 10 % with a Tc-99 content of 10 % and 20 %.

The main results of these calculations were the following:

- The optimum $ZrH_{1.7}$ content is close to 3 % since larger values cause a stronger flux depression in the target S/As so that the capture rate decreases although the one-group cross-section is further increasing. This increase is, however, less pronounced than in the peripheral positions because the surrounding fuel S/As are mainly determining the spectrum.

- The Tc-99 content has again controversial influences on the transmutation half-time and the transmuted mass per year since both parameters are increasing with increasing Tc-99 content. Typical values for 3 % $ZrH_{1.7}$ are:

	$T_{1/2}$ [a]	ΔM [kg/a]
10 % Tc-99:	19	110
20 % Tc-99:	25	170

In view of the facts that the fast reactor itself produces about 20 kg/a and that one LWR produces about 30 kg/a one can conclude that with 10 - 20 % Tc-99 content the production of 3 - 5 LWRs could be transmuted. The related transmutation half-times are in the range of 20 - 25 years.

The total amount of Tc-99 which would be loaded into the reactor with 84 target S/As and 20 % Tc-99 volume fraction is roughly 6000 kg for a 1100 MW_e reactor.

In order to get an idea about the influence of the moderated Tc-99 subassemblies on the safety parameters of the core, the sodium void effect, the Doppler coefficient and the control rod worth have been calculated for a 1.2 m high core with 84 Tc-99 S/As with 20 % Tc-99 and 3 % $ZrH_{1.7}$ content. An end-of- equilibrium-cycle condition was simulated by adding appropriate fission product concentrations to the fresh fuel. The table below gives the results in comparison to the corresponding values of the 0.7 m high core 2 in chap 3:

	core with Tc-99 (1.2 m high)	core 2 (0.7 m high)
sodium void effect [$]	3.9	4.4
Doppler constant [$]	- 1.2	- 1.6
control rod worth [$]	28	23

Despite the relatively large core height of 1.2 m, the SVE is 12 % smaller compared to the flat homogeneous layout. This is a consequence of the higher Pu enrichment, the moderating effect of the $ZrH_{1.7}$ and of the strong heterogeneity of the core with 84 Tc-99 target S/As. On the other hand the Doppler constant is reduced by 27 % due to the higher Pu-enrichment in the core S/As and the harder neutron spectrum. But this should be acceptable in view of the smaller SVE.

The control rod worth is more than 20 % larger compared to the flatter core 2, which, however, is necessary because a larger burnup reactivity swing has to be compensated.

As a conclusion, one can state that the transmutation of the long-lived fission products Tc-99 or I-129 in special in-core positions seams to be in principal feasible since it is acceptable from the core design and safety point of view. A core with 2600 thermal MW could be loaded with about 6000 kg of Tc-99 and I-129 and it would transmute about 170 kg/a which corresponds to the production of 5 LWRs with 1300 MW_e each plus the production of the fast reactor itself. With respect to these absolute values the result is quite encouraging, but in relative terms it is very disappointing which is expressed by the transmutation half-times of 20 - 25 years for Tc-99 and 30 - 40 years for I-129. For a quantitative mass reduction of

these isotopes one would need at least 5 half-times (giving a reduction to 3 % of the initial mass) and this would require an irradiation time of 100 - 200 years. Since the residence time of the target S/As will be not larger than 10 years the fission products have to be recycled more than 10 times. Each time the S/As have to be reprocessed and refabricated and the daughter product Ru-100 of Tc-99 has to be separated from the target material. Therefore, it is highly questionable whether such a recycling concept is technically feasible.

6 Conclusion

The investigations of the capabilities of fast reactors with respect to the transmutation of MA and long-lived fission products have lead to the following conclusions:

- The concepts of homogeneous and heterogeneous recycling of MA have similar transmutation efficiencies: 30 - 35 % of the initial MA content are transmuted during a S/A lifetime of 6 years. Therefore multiple recycling will be necessary for a quantitative reduction of the MA masses.

- Small cores are less efficient than larger cores because of their lower flux level. But they have the advantage to limit the positive sodium void effect, which is increasing with increasing MA content, to acceptable values.

- An upper limit of 5 % MA content is recommended for the homogeneous recycling which limits the deterioration of the sodium void effect and the Doppler effect to an acceptable amount.

- For 5 % MA content a transmutation efficiency of 12 kg/TWh can be deduced which corresponds to the production of about 5 Light Water Reactors.

- Special attention has to be paid to the build-up of Pu-238 in the fuel which reaches 7 % of the total Pu in case of 5 % MA, which requires efficient measures for shielding and cooling during refabrication.

- Heterogeneous recycling of MA is recommended for further studies, since it limits the number of MA containing S/As to about 5 % of all fuel S/As and

since it avoids the deterioration of the sodium void effect and the Doppler effect. Measures with respect to the extremely high Pu-238 content have, however, to be taken in this concept.

- The comparison of oxide and metal fueled fast reactors with respect to their transmutation efficiency has shown no clear advantage for one of these concepts.

- The transmutation of fission products has been studied for the two cases where the target subassemblies were located either in peripheral or in in-core positions. It was shown that when using peripheral positions, the masses of Tc-99 and I-129, which can be transmuted per year, are in the range of the production of 2 - 5 LWRs, but the transmuting half-times are rather poor: 20 - 25 years for Tc-99 and 30 - 40 years for I-129 in in-core position whereas they are by a factor of 2 larger in peripheral positions. For a quantitative reduction 5 half-times would be necessary which mean an irradiation time of 100 - 200 years. This indicates that these recycling concepts seem not to make sense.

- Cross sections of ENDF/B–IV and –V, ENDL were used; some isotopes were taken from the ORIGEN2 library "Advanced oxide, LWR–Pu/U".
- Eigenvalue, flux and power calculations were performed in 3d–geometry using the diffusion code DEGEN.
- 26 group spectra were used to produce one–group cross–sections for burnup calculations with ORIGEN2. Burnt concentrations were then used again in flux and power calculations in DEGEN.
- Absorbers were inserted to keep k_{eff} between BOC and EOC almost constant. The relative insertion depths of the inner and outer control rods were adjusted to have almost equal power peaks in both core zones.
- Plutonium contents were adapted so that k_{eff} was about 1.01 at EOC with rods out and that the maximum linear ratings were similar in both core zones.
- Sodium void effects and Doppler effects were deduced via direct k_{eff} calculations.

VG 1 : Methods of calculations

parameter		unit	core 1	core 2	core 3	core 4
thermal power		MW	3600	2600	2000	450
core height		m	1.0	0.7	0.5	0.7
core diameter		m	4.0	4.0	4.0	1.7
no. of fuel S/As		–	375	375	375	84
no. of control rods		–	24	24	24	6
no. of pins per S/A		–	331	331	331	271
fissile	MA=0	$	5.9	4.4	3.0	2.5
void	MA=5%	$	6.5	5.1	–	3.1
effect	MA=15%	$	7.5	6.3	–	4.8
Doppler	MA=0	$	–1.7	–1.6	–1.4	–1.1
constant	MA=5%	$	–1.3	–1.2	–	–0.8
	MA=15%	$	–0.7	–0.6	–	–0.5
$\Delta\varrho$ per	MA=0	$	–8.9	–10.4	–12.8	–12.3
full power	MA=5%	$	–4.6	–6.7	–	–8.9
year	MA=15%	$	+1.6	–1.5	–	–4.9
control	MA=0	$	25	23	19	31
rod	MA=5%	$	24	21	–	30
worth	MA=15%	$	20	17	–	27

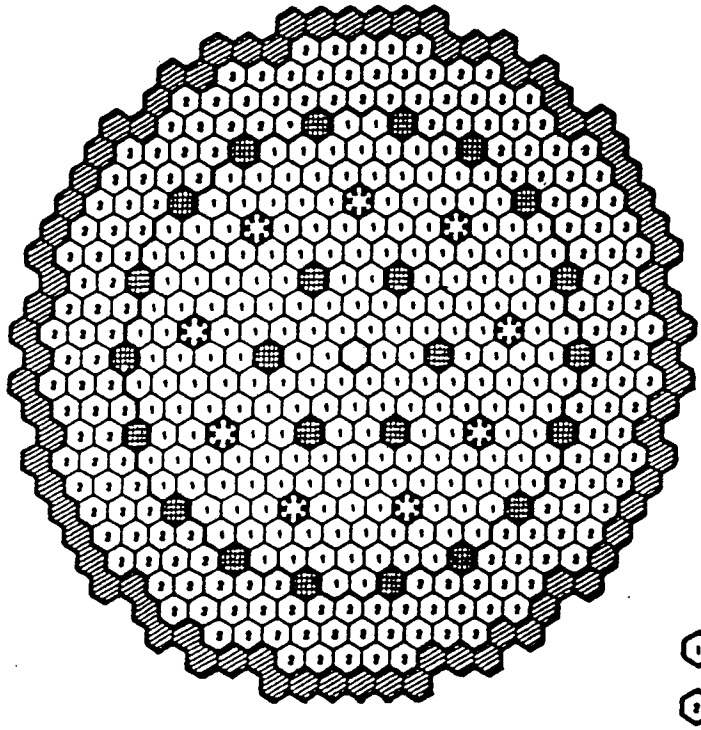
VG 2 : Design and EOEC performance parameters of cores with different size

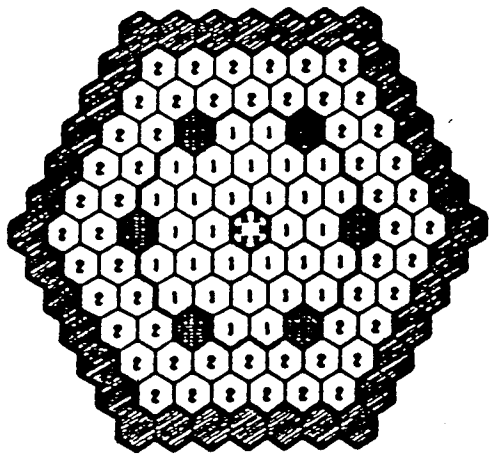
EFR – like :

Core 1 :
1 m height,
3600 MW_{th}

Core 2 :
0.7 m height,
2600 MW_{th}

Core 3 :
0.5 m height,
2000 MW_{th}





inner core S/As
outer core S/As
radial breeder S/As
control and shutdown rods
diverse shutdown rods
dummy S/A

Small Core :

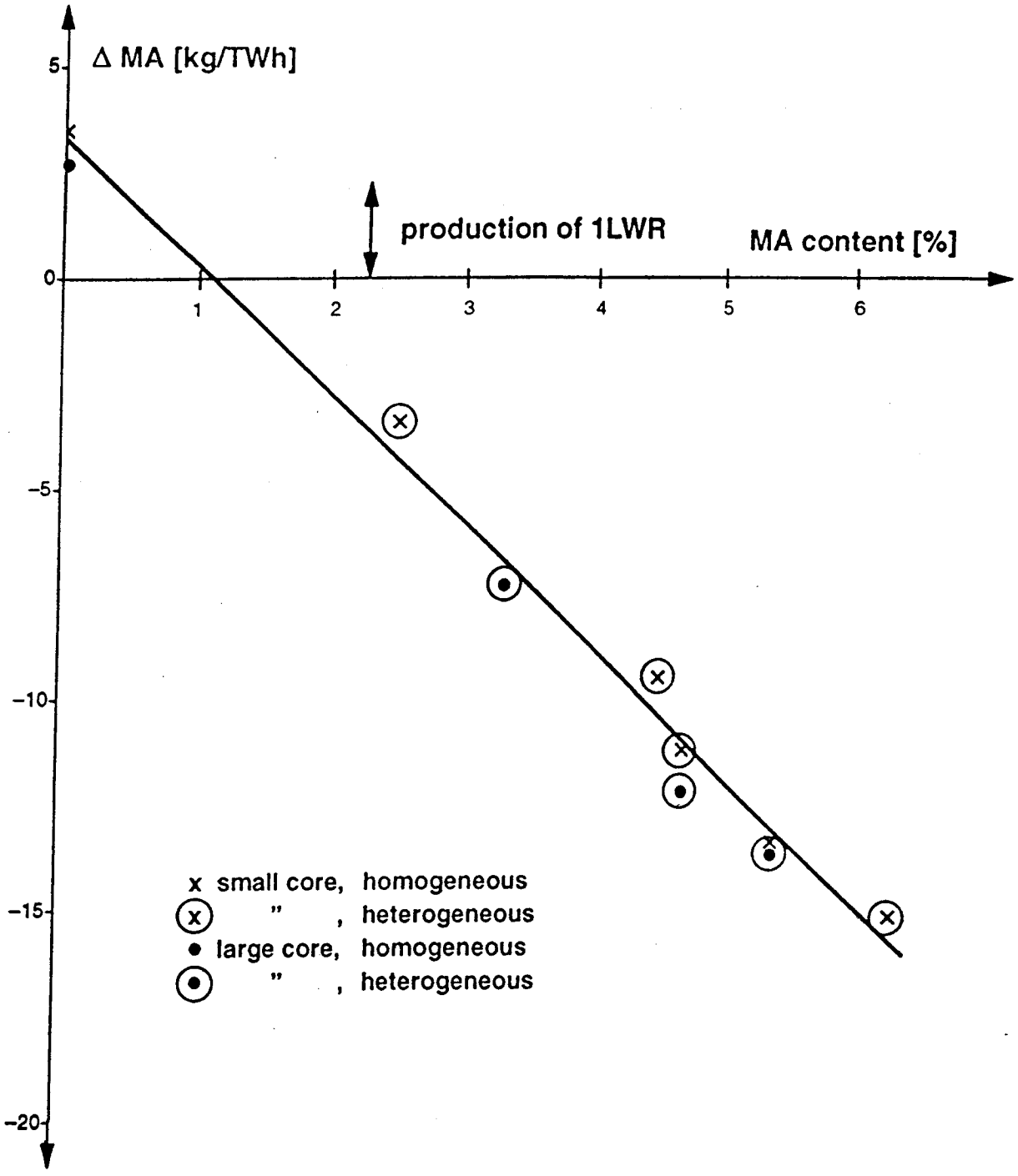
Core 4 :
0.7 m height,
450 MW_{th}

VG 3 : Core layouts with different core size

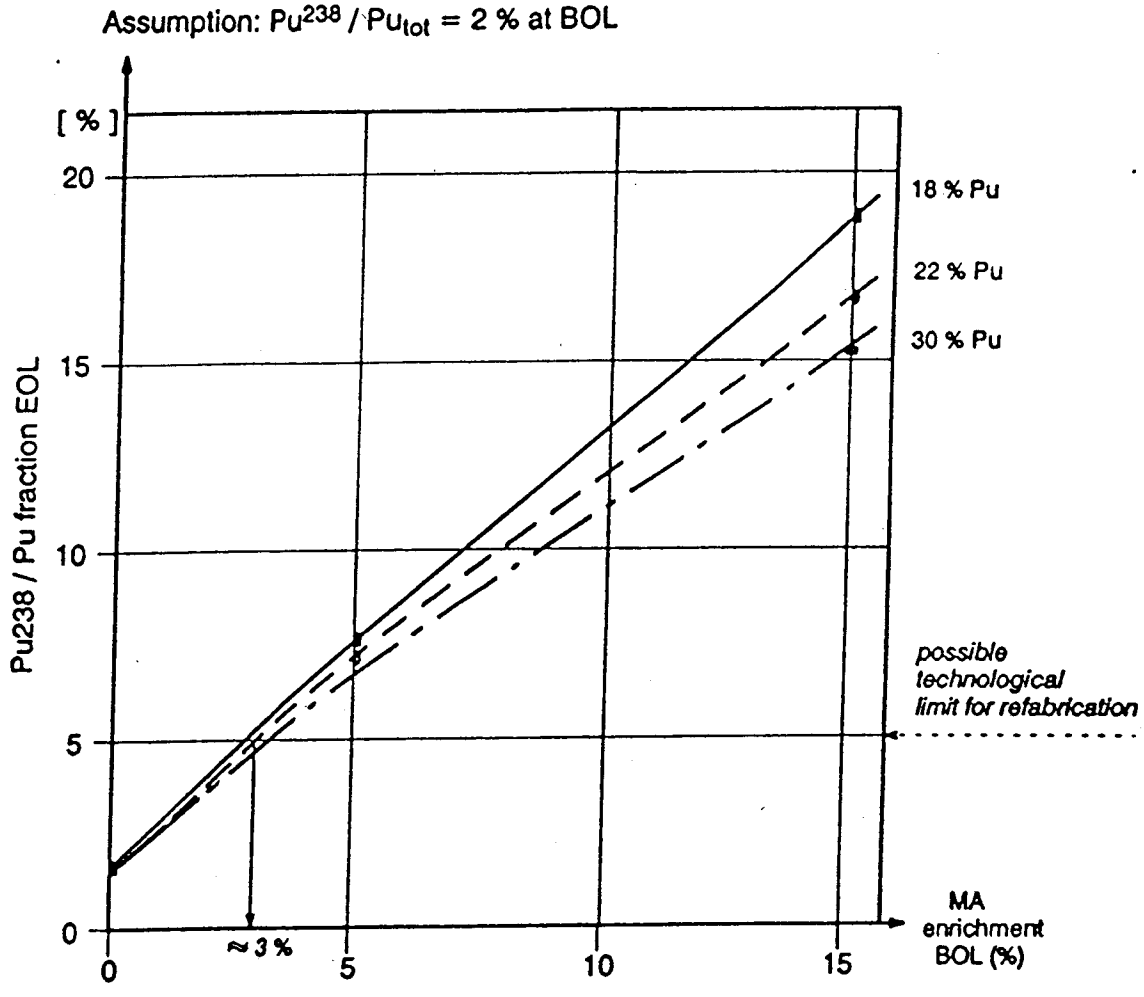
parameter		unit	600 MW core			2600 MW core		
			0.5% HOM	5.4% HOM	5.4% HET*	0.5% HOM	5.4% HOM	5.4% HET*
BOL inventory	Pu	kg	2044	2044	2030	6871	6871	6617
	MA	kg	41	448	460	138	1586	1590
ΔM per cycle	Pu	%	–2.7	–2.2	–2.3	–2.9	–2.2	–2.1
	MA	%	+9.8	–6.2	–5.6	+9.8	–8.0	–8.0
ΔM / TWh	Pu	kg	–31.2	–25.0	–26.8	–26.0	–20.0	–16.0
	MA	kg	+3.5	–13.4	–12.5	+2.7	–13.7	–12.6
$\Delta\varrho_{VOID}$ (total)		pcm	1150	1530	1300	1370	1670	1550
Doppler constant		pcm	–560	–380	–475	–600	–410	–450

* inter– or extrapolated from calculated values

VG 4 : Comparison of core properties of 600 MW_{th} and 2600 MW_{th} Minor Actinides transmuting fast reactors



VG 5 : MA transmutation capacity as a function of MA content



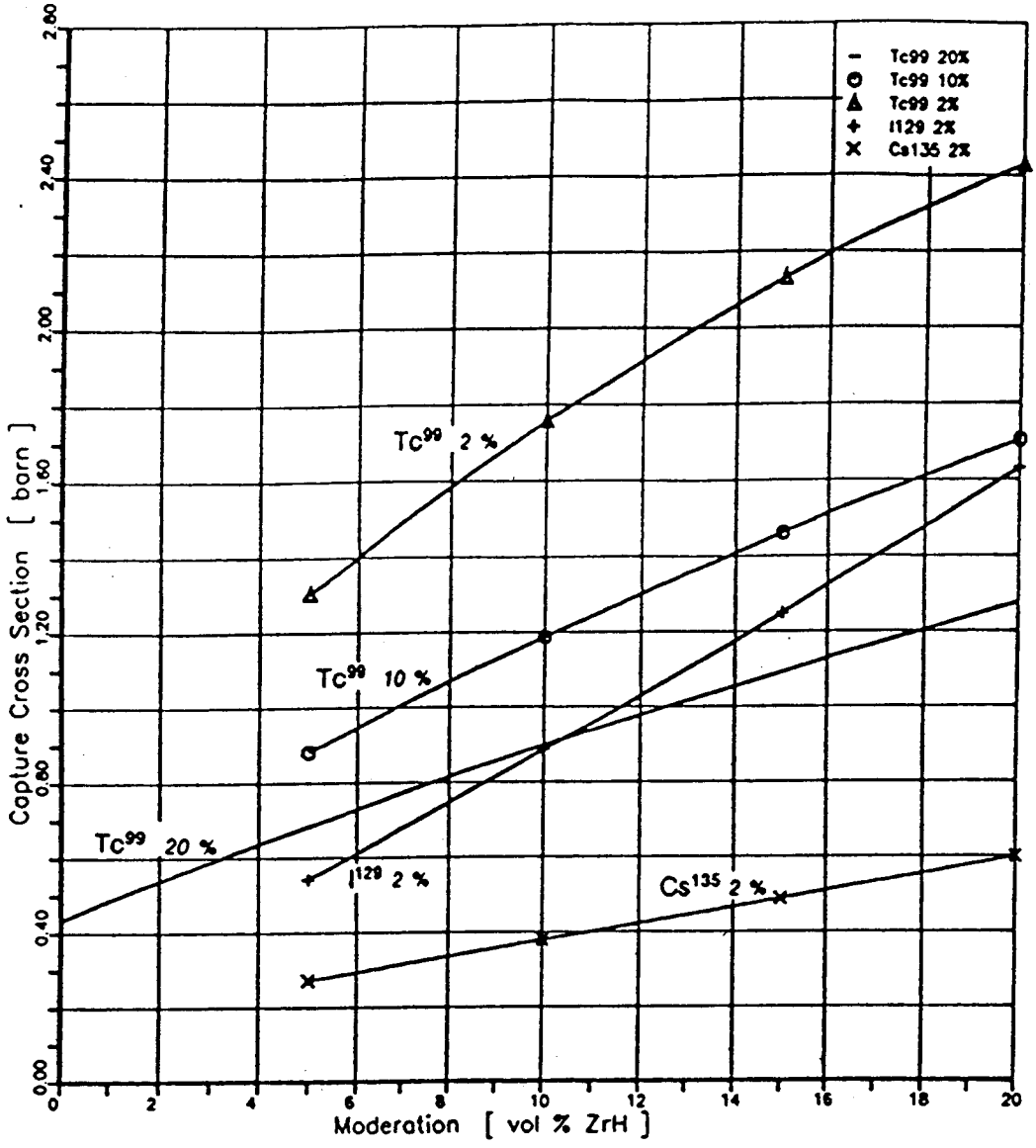
VG 6 : Pu238 generation in MA containing cores

parameter	unit	oxide	metal
MA inventory BOL	kg	1521	2004
MA inventory EOL	kg	934	1121
changes per year	kg	–98	–122
changes per TW_e h	kg	–12.8	–15.9
changes per year	%	–6.4	–6.1
transmutation half–life	a	10.5	11.0
Pu–238/Pu–tot EOL	–	0.078	0.076

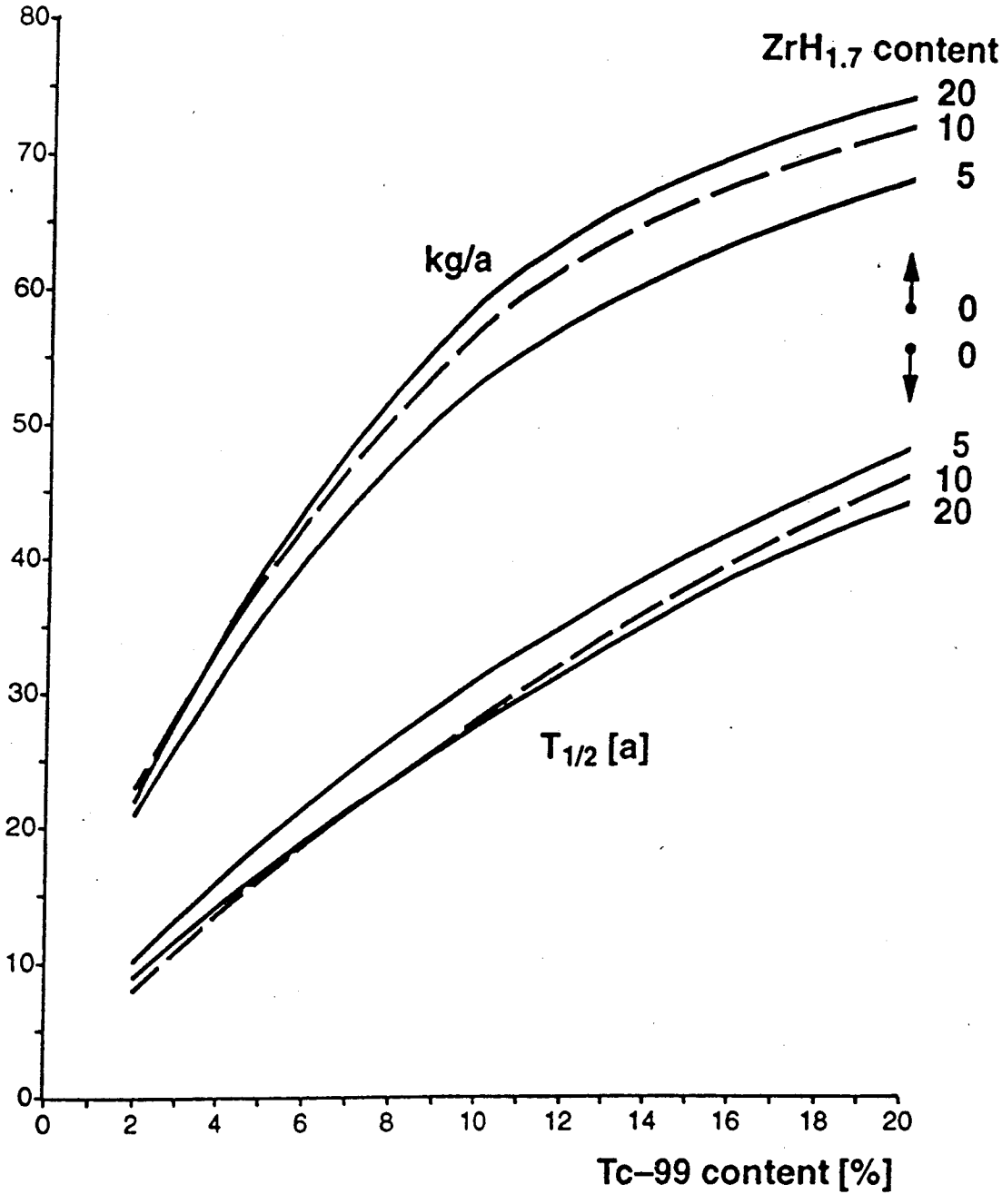
Mass balances in metal and oxide cores of 2600 MW_{th} with 5% MA at BOL

parameter	unit	oxide		metal	
		0% MA	5% MA	0% MA	5% MA
fissile void effect, EOC	$	4.4	5.1	7.4	7.9
Doppler constant, EOC	$	–1.6	–1.2	–0.8	–0.6
reactivity loss per cycle	$	–8.3	–5.4	–4.6	–2.7
control rod worth	$	23	21	20	18

VG 7 : Performance parameters of cores with metal and oxide fuel for 0 and 5% Minor Actinides

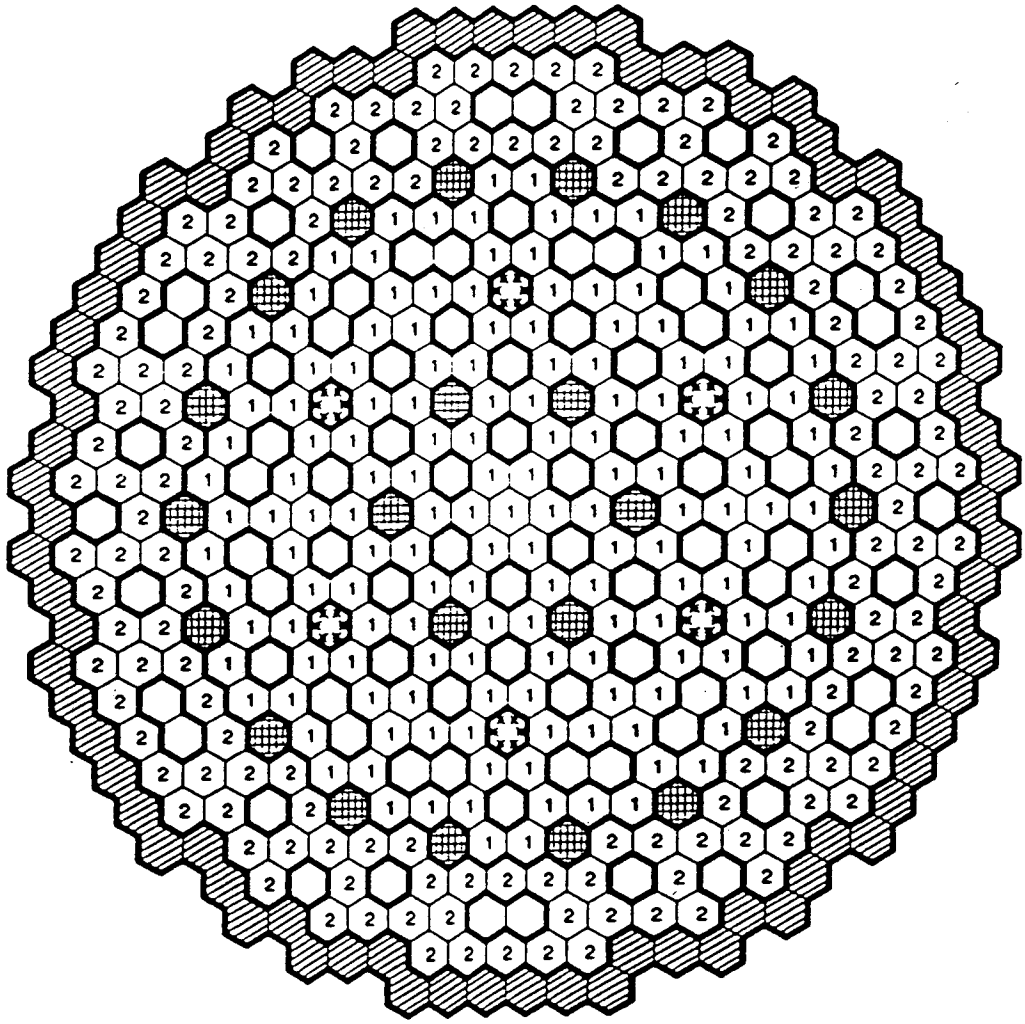


VG 8 : One–group capture cross–sections for Tc–99, I–129 and Cs–135 in dependence on moderator content



Half–life and rate of Tc–99 transmutation in peripheral positions

VG 9



Explanations

- inner core S/As
- outer core S/As
- reflector S/As
- control and shutdown rods
- diverse shutdown rods
- Tc–99 S/As

VG 10 : Cross–section of the core with 84 Tc–99 containing subassemblies

STUDIES ON NITRIDE FUEL CYCLE FOR TRU BURNING

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The research on a nitride fuel cycle for burning transuranium elements(TRUs) in actinide burning reactors(ABR) has been carried out in JAERI. With regard to fuel fabrication, the techniques of direct conversion of actinide nitrate solution from the reprocessing to nitride have been studied by applying the sol-gel method. It was confirmed that pure neptunium mononitride can be prepared by carbothermic reduction of the dioxide. The CVD process is also being developed for coating low density TiN to nitride fuel particles. The properties necessary for fuel design were investigated by using the samples synthesized by carbothermic reduction. The thermal conductivity of NpN was estimated from the thermal diffusivity determined by a laser-flash method. The partial pressures of Np and Am over the nitrides were measured by Knudsen-effusion mass spectrometry. The status of the research on the pyro-chemical process with fused salt electrorefining is also described.

1. Introduction

Nitride fuel will enable the adoption of the cold fuel concept, because of its good thermal properties, which result in lower fuel temperature and hence lower fission gas release. Furthermore, the cold fuel may accept a thinner cladding and make the neutron spectrum harder. The mutual miscibility of actinide mononitrides also leads to the fuel design of high concentration of transuranium elements(TRUs). Actinide burner reactor(ABR) concepts with Na-bonded pelletized fuel and TiN-coated particle fuel have been proposed from JAERI[1], based on the excellent thermal characteristics of actinide nitrides. For reprocessing, an innovative fuel cycle combined with fused salt electrorefining will be available in the case of nitride fuel as well as metallic fuel[2].

Fabrication method of TRU nitride fuel and their properties have been studied in JAERI following the concept of the transmutation of TRUs at ABR. In the present paper, the status of the research on nitride fuel for TRU burning is described.

2. Fabrication of fuels for TRU burning

2.1 Sol-gel method

Synthesis of the nitrides can be made by carbothermic reduction of the oxides in a nitrogen atmosphere. Minor actinides (Np, Am and Cm) will be separated from the high-level waste as nitrates. The actinide nitrates will then be converted to the mixture of oxides and carbon via the sol-gel techniques, which is to be fed to the carbothermic synthesis of the nitrides.

The sol-gel process is studied with particular emphasis on the application of microwave techniques first introduced by PSI, Switzerland[3]. The process is basically the internal gelation process. The ammonia donor, HMTA(hexamethylenetetramine), is added to the nitrate solution. By heating the liquid droplets to ~80°C, HMTA is decomposed to form ammonia. The heating of the droplets by microwave is almost spontaneous: gelling of droplets is complete while they fall through a microwave cavity of ~20 mm length(fig. 1). In our apparatus, an industrial frequency of 2.45 GHz has been successfully employed for the gelling of uranium nitrate solution[4]. Further efforts are being made:

(1)Mixing of the feed solution right above the nozzle in order to prevent the premature gelling by alpha-decay heat.

(2)Designing microwave cavity to minimize the droplet falling distance.

2.2 Preparation of neptunium nitride by carbothermic reduction

Actinide nitrides such as UN and PuN can be synthesized by carbothermic reduction of the dioxides with graphite in a N_2-H_2 mixed gas stream[5,6]. The same techniques were applied to the preparation of the mononitride of Np-237 from the dioxide[7]. The mixture of NpO_2 and graphite with a C/Np ratio of 2.2 was compacted into thin discs of 12 mm in diameter and about 2 mm in height. The compacts were first heated at 1,823 K in a N_2 stream for 36 ks to convert oxide to nitride and then heated at 1,723 K in N_2-8%H_2 for removing residual carbon in nitride.

The product was confirmed by X-ray analysis to be a nearly single phase with NaCl-type structure having the lattice parameter of 0.48968±0.00006 nm, which is close to the value of the mononitride prepared by hydriding-nitriding of the metal, 0.48979±0.00003 nm[8] and 0.48987±0.00005 nm[9]. Chemical analysis of the product[10,11] revealed that the impurity contents of carbon and oxygen are low as shown in table 1. It is shown from the result that NpO_2 could be converted to pure NpN by a similar procedure to the cases of UN and PuN.

It is notified from the experience of nitride fuel fabrication that residual carbon contents in actinide mononitrides synthesized by carbothermic reduction have a tendency to decrease with increasing atomic number. It could be explained from thermodynamic estimation that the removability of carbon depends on the solubility of carbon in nitride phase. In the case of UN, the solubility of carbon in the nitride phase is calculated to be around 0.10 at 1,723 K. On the other hand, the solubility of carbon in PuN is below 0.01. Such differences in carbon solubility may affect the content of residual carbon in UN, NpN and PuN.

The mononitride synthesized from the oxide was ground and compacted into a green pellet, which was heated at 2,000 K for 72 ks in a N_2-8%H_2 atmosphere. The sintered density reached over 90 % of theoretical density(T.D.). Sample preparation was carried out in gloveboxes filled with purified argon gas[12]. The oxygen and water levels of the atmosphere in the gloveboxes were less than 3 and 5 ppm, respectively. The NpN sample prepared was supplied for the studies on the properties such as thermal conductivity and vapor pressure as described below.

2.3 CVD coating of TiN

One of the burner concepts employs the TiN-coated fuel particles whose surface is directly cooled by helium. The design permits the use of the cold fuel concept where the fuel temperature is kept below 1,000 K, that is, ~1/3 of the melting point of the fuel nitrides. The coating is of a duplex type: the fuel kernel is coated with low-density and high-density TiN layers. The inner low-density layer acts as a buffer to prevent the outer high-density layer from the fission recoil damage.

In JAERI, techniques for the CVD coating of the refractory ceramics such as ZrC have been developed within the high-temperature gas-cooled reactor program[13]. The techniques will be readily applied for the TiN coating. Besides, there is a range of industrial application of CVD-TiN such as a coating for cutting tools. However, there have been little experience in the CVD synthesis of the low-density TiN.

In order to demonstrate the feasibility of the CVD process for the low-density TiN coating, a study has been made jointly with Mitsubishi Materials Corporation. By using a mixture of $TiCl_4$, NH_3 and H_2, the TiN coating whose density is as low as 40%T.D. has been formed on static graphite substrates.

3. Properties of TRU nitrides

3.1 Thermal conductivity of NpN

Thermal conductivity is one of the most important properties for fuel design. Especially the information on Np is essential because of its large production and long half life time compared with other minor actinides. The thermal conductivity of NpN was determined from the thermal diffusivity measured by a laser-flash method[14].

Thermal diffusivity of NpN was measured from 740 K to 1,600 K in a vacuum on both heating and cooling runs. Details of the apparatus used were described elsewhere[15]. Thermal conductivity is given as a product of thermal diffusivity, bulk density and specific heat capacity. Unfortunately the last one of NpN is not available. Therefore, in this study the specific heat capacity, Cp is derived from the empirical equations presented by Kelley[16] and Kubaschewski[17] as follows;

$$Cp(J/mol\ K) = 52.85 + 2.55\times10^{-3}T - 8.37\times10^{5}T^{-2}.$$

Thermal conductivity of NpN is shown in fig. 2, together with those of UN and PuN[18]. The correction of porosity was made by use of the Maxwell-Eucken equation. The thermal conductivity of NpN gradually increases with temperature in a similar way to UN and PuN. The thermal conductivity of actinide mononitrides shows a tendency to decrease with the atomic number of actinides and also that the conductivity of NpN lies between those of UN and PuN.

3.2 Vaporization behavior of NpN and AmN

Vaporization behavior of actinide nitrides affects the migration of actinides during the fabrication process and the performance of failed fuel pins. From these viewpoints, the partial pressure of Np over NpN was determined by mass spectrometry with a Knudsen effusion cell.

Several mg of NpN synthesized by carbothermic reduction was used for mass spectrometry. The experimental procedure is similar to the determination of Pu pressure over PuN and (U,Pu)N described in an earlier paper[19]. The observed partial pressure of Np over NpN is shown in fig. 3, which indicates the change of the slope of the partial pressure plotted against the reciprocal of absolute temperature around 1,950 K. The phenomena was observed only at the first heating. On the following cooling and repeated runs, Np pressure was found to

be close to that over neptunium metal[20]. Therefore, it is suggested from these results that NpN evaporates congruently below 1,950 K, but at higher temperature it forms a liquid metal phase as UN does[21]. From mass spectrometric studies, PuN is reported to evaporate congruently[19,22], while UN forms a liquid phase and the partial U pressure over UN is similar to that over U metal. It could be said from the present study that NpN shows an intermediate behavior between UN and PuN.

Vaporization of Am can be a problem in the fabrication of the Am-containing fuels regardless of their chemical forms. Evaporation of americium from the nitride was also analyzed using the vaporization data on PuN containing Am[19]. The PuN sample contained about one percent Am-241 of the total heavy metal. By the analysis of the vaporization behavior, it was estimated that the second-law enthalpy of formation of AmN is -294kJ/mol at 1,600 K, which is close to those of UN and PuN.

4. Electrorefining of TRU nitrides

A pyro-chemical process has been developed for the metallic fuel cycle represented by the IFR program[23]. The introduction of the pyro-chemical process will lead to the design of a compact reprocessing facility combined with a fuel fabrication process. Recently it was indicated from thermodynamic considerations that nitride fuel might be also able to be treated by a pyro-chemical process, where actinide nitrides dissolve into fused salt as metal ions at an anode, while actinide metals are deposited at a cathode[24].

Uranium mononitride was treated by electrorefining with LiCl-KCl eutectic salt. Uranium mononitride powder was charged in a graphite anode basket. The cathode was a thin molybdenum rod. As an oxidizer, $CdCl_2$ was initially added to LiCl-KCl. Electrolysis was made at 450°C. It was confirmed that UN was completely dissolved and uranium metal was deposited on the cathode. Recently Russian researchers also reported the recovery of 500 g uranium metal from UN by a pyro-chemical process[25]. These results confirm the validity of the nitride fuel cycle combined with fused salt electrorefining.

For fast reactors, a nitride fuel cycle combined with a pyro-chemical process with fused salt electrorefining and refabrication has been proposed[2]. The same concept might be available for the nitride fuel cycle for TRU burning. At present, an electrorefiner and gloveboxes with an atmosphere of high purity argon gas are being installed in order to apply the method of fused salt electrorefining to the nitrides of Np and Pu. The preliminary experiments are planned to start in spring, 1995.

5. Summary

The recent research and development of nitride fuel and its fuel cycle for the transmutation of minor actinide are described. The fuel fabrication method investigated in JAERI is as follows:

(1)Direct conversion of actinide nitrate salt from reprocessing process to fuel particles
(2)Preparation of minor actinide nitride by carbothermic reduction
(3)Coating of TiN to fuel particles by the CVD method.

The basic fuel properties of mononitride of minor actinides were determined using the samples synthesized carbothermic reduction. It is shown that the thermal conductivity of NpN lies between those of UN and PuN. With regard to the vaporization behavior of NpN, it is suggested that NpN evaporates congruently below about 1,950 K, while it forms a liquid phase at higher temperature. The vapor pressure of Am over AmN is also estimated using the data on the Am-containing mononitride of reactor-grade plutonium. Finally, the studies on an innovative nitride fuel cycle combined with fused salt electrorefining and refabrication of particle fuel are introduced.

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Table 1 Composition of NpN synthesized by carbothermic reduction

nitrogen	carbon	Oxygen
5.70 wt%	0.09 wt%	0.03 wt%

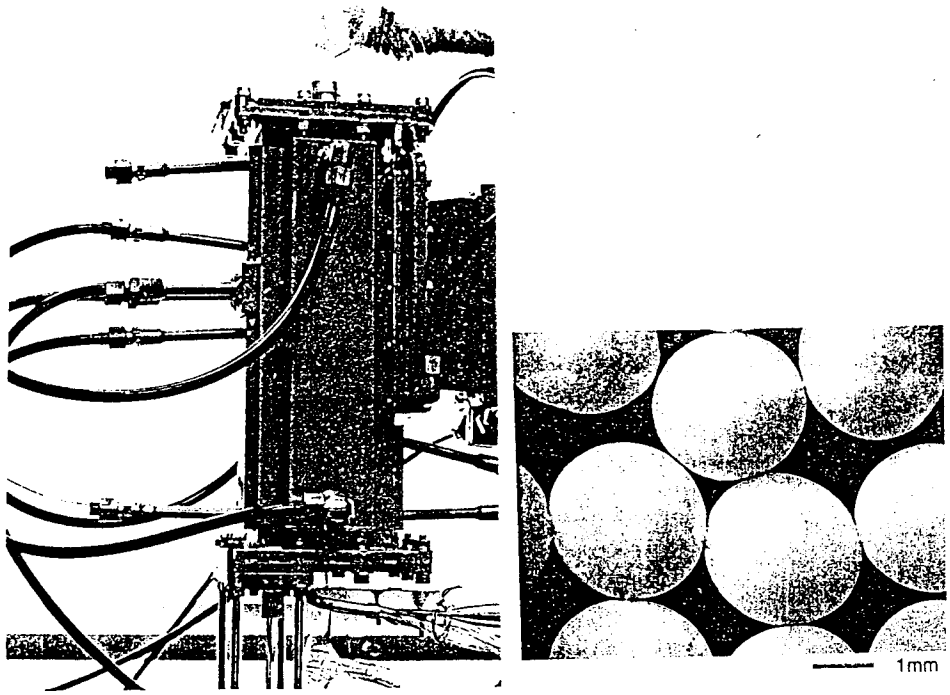

Fig. 1 A 2.45GHz microwave gelation apparatus and microspheres of ammonium uranates

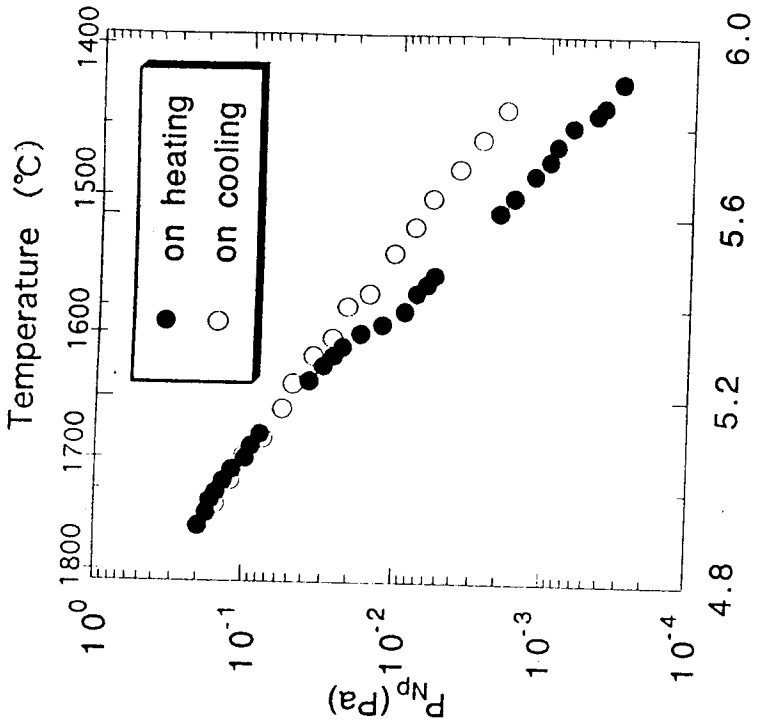

Fig. 3 Vapor pressure of Np over NpN

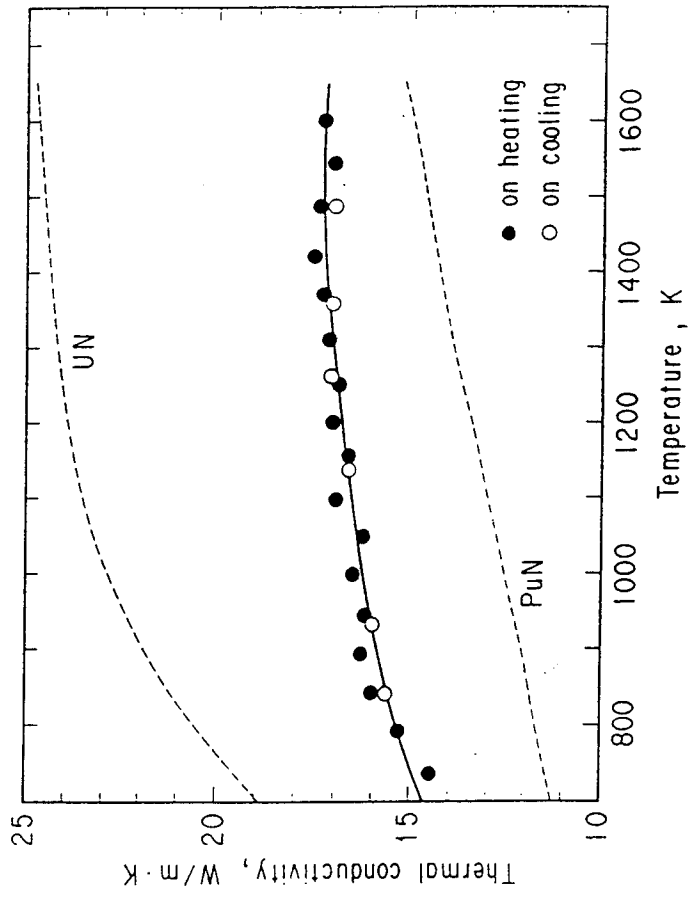

Fig. 2 Thermal conductivity of NpN

Study on Core Characteristics for MA recycling in Fast Reactors

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Abstract

Effects of core characteristics (burnup reactivity loss, Doppler coefficient, sodium void reactivity, control rod worth, power distribution, transmutation rate, breeding ratio etc.) on MA recycling were studied in a 1000MWe - class LMFBR. Minor actinides produced by the standard PWR were used in the initial core of the MA recycling. The minor actinides that have been irradiated previously but not fissioned only introduced in this recycle system from 2nd cycle. It was found that the MA recycling in an LMFBR is feasible from neutronic and thermal-hydraulic points of view. However, the Np at the 8th recycle is significantly depleted compared to the unirradiated feed, and the fraction of Cm is greatly increased because of neutron captures in Am. The accumulation of Cm by the MA recycling may bring some design issues concerning the fuel handling and reprocessing due to increase both decay heat and neutron emission rate from ^{244}Cm.

1. Introduction

Some of the transuranium nuclides (minor actinides : MA) produced by the operation of nuclear power plants have extremely long-term radiotoxicity[1], and their management is one of the key issues for nuclear power to be accepted by the public. There are some means of reducing radiotoxicity of MA nuclides under investigation. Because of the hard neutron spectrum, LMFBRs are considered to have the potentiality of transmuting MA nuclides effectively.[2-6]

The following studies are implemented to establish MA transmutation technology by an LMFBR in PNC-Japan:

(1) Feasibility studies of MA burner core concepts and evaluation of MA material balance,
(2) Nuclear data evaluation of MA nuclides in sample irradiation experiments,
(3) Measurement and evaluation of physical and chemical properties of MA compounds,
(4) Development of fabrication technology of MA fuel,
(5) Evaluation of fuel behavior by MA fuel pin irradiation experiments.

Concerning the feasibility studies of core concepts, the following items have been carried out:

1) Optimization of loading method of MA,
2) Effect of rare earth(RE) in MA on core characteristics,
3) Influence of uncertainties of MA nuclear data,
4) Influence of MA containing fuel on reactor plant and fuel cycle,
5) Effect of MA recycling on core characteristics,
6) Evaluation of properties of MA containing fuel.

In this paper, the effect of MA recycling on core characteristics in an LMFBR is presented.

2. Calculational Method and Conditions

A 1000MWe homogeneous core with two enrichment zones was employed as a reference core. The main specifications of the reference core of MOX fuel are shown in Table 1.

The minor actinides produced by the standard PWR were used in the initial core of MA recycling as shown in Table 2. The minor actinides that have been irradiated previously but not fissioned only introduced in this recycle system from 2nd cycle after 5 years of cooling time. When adding MA in the recycled cores, these new isotopes were homogeneously distributed all over the fuel in replacement of heavy nuclides of uranium and plutonium. The fuel enrichment in PuO_2 was adjusted to get the same cycle length and overall fuel residence time and the same reactivity at the end of equilibrium core. The number of MA recycling in the core life is expected 8 times.

The nuclear characteristics of MA - recycled cores were calculated by a burnup code on two-dimensional RZ geometries. Burnup characteristics, power distribution and reactivity coefficients were obtained from the analyses. Cross sections were collapsed from JFS-3-J2 library [7] based on JENDL-2 [8]. Seven-group effective cross sections were used in the calculation of burnup characteristics and power distribution. Reactivity coefficients were calculated using 18-group effective cross sections.

The core configurations of the reference core of MOX is shown in Fig. 1 .

3. Results and Discussion

The effect of MA recycling on core characteristics is shown in Table 3 . The effect of MA recycling on Pu and MA isotopic composition is also shown in Table 4. The results of the neuron emission rate and decay heat in irradiated fuels by the MA recycling are shown in Table 5. The main results of the study are summarized as follows.

(1) Burnup reactivity loss is ~1.6%Δk/kk' in the initial core, but the values from the 4th to the 8th recycle can keep around 0.4 % Δk/kk'. The influence of MA recycling on burnup reactivity loss is small in the equilibrium cores. The plutonium enrichment is reduced from 19.7wt% to 18.7wt% by MA recycling because of decreasing Np and increasing Cm nuclides which have large fission cross sections .

(2) Absolute value of Doppler coefficient becomes large by MA recycling, and the value of the 8th recycle is approximately 14% larger in comparison with that in the initial core. This be caused by the reduction of Pu enrichment by MA recycling, and the increase of resonance absorption of ^{238}U.

(3) Na void reactivity decreases with MA recycling, and the value of the 8th recycle is ~7% smaller than that in the initial core. It seems that the influence of MA recycling on Na void reactivity is generally small .

(4) Control rod worth is increased by ~5% in the 8th recycling. The influence of MA recycling on control rod worth is small.

(5) MA-loaded cores can transmute MA by ~10% , and there is not large difference in MA recycling. The amount of MA transmutation is approximately 4t for 30 years.

(6) Neptunium at the 8th cycle is significantly depleted compared to the unirradiated feed, and the fraction of Cm is greatly increased because of neutron captures in Am as shown in Table 4. Both the neutron emission rate and decay heat of the discharged fuels at the 8th recycle are very large in comparison with the fuels loaded in initial core as shown in Table 5. The accumulation of Cm by MA recycling will bring some problems concerning the fuel handling and reprocessing due to increase both decay heat and neutron emission rate from ^{244}Cm. If it is possible to remove Cm from the MA-loaded fuel, the decay heat value will decrease by one order, and the neutron emission rate by three orders. Since the dominant isotope, ^{244}Cm, has a relatively short half-life of 18 years, there might be another possibility of the fuel cycle, that is , partitioning of Cm and Am from MA in the reprocess and storing of Cm and Am for a period. Some study will be needed to estimate the trade-off between the plant modification and the reprocessing.

4. Concluding remarks

It was found that the MA recycling in an LMFBR is feasible from neutronic and thermal-hydraulic points of view . However, the Np at the 8th cycle is significantly depleted compared to the unirradiated feed, and the fraction of Cm is greatly increased because of neutron captures in Am. The accumulation of Cm by the MA recycling may bring some design issues concerning to the fuel handling and reprocessing due to increase both neutron emission rate and decay heat from ^{244}Cm.

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Table 1 Main Design Parameters of the 1000MWe Reference LMFBR

Design Parameters	Data
1. Plant Parameters	
·Reactor Thermal Power	2517 MWt
·Coolant Temperature (Reactor Outlet / Inlet)	530 / 375 °C
·Operation Cycle Length	15 Months
2. Core parameters	
·Core Concept	2-region Homogeneous
·Average Fuel Burnup	91 GWD/T
·Max. Linear Heat Rate	430 w/cm
·Core Diameter / Core Height	3.68/1.00 m
·Thickness of Axial Blanket (Upper/Lower)	0.20/0.20 m
3. Core fuel parameters	
·Fuel Composition	PuO_2-UO_2
·Pu Isotope Ratio (238/239/240/241/242)	2.4/51.1 / 26.9 / 12.0 / 7.6 (LWR Discharged)
·Pattern of Fuel Exchange	3 Dispersed Batches
4. Blanket fuel parameters	
·Fuel Composition	UO_2
·U Isotope Ratio (235 / 238)	0.3 / 99.7
·Pattern of Fuel Exchange	4 Dispersed Batches

Table 2 Composition of MA Loaded in the Initial Core

Nuclide	Data(wt%)
Np-237	49.14
Am-241	29.98
Am-242m	0.08
Am-243	15.50
Cm-242	-
Cm-243	0.05
Cm-244	4.99
Cm-245	0.26

Discharged from PWR (35GWD/T) and Cooled for 5 Years Before Reprocessing

Table 3 Effect of MA Recycling on Core Performance

Item	Initial Core	4th Recycle	8th Recycle
MA Content (wt%)	5	5	5
Pu Enrichment (Average、wt%)	19.7	19.5	18.7
Burnup Reactivity (%Δk/kk')	1.6	0.4	0.5
Maximum Linear Power (W/cc)	383	403	410
Breeding Ratio	1.12	1.26	1.23
Doppler Coefficient	1.0	1.07*	1.15*
Na Void Reactivity	1.0	0.97*	0.93*
Control Rod Worth	1.0	1.03*	1.05*
MA Transmutation Rate(%)	10.3	10.5	10.1

* Relative Value

Table 4 Effect of MA Recycling on Pu and MA Isotopic Composition

Item	Reference (Initial Core)	4th Recycle	8th Recycle
^{238}Pu	2.4	7.1	4.8
^{239}Pu	51.1	53.7	53.8
^{240}Pu	26.9	30.8	33.9
^{241}Pu	12.0	3.2	3.4
^{242}Pu	7.6	5.2	4.1
^{237}Np	49.1	18.9	8.8
^{241}Am	30.0	27.0	23.4
^{243}Am	15.5	28.0	28.8
^{244}Cm	5.0	16.2	20.1
^{245}Cm	0.3	5.9	8.8

Table 5 Effect of MA recycling on Neutron Emission and Decay Heat

Item	Reference (Initial Core)	4th Recycle	8th Recycle
Neutron Emission (n/sec/kg)	3.2×10^{7} (1.0)	6.9×10^{7} (2.2)	11.1×10^{7} (3.5)
Decay Heat (W/g)	16.5 (1.0)	25.2 (1.5)	27.3 (1.6)

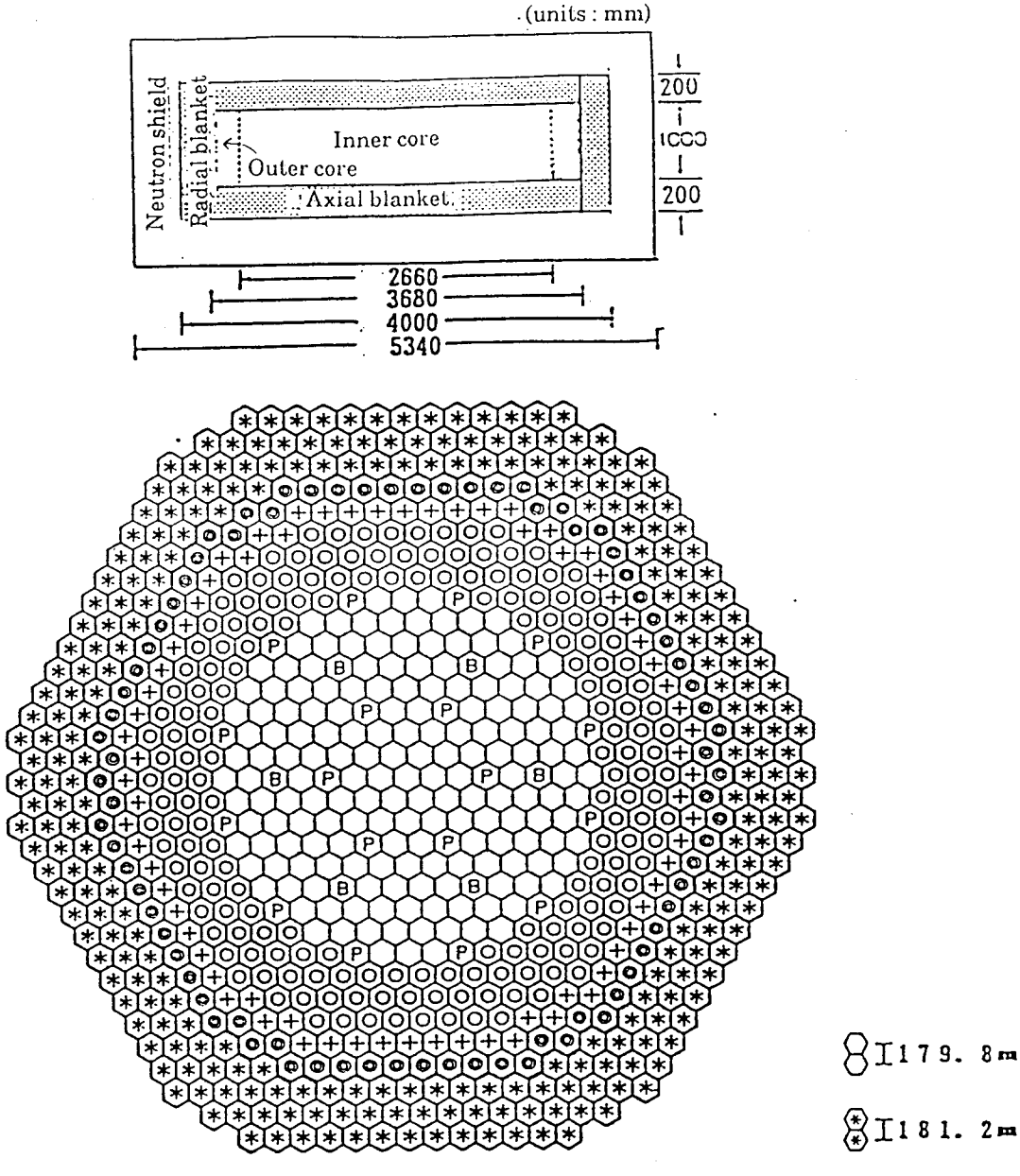

Fig. 1 Core layout of 1000MWe-class reference LMFBR

ANALYSIS OF TRANSMUTATION RATES OF LONG-LIVED RADIONUCLIDES IN THE KOREAN MULTI-PURPOSE RESEARCH REACTOR

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ABSTRACT

Transmutation rates of Minor Actinides(MA) and long-lived fission products loaded in the irradiation thimble of the Korea Multi-purpose Research Reactor (KMRR) were estimated by using the "TRIFON" code. The transmutation rates of Np-237 and a mixture of Am and Cm calculated were 65% and 75 %, respectively, over the burning period of six fuel cycles (203 days) in the central flux trap of the KMRR. However, the net burning rates, which were offset by production rates of new transuranic nuclides, were 35 % and 27 %, respectively. The transmutation rates of long-lived fission products of Tc-99 and I-129 were estimated as 13 % and 9.5 %, respectively over the same burning period. In the cases of partially replacing six central fuel elements among the 36-element assembly of KMRR by Np-237 or a mixture of Am and Cm, the transmutation rate of Np-237 is 26 % for 17.5g replacement and 20 % for 52.4g replacement. The rate of Am-241 is 46 % for 17.5g replacement and 26 % for 52.4g replacement.

1. INTRODUCTION

Several types of reactors and/or accelerators are being suggested as practical transmutation facilities in several countries. For example, the actinide burner, hybrid system of a subcritical reactor combined with an accelerator or conventional power reactors are being considered for nuclear transmutation. However, the best way of transmutation is yet to be determined. Both economic and technical advantages or disadvantages should be taken into account to select the most suitable way of transmutation.

This study is focused on the estimation of long-lived radionuclide transmutation rates in the research reactor, KMRR which will be commissioned early next year. Since the neutron flux in the core of KMRR is higher than those of conventional PWR's, it may provide more favorable conditions for the transmutation of Minor Actinides which have lower fission cross sections than those of fissile uranium and plutonium.

The computer code, TRIFON, developed in Russian Institute of Theoretical and Experimental Physics (ITEP) was used to estimate the transmutation rates of MA and long-lived fission products such as Np-237, a mixture of Am and Cm, Tc-99 and I-129.

The fission products of Tc-99 and I-129 were assumed to be placed in the central flux trap. The minor actinides of Np-237 and a mixture of Am and Cm were assumed to be either placed in the central flux trap or partially mixed up into six central fuel elements among the 36-element assembly of KMRR. Both cases were assumed to be irradiated during the period of six fuel cycles (203 days).

2. REACTOR CORE AND IRRADIATION THIMBLES OF THE KMRR [1]

The reactor physics design was carried out to achieve the maximum neutron flux with the maximum available fuel enrichment, 20% LEU and proper heat removal. The core features a symbiosis of light water reactor lattices as well as heavy water reactor lattices. This combination provides extensive variety and flexibility of neutron quality in terms of its energy and spatial distributions. Table 1 shows the design concept of the KMRR core.

As shown in Fig. 1, the inner core, an assembly of light water reactor lattices, is enclosed by the corrugated parallelepiped inner shell of the reflector tank. The 8 out of 31 sites are assigned for residences of 4 control absorbers and 4 shut off shrouds. Each of them has 18-element fuel assembly and circular flow tube. The nominal core configuration allows 20 hexagonal flow tubes to accept 36-element fuel assemblies, and 3 vacant sites are reserved for capsule, rig, or loop installations.

The outer core is clustered with 8 circular flow tubes vertically passing through the reflector tank at the narrow sides of the inner core periphery. Loaded with 18-element fuel assemblies, the H_2O-cooled and D_2O-moderated outer core enhances total core excess reactivity. The outer core sites, used for irradiation purpose, provide high epithermal neutron fluence.

The central trap, one of the three flux traps in the inner core, has the highest flux, 5.3×10^{14} n/cm^2 s. It will be valuable to install the fuel and material test loop. The D_2O-filled reflector tank is equipped with a number of vertical irradiation thimbles. Their sizes and locations are optimized to maintain the required neutron quality and level without significant disturbance by the reactor operating condition changes and other near-by experiments.

One fuel cycle consists of 28 days' burning and 7 days' shutdown which is necessary for fuel exchange. Most of 36-element assemblies and all of 18-element assemblies are burned up for 6 fuel cycles, but part of the 36-element assemblies are used for 7 fuel cycles. Material and specifications of KMRR standard fuel are shown in Table 2.

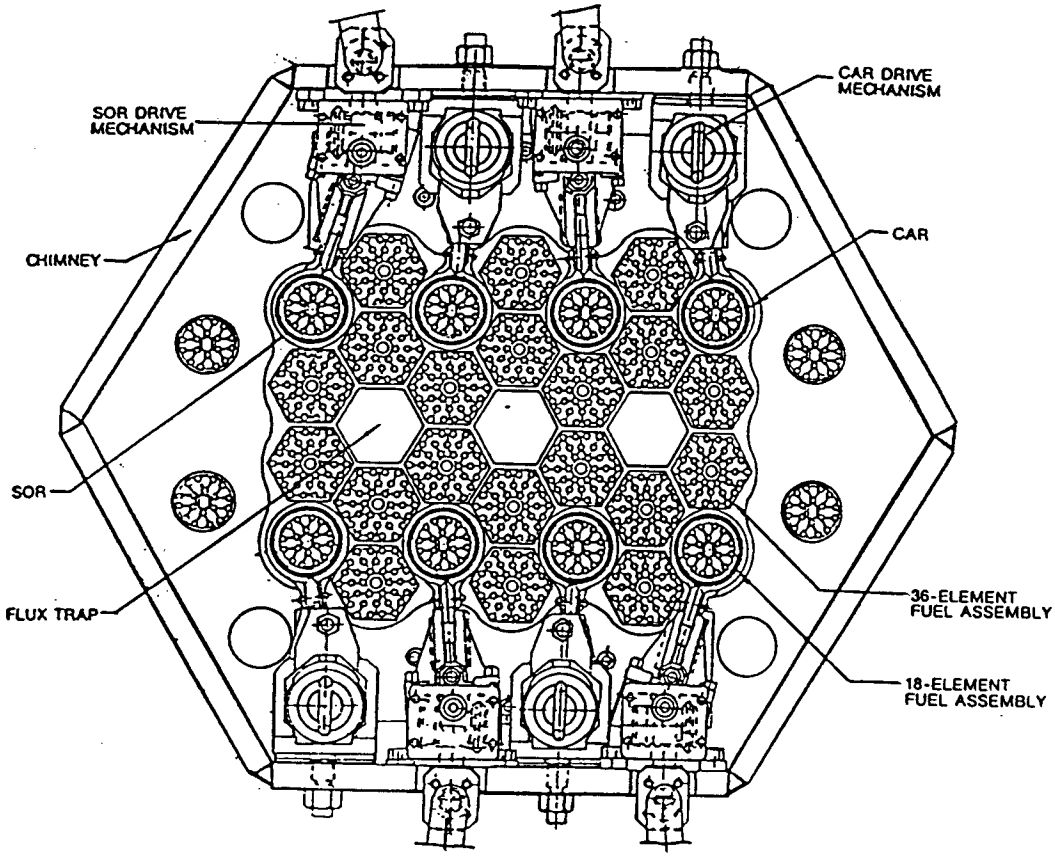

Fig.1 Schematic diagram of KMRR core

Table 1. Major design concept of KMRR

Reactor Type	Open-Tank-In-Pool, LWR/HWR Hybrid
Thermal Power	30 MWth
Fuel	U_3 Si-Al, 19.75 W/O ^{235}U Enriched
Coolant	H_2O
Moderator	H_2O/D_2O
Reflector	D_2O
Cooling Method	Convective Up-Flow
Secondary Cooling	Cooling Tower
Reactor Building	Confinement
Power Regulation and Xenon Poison Compensation	Four Hafnium Tubes Sliding over the 18-Element Fuel Assembly
Shutdown System	Four Hafnium Tubes Sliding over the 18-Element Fuel Assembly. Dropped by Gravity
Heat Generated in Reactor Fuel	27.5 MW
Heat Generated in Moderator and Structure	Approx. 2.5 MW
Total Fission Power	30.0 MW
Heat Removed by Primary Coolant System	27.5 MW

Table 2. Specifications of KMRR standard fuel

Composition Rates in Fuel Meat (W/O)	
^{235}U	11.7
^{238}U	46.9
Si	2.4
Al	39.0
Fuel Meat	
enrichment(w/o)	19.75
density(gr/cc)	5.4
diameter(mm)	6.35/5.5
length(mm)	700.0
mass of uranium(gr)	69.8/52.4
LEU density (gr/cc)	3.15
Cladding	
material	Al(co-extruded)
thickness(mm)	0.76/1.19
Fin	
number	8
height(mm)	1.02
width(mm)	0.76

3. TRIFON CODE [2]

The one-dimensional code TRIFON, which is based on the collision probability method was developed in ITEP, Russia. Neutron energy spectrum and spatial distribution of neutron flux can be computed with this model and transmutation rates of various nuclides can also be estimated. The cross section data, used in this code, are mainly from the Russian ABBN and also partly from the other additional information. They are composed of 26 group cross section data and resonance parameters.

ITEP validated this code by comparing the calculation results from other codes, as shown in Table 3 [3]. The results of this code are closer to the results of Monte-Carlo simulations than that of WIMS code. Also, the quantities of uranium and transuranium contained in LWR spent fuel were also calculated with this code. The comparison of the calculated results with experimental results is shown in Fig. 2. The agreement between the calculated and experimental results is quite good. On the basis of this demonstration, ITEP carried out transmutation studies with this code for the heavy water reactor, where high enriched uranium is used as fuel, as well as the thorium fuel cycle reactor[4,5].

Table 3. Comparison of the parameter values calculated from TRIFON and other codes

Lattice	Parameter	TRIFON	WIMS	EPRI-CELL	Monte Carlo(%SD)
NB-1, Low Enrichment	K_∞	1.1344	1.254	1.1449	1.1471(0.14)
	ρ_{28}	1.375	1.458	1.360	1.363(0.6)
	CR	0.804	0.829	0.796	0.798(0.4)
NB-2, (PU/U=0.02)	K_∞	1.1734	1.1640	1.1698	1.1748
	ρ_{28}	2.526	2.372	2.613	2.612
	CR	2.110	2.319	2.148	2.148
NB-4, PWR Type	K_∞	1.3363	-	1.3415	1.3424(0.26)
	ρ_{28}	2.666	-	2.632	2.654(0.6)
	CR	0.556	-	0.548	0.549(0.4)
NB-5. Hard Spectrum	K_∞	1.1302	-	1.1421	1.1456(0.15)
	ρ_{28}	8.452	-	8.534	8.503(0.4)
	CR	1.015	-	1.006	1.006(0.3)

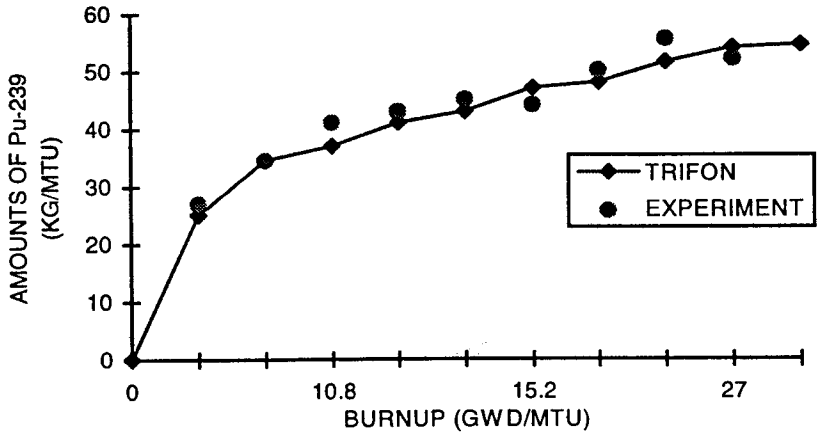

Fig. 2 Comparison of the experimental and calculated results for the amount of Pu-239 with burnup in PWR.

4. TRANSMUTATION RATE ANALYSIS IN THE KMRR

4.1. Calculation Model

For the convenience of calculation, in the transmutation study with the central irradiation thimble the shapes of hexagonal thimble, fuel assembly tubes, and surrounding moderator were assumed to be the annular rings with corresponding equivalent volumes. The schematic of the cell model reflected by these assumptions is illustrated in Fig. 3. The net current was assumed to be zero at the outer boundary of six assemblies around the central trap in the core.

In the transmutation study with minor actinides partially mixed up into six central fuel elements out of the 36-element fuel assembly as shown in Fig. 4, the central support rod, moderator, cladding, structure, and MA and fuel were also assumed to be the annular rings with corresponding equivalent volumes in the order of their locations.

4.2. Calculation and Results

The transmutation rate of Np-237 with burning time calculated by TRIFON code was compared, in Fig. 5, with that obtained from WIMS-KAERI code [6] which had been used in the design of KMRR. The two results were found to be in accordance within 7% error.

Small quantities of long-lived radionuclides were considered to be loaded as the target to minimize the impact on core behavior. The atomic number density of a target nuclide is fixed as $0.0001/cm^2$ barn. Since the code was developed on the basis of one dimension, the thermal power was converted to the power per unit length. As noted in previous section, one fuel cycle is composed of 28 days' burning and 7 days' shutdown in the KMRR. Thus the thermal power during the irradiation period of six fuel cycles is assumed as 30 x (168/203)/70 ≈ 0.355 MW/cm.

Fig. 6 shows the transmutation rate of small quantity of Np-237 in the central flux trap. The transmutation rate was estimated as 64.6% and the net burning rate, offset by the production of new transuranium from the Np target, was 34.5%.

Fig. 7 illustrates the transmutation rates of a mixture of Am and Cm in the central flux trap with burning time when loaded as the same component ratio of the quantity as in typical PWR spent fuel at 10 year cooling time. The amount of Am-241 as well as Am-243 was found to decrease with time whereas that of total Cm nuclides increases due to the transmutation of Am into Cm by neutron capture and then β-decay during the irradiation. Consequently, the total net burning rate of Am and Cm was 27.3% during the irradiation period of six fuel cycles.

On the other hand, the transmutation rates of Tc-99 and I-129 loading in the central flux trap were 13% and 9.5%, respectively, as shown in Fig. 8. They were lower than those of minor actinides due to lower neutron cross sections of Tc-99 and I-129.

In the cases of partially mixing up Np-237 or a mixture of Am-Cm into six central fuel elements out of the 36-element assembly of KMRR fuel, the transmutation rate of Np-237 is 26 % for 17.5 g replacement and 20 % for 52.4 g replacement as shown in Fig. 9. The rate of Am-241 is 46 % for 17.5 g replacement and 26 % for 52.4 g replacement as shown Fig. 10. As the amounts of test materials inserted in the fuel assembly are increased, neutrons are captured in test materials considerably and the transmutation rates in such cases are decreased.

The estimated transmutation rates are much lower than those from the transmutation in the irradiation thimble because of the 4~5 times lower neutron flux.

5. CONCLUSIONS

It was found that transmutation of minor actinides in the KMRR can be achieved to some extent, showing the possibility for using KMRR as a facility for transmutation test. The calculation results obtained from this study can also be used as basic information for target design or transmutation test.

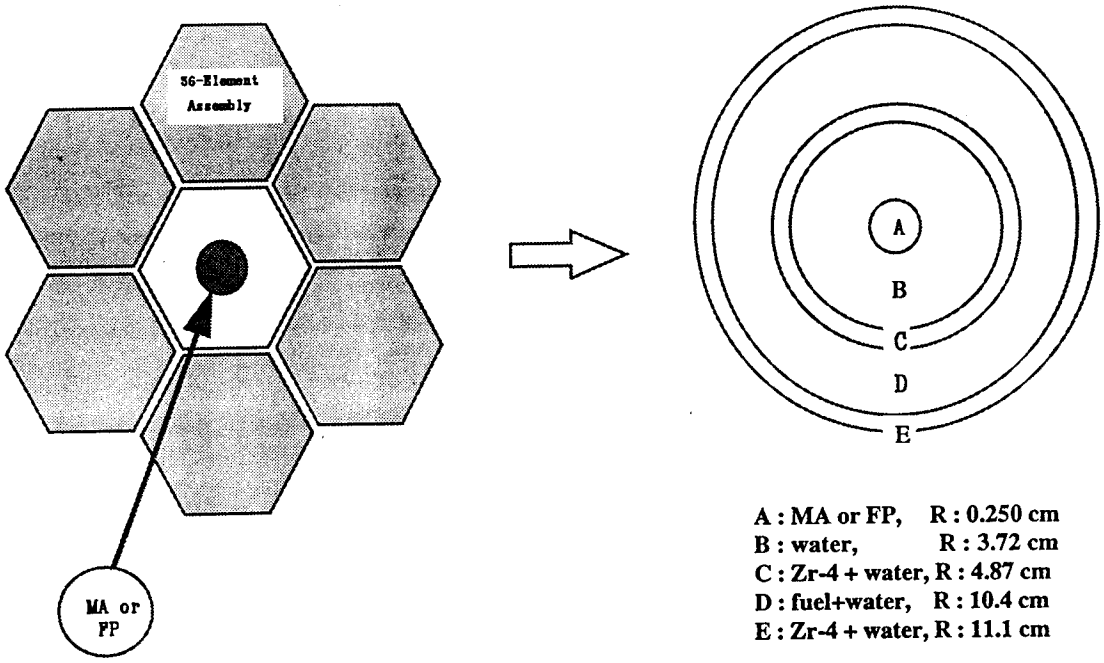

Fig. 3. Cell calculation model of central flux trap and six 36-element Assemblies

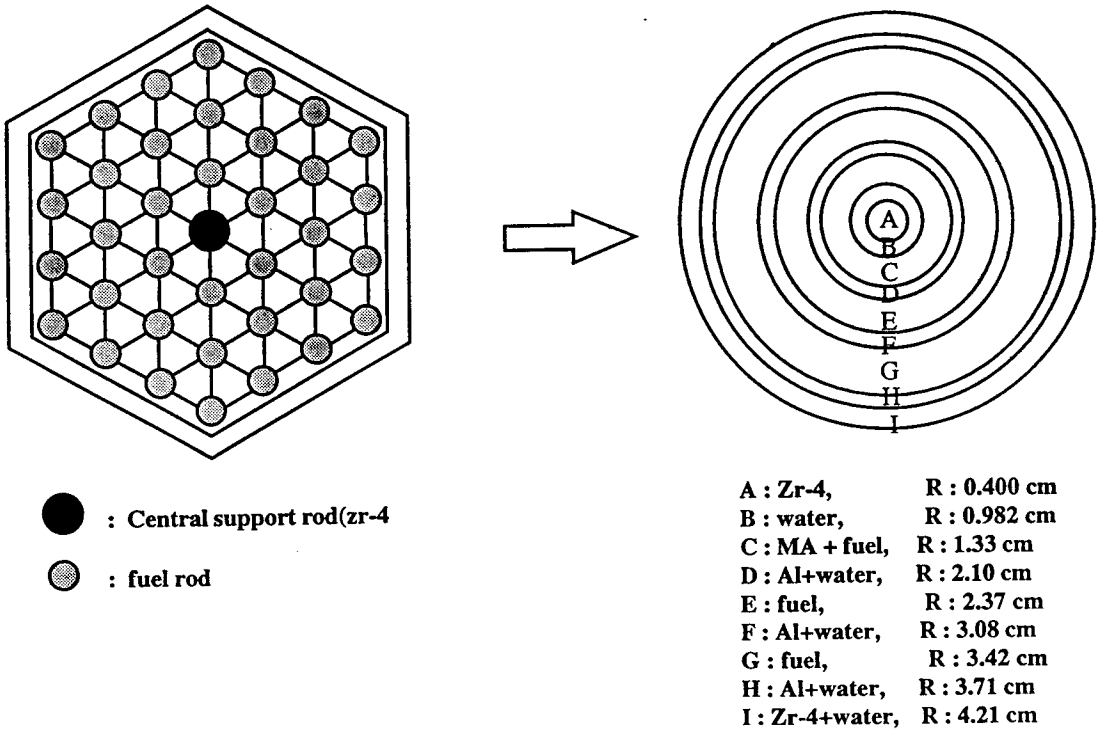

Fig. 4. Cell calculation model of 36-element assembly with partly replacment of fuel by MA

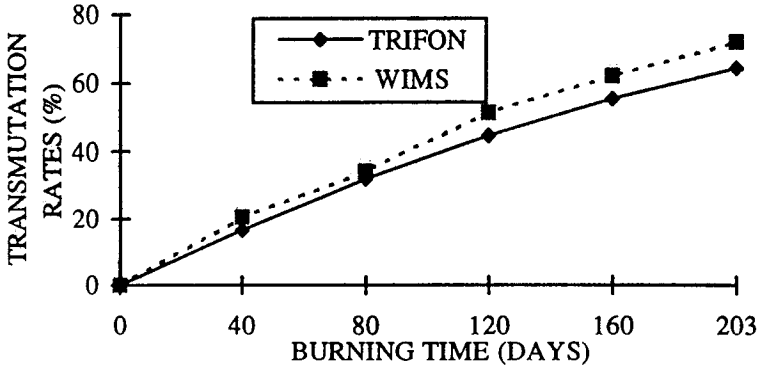

Fig. 5 Comparison of the calculated results of TRIFON and WIMS codes for transmutation rate of Np-237

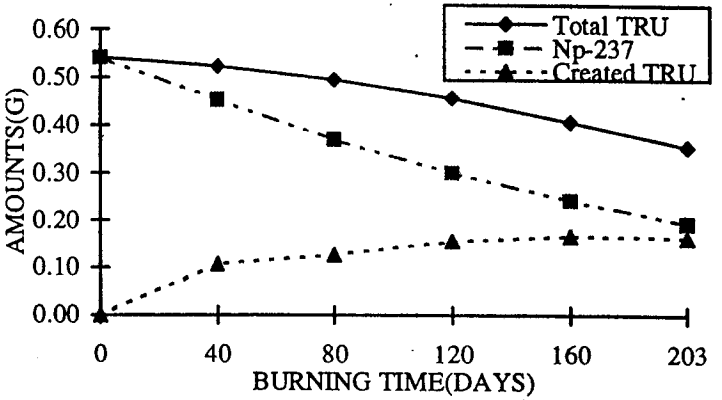

Fig. 6 The changes in the amounts of Np-237 and total TRU with burning time.

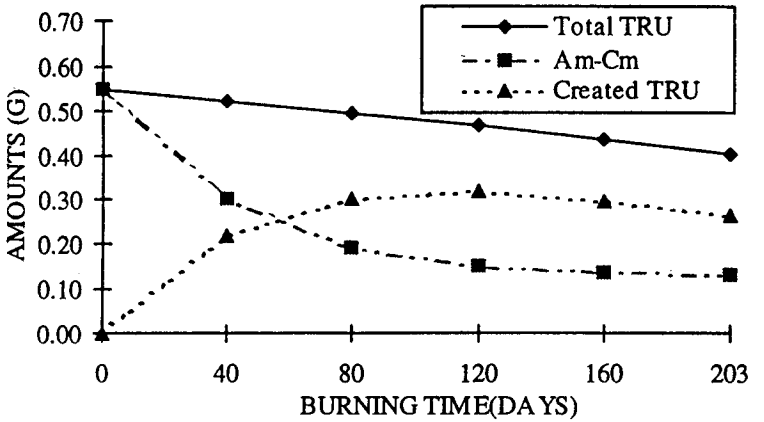

Fig. 7 The changes in the amounts of Am-Cm and total TRU with burning time.

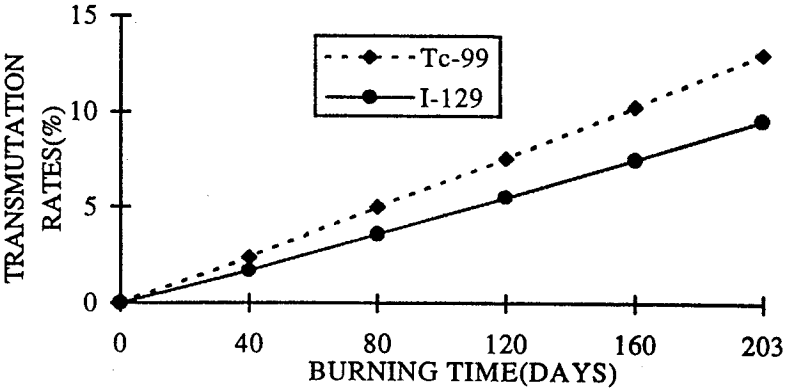

Fig. 8 Transmutation rates of Tc-99 and I-129 with burning time.

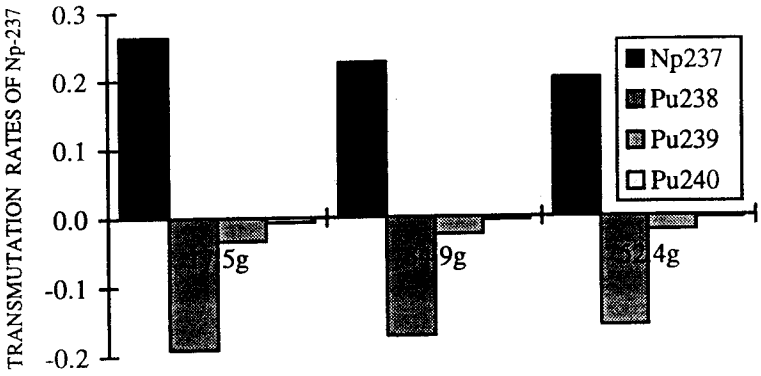

Fig. 9 Transmutation rate of Np-237 with loading quantity in the case of replacement of KMRR fuel

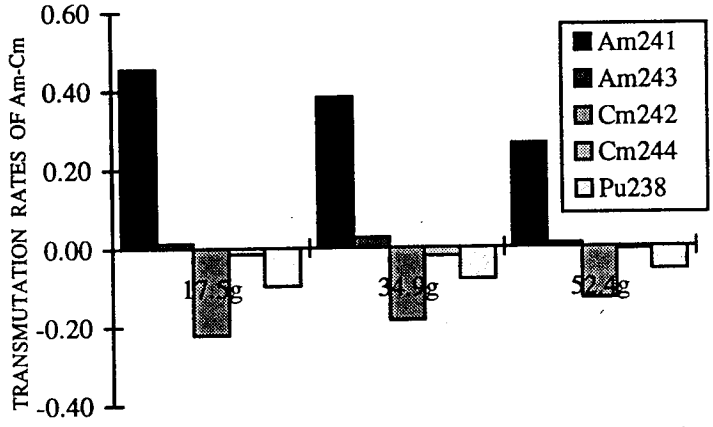

Fig. 10 Transmutation of Am-Cm with loading quantity in the case of replacment of fuel.

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TRANSMUTATION OF AMERICIUM IN THERMAL REACTORS

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Abstract
If all plutonium is partitioned from spent fuel and transmuted, then americium and its daughter products are the main contributors to the radiotoxicity of spent fuel up to several thousands of years of storage. This paper describes the possible benefits when americium is irradiated in a thermal reactor.
After one million years of storage, Np-237 dominates the possible dose to the population due to leakage of actinides from underground disposal sites. Because Am-241 decays to Np-237, the radiotoxicity due to Np-237 is reduced by about 50% when Am-241 is transmuted to other nuclides upon irradiation in a thermal reactor.
Secondly, the radiotoxicity of an americium sample can be reduced with a factor of 10 to 20 after irradiation for 3 to 6 years in a thermal neutron flux of 10^{14} $cm^{-2}s^{-1}$. The strongly alpha and neutron emitting transmutation products can most probably not be recycled again, so a process is suggested in which americium is first irradiated and then put to final storage. It is shown that after a storage time of about one hundred years, the radiotoxicity of an irradiated americium sample is considerably reduced compared to the radiotoxicity of an unirradiated sample. The same holds for the alpha activity and the heat emission. This statement has been confirmed by a calculational case study on an americium containing sample, which was supposed to be irradiated for about three years in an experimental facility in the Petten High Flux Reactor.

Introduction

Partitioning and Transmutation (P&T) of actinides are generally said to have three main objectives:

1 reduction of the possible dose to the population due to leakage of actinides from underground disposal sites;
2 reduction of the total radiotoxic inventory of underground disposal sites;
3 using fissile actinides for energy production.

Each of these three objectives has its specific impact for the P&T strategy to be followed. The first objective implies that the initial inventory of Np-237 and its precursors shall be reduced, because Np-237 is a nuclide which is easily transported to the biosphere without much adsorption in the underground once it is released from the underground disposal site. Studies on underground disposal options in salt domes show that Np-237 dominates the possible dose to the population living some millions of years from now [1].

The second objective is met by heavily reducing the total amount of actinides in High Level Waste (HLW). It is believed that reducing the radiotoxic inventory per disposal site or reducing the number of disposal sites has some positive impact on the public opinion with regard to the nuclear waste problem. It also leads to lower probabilities of human intrusion in repositories and to reduced accompanied doses received by men after intrusion. For human intrusion scenarios the contribution of americium to the dose is dominant [1]. Also from the economic point of view it is advantageous to reduce the inventory or the number of underground disposal sites to be licensed. This can be accomplished by reduction of the heat emission of the waste.

The third objective is met by fissioning of actinides in nuclear energy parks with thermal and fast reactors. This was the main reason to apply reprocessing. Future incentives for partitioning of actinides, especially of plutonium, depend primarily on future prospects of nuclear energy, which are highly affected by political standpoints.

This paper attempts to give incentives for heterogenous transmutation of americium in thermal reactors based on the first two given objectives. It deals explicitly with transmutation of americium in thermal reactors. Although fast reactors may be more appropriate for transmutation of americium, it is expected that only few fast reactors will be available in the next decades of years. Unless otherwise stated, calculations have been done with the fuel depletion code ORIGEN-S [2] with accompanied data libraries based on JEF2.2 and EAF3 [3]. Radiotoxicities are based on Annual Limits on Intake (ALI) values for ingestion based on ICRP-61 [4]. References 5 and 6 give more details about the material presented in sections 2 to 5, and 6, respectively.

2 Production of americium

Table 2.1 gives the yearly produced masses of neptunium, plutonium, americium and curium in the Borssele Nuclear Power Plant (The Netherlands), which has an electric power of 450 MW. It is seen that americium is produced in only small quantities during reactor operation. However, it is produced in much larger quantities in spent fuel due to decay of Pu-241 with a half life of 14.7 years. The yearly produced quantities of americium as a function of the interim storage time after unloading are given in Table 2.2. It is seen that the yearly produced Am-241 mass increases from 0.7 kg at EOC to 8.2 kg after 10 years of interim storage due to decay of Pu-241. From these numbers it follows that plutonium once partitioned from spent fuel should not be stored for a long time, because a large fraction of Pu-241 will have decayed to Am-241, which decreases the fraction of fissile isotopes in the plutonium.

If plutonium is not used in MOX fuel, all Pu-241 will eventually decay to Am-241, leading to a yearly produced Am-241 mass of 20.6 kg. Because all Am-241 will eventually decay to Np-237, fissioning of Pu-241 in MOX fuel seems the most effective way to prevent the buildup of Np-237. Because partitioning of plutonium is done only some years after interim storage of spent fuel, the amount of Am-241 produced during interim storage equals about the yearly produced amount of Np-237 (4.9 kg). To meet the first objective (reduction of the possible dose to the population due to leakage of actinides from underground disposal sites) transmutation of Am-241 seems as important as transmutation of Np-237 itself. Unless otherwise stated, all results in the remainder of this paper refer to an initial amount of 4.9 kg Am-241 and 0.9 kg Am-243, corresponding with an interim storage time of five years. It must be kept in mind, however, that the yearly produced Am-241 mass changes when another interim storage time is used.

3 Radiotoxicity of americium

Figure 3.1 gives the radiotoxicity of the yearly produced spent fuel (12.5 t) in the Borssele NPP compared with the radiotoxicity of the total uranium ore needed for manufacturing this fuel (82.2 t). The radiotoxicity after several hundreds of years is completely determined by the actinides in the spent fuel. The contribution of each

actinide element and its daughter products to the radiotoxicity of actinides is shown in Figure 3.2. It is seen that plutonium is the main contributor (80 to 95%) up to one million years of storage. Americium contributes for about 20% up to 1,000 years, and the initial amounts of Am-241 and Np-237 equally contribute after 2 10^5 years of storage.
When partitioning of plutonium is already adopted, as is the case for many countries in Europe, and when the radiotoxicity of plutonium can be considerably reduced by transmutation, americium is the most important nuclide dominating the radiotoxicity of the HLW up to 50,000 years.

Table 2.1: *Yearly produced masses of neptunium, plutonium, americium and curium in the Borssele NPP.*

Nuclide	Half Life (a)	Mass (kg)
Np-237	2.14 10^6	4.9
Pu-238	8.78 10^1	1.6
Pu-239	2.41 10^4	79.5
Pu-240	6.54 10^3	28.2
Pu-241	1.47 10^1	19.9
Pu-242	3.76 10^5	5.5
Am-241	4.32 10^2	0.7
Am-243	7.39 10^3	0.9
Cm-242	4.46 10^{-1}	0.2
Cm-244	1.81 10^1	0.2

Table 2.2: *Yearly produced americium masses (kg) in the Borssele NPP as a function of the interim storage time after unloading.*

Nuclide	Storage Time (a)		
	2	5	10
Am-241	2.5	4.9	8.2
Am-243	0.9	0.9	0.9
Total	3.4	5.8	9.1

4 Transmutation of americium

Introduction
The most abundant americium isotope in spent fuel after a few years of interim storage is Am-241. Because it is not fissile, it is transmuted by neutron capture to Am-242m (10%) or to Am-242 (90%). The first-mentioned activation product has a relatively long half life of 141 years, is highly fissile, and can therefore easily be fissioned in a thermal neutron flux. The second activation product also has a high fission cross section, but decays with a half life of 16 hours to Cm-242 (83%) or Pu-242 (17%). For the Am-242 fission rate being equal to the decay rate, a thermal neutron flux as high as 7 10^{15} $cm^{-2}s^{-1}$ is required.
The transmutation product Cm-242 decays to Pu-238 with a half life of 163 days, but can also be transmuted by neutron capture to the fissile isotope Cm-243. A thermal neutron flux as high as 5 10^{15} $cm^{-2}s^{-1}$ would be needed

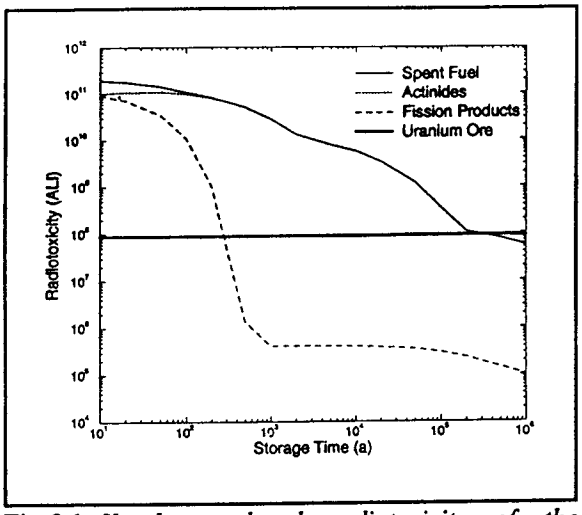

Fig 3.1: *Yearly produced radiotoxicity of the Borssele NPP as function of storage time.*

for the capture rate being equal to the decay rate. The formed amount of Pu-242 can be transmuted into the fissile Pu-243. However, this plutonium isotope decays with a half life of 5 hours to Am-243. In case of irradiation of Am-241 in low thermal neutron fluxes and moderate values of the neutron fluence, transmutation will result in production of Pu-238 (75%) and Pu-242 (15%), and fissioning of Am-242m (10%).

Transmutation of Am-241 in thermal reactors seems only attractive in a high thermal neutron flux of about 10^{16} $cm^{-2}s^{-1}$. The argument that transformation of Am-241 into short-lived Pu-238 is beneficial because it would reduce the production of 'mobile' Np-237 (the first objective mentioned in section 1) violates the second objective since the radiotoxicity or heat emission is not reduced. However, there could still be some incentive to transmute Am-241 in a thermal reactor when a very high neutron *fluence* (flux times irradiation time) is achieved. Then there will be successive transformation of the formed Pu-238 and Pu-242. The produced Pu-238 can be further transmuted to fissile Pu-239 and even Pu-241. However, as explained above, the formed Pu-242 is mainly transmuted to Am-243, and this amount of Am-243 (max 15% of the initially present Am-241) together with the initially present Am-243 (about 0.9 kg) is again transformed to Cm-244 upon neutron capture. Cm-244 is a short-lived neutron emitting isotope and another neutron capture would be necessary to transmute this isotope into fissile Cm-245. The whole process is illustrated in Figure 4.1, where the fission rate as a function of time is shown for 4.9 kg Am-241 and 0.9 kg Am-243 being irradiated in a thermal neutron flux of 10^{14} $cm^{-2}s^{-1}$.

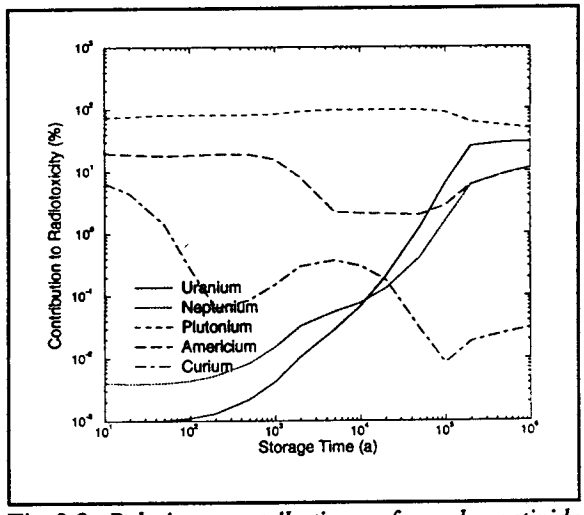

Fig 3.2: *Relative contribution of each actinide element and its daughters products to the radiotoxicity of actinides in spent fuel.*

Transmutation of americium in thermal reactors leads to a mix of nuclides with a high α activity and a high spontaneous fission rate, which is practically impossible to reprocess, and which can not be used for manufacturing of new targets or fuel assemblies. Successive reprocessing is avoided by adapting a transmutation scheme in which the irradiated americium is disposed of.

First objective

To meet the first objective mentioned in section 1 (reduction of the possible dose to the population due to leakage of actinides from underground disposal sites) the amount of Np-237 and its precursors (Am-241, Pu-241 and Cm-245) should be reduced as much as possible. Table 4.1 gives the sum of radiotoxicities due to Np-237 and its precursors originating from americium after irradiation with a thermal neutron fluence of about 1.9 10^{22} cm^{-2} (corresponding with an irradiation of 6 years in a thermal neutron flux of 10^{14} $cm^{-2}s^{-1}$), and after a storage time of one million years. Almost all initial Am-241 is transmuted, and the radiotoxicity due to Np-237 and its precursors in the irradiated americium after a storage time of one million years is reduced with a factor of 100. It must be noted however that Am-241 in spent fuel contributes for 50% to the radiotoxicity due to Np-237 after one million years, and that transmutation of Np-237 itself, also with yearly produced mass in the Borssele NPP of 4.9 kg (see Table 2.1), would be necessary to reduce the other contribution of 50%. This latter percentage

decreases when longer interim storage times are applied for the spent fuel (see Table 2.2).

Second objective

The second objective (reduction of the total radiotoxic inventory of underground disposal sites) can be met by reducing the radiotoxicity and the α activity of the waste. In Figure 4.2, the radiotoxicity due to americium is shown before and after irradiation in a neutron flux with a thermal neutron fluence of 9.5 10^{21} cm^{-2} (corresponding with an irradiation of 3 years in a thermal neutron flux of 10^{14} $cm^{-2}s^{-1}$). After about 70 years, the radiotoxicity due to the irradiated americium is lower than the radiotoxicity due to the unirradiated americium. It is also seen that the radiotoxicity due to the irradiated americium is lower for lower thermal neutron fluxes. This is mainly due to the fact that Cm-242 decays with half life of 163 days and that lower neutron fluxes lead to longer irradiation times when the neutron fluence is kept constant. More Cm-242 can be transmuted to Pu-239 (via α decay and subsequent neutron capture in Pu-238), and subsequently be fissioned when the irradiation time is longer.

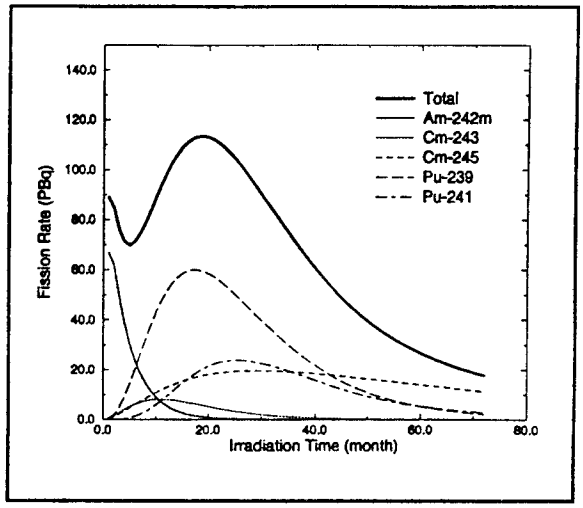

Fig 4.1: *Fission rate as function of time of 5.8 kg americium irradiated in a thermal neutron flux of 10^{14} $cm^{-2}s^{-1}$.*

Figure 4.3 gives the similar curves for a thermal neutron fluence which is twice as large; thus corresponding with an irradiation of 6 years in a thermal neutron flux of 10^{14} $cm^{-2}s^{-1}$. The radiotoxicity due to the irradiated americium is already within 10 years lower than the radiotoxicity due to the unirradiated americium. For irradiation in thermal neutron fluxes of 10^{14} $cm^{-2}s^{-1}$ and lower, the radiotoxicity due to the irradiated americium after 100 years of storage is about a factor of 20 lower than the radiotoxicity due to the unirradiated americium. It must be noted that Figures 4.2 and 4.3 give the radiotoxicity as function of storage time after irradiation. This means specifically that the irradiation time corresponding with each thermal flux is neglected.

Table 4.1: *Total radiotoxicity and radiotoxicity due to Np-237 and its precursors after irradiation of 5.8 kg americium with a thermal neutron fluence of 1.9 10^{22} cm^{-2} and after a storage time of one million years.*

Thermal Flux	Radiotoxicity (ALI)	
($cm^{-2}s^{-1}$)	Total	Np-237+Precursors
0	6.1 10^{6}	6.1 10^{6}
4 10^{14}	7.0 10^{5}	7.5 10^{4}
2 10^{14}	3.8 10^{5}	7.0 10^{4}
1 10^{14}	2.7 10^{5}	6.4 10^{4}
4 10^{13}	2.5 10^{5}	6.3 10^{4}
2 10^{13}	2.6 10^{5}	6.4 10^{4}

Apart from other reasons, the second objective (reduction of the total radiotoxic inventory of underground disposal sites) may have some economic benefit when the number or the size of underground disposal sites can be reduced. Because the maximum amount of HLW to be stored at one disposal site is mainly determined by the total heat emission of the waste, and because the heat emission of actinides is mainly determined by α activity, it is assumed that the α activity at the time of disposal determines the maximum amount of actinide material

which can be stored at one specific disposal site. The α activity of the americium decay chain as a function of time is shown in Figure 4.4. It is seen that a considerable reduction of the α activity after about 100 years of storage can be achieved by irradiation with a thermal neutron fluence of 1.9 10^{22} cm^{-2}.

Table 4.2: *The β⁻ activities of the yearly produced fission products of the Borssele NPP after a storage time of 50, 100 and 200 years.*

Time (a)	β⁻ activity (PBq)	Contribution (%)		
		Sr-90	Y-90	Cs-137
50	37	29.1	29.1	40.7
100	11	28.8	28.8	41.6
200	1.1	27.0	27.8	42.0

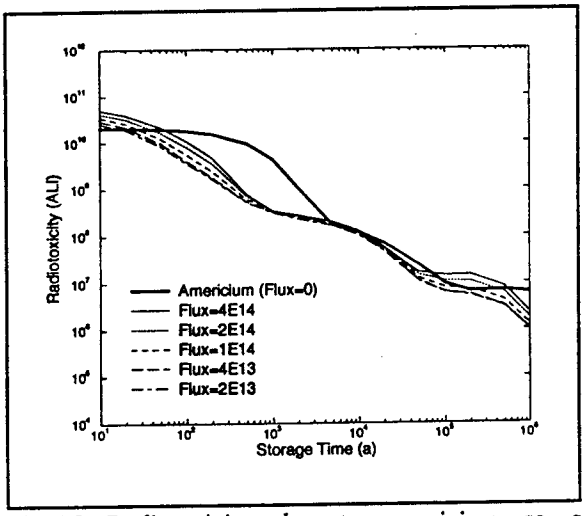

Fig 4.2: *Radiotoxicity due to americium as a function of storage time after irradiation with a thermal neutron fluence of 9.5 10^{21} cm^{-2}.*

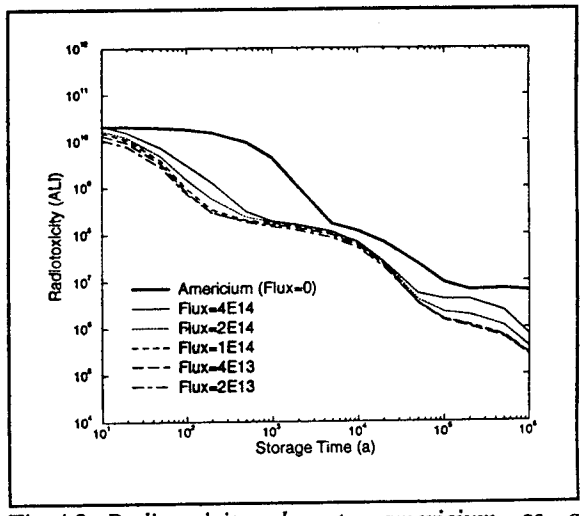

Fig 4.3: *Radiotoxicity due to americium as a function of storage time after irradiation with a thermal neutron fluence of 1.9 10^{22} cm^{-2}.*

The heat emission of the actinides has to be compared with the total heat emission of the deposited waste, which is mainly determined by the β⁻ activity of the fission products. This activity is given in Table 4.2 for storage times of 50, 100 and 200 years. It is seen that the main contributors to the β⁻ activity are Sr-90, Y-90 and Cs-137. Because the proportional contributions of these isotopes to the β⁻ activity are almost independent of time, the average emitted energy per β⁻ decay for this mix of isotopes is constant and equals about 0.4 MeV. In Table 4.3, the average energy emission by β⁻ and γ decay of the yearly produced fission products of the Borssele NPP are given and compared with the energy emission by α decay of americium, for which it is assumed that the average emitted energy per α decay is 5.5 MeV. After a storage time of 100 years, americium contributes for about 30% to the heat emission. This percentage increases up to 80% after a storage time of 200 years. Therefore, transmutation of americium in order to reduce the heat emission of the waste at the time of final disposal seems only meaningful when an interim storage time of 100 years or longer is applied to fission products during which they are put to temporary storage (e.g. above ground). This interim storage time is about the same as the storage time required to get a considerable reduction of the α activity after transmutation of americium (see Figure 4.4). The α activity at the time of final disposal can be reduced with about 90% and the total heat emission with about 25% if an interim storage time of 100 years is applied, during which both fission products and transmutation

products of americium are put to temporary storage.

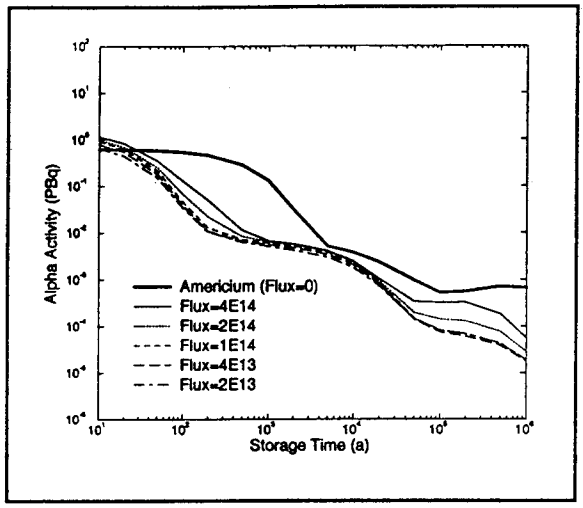

Fig 4.4: *The α activity due to americium as function of storage time after irradiation with a thermal neutron fluence of 1.9 10^{22} cm^{-2}.*

5 Inert Matrices

The radiotoxicity due to americium is only reduced considerably when a large percentage (about 75%) of the initial amount of americium has been transmuted into fissile isotopes and eventually fissioned. Such high burnups cannot be reached when americium is irradiated as pure Am_2O_3 with density of 10 g cm^{-3}, because damage to the Am_2O_3 matrix due to recoil and buildup of fission products will limit the burnup to much lower values. Therefore, americium should be irradiated at low concentrations, which can be achieved by diluting americium in some other matrix. Uranium oxide cannot be used as a matrix because the buildup of fissile plutonium isotopes and subsequent fissioning of these isotopes will considerably limit the burnup of americium. Multiple recycling of americium and transmutation products is necessary then. This is a strong incentive to consider transmutation of americium at low concentrations in some neutron inert matrix (e.g. Al_2O_3). Diluting americium in inert matrices can also be used to control the power density in the target containing the americium. Furthermore the relative transmutation efficiency increases considerably when americium is diluted. Of course, for the extended irradiation times assumed in this paper, there are also huge requirements on the cladding, which preferably is made of stainless steel.

Table 4.3: *Energy emission of the yearly produced fission products (only $\beta^-+\gamma$ decay) and americium (only α decay) of the Borssele NPP after storage times of 50, 100 and 200 years.*

Time	Energy Emission (MeV s^{-1})				
(a)	β^-	γ	$\beta^-+\gamma$	α	$\alpha+\beta^-+\gamma$
50	$1.5\ 10^{16}$	$8.6\ 10^{15}$	$2.4\ 10^{16}$	$3.2\ 10^{15}$	$2.7\ 10^{16}$
100	$4.4\ 10^{15}$	$2.7\ 10^{15}$	$7.1\ 10^{15}$	$3.0\ 10^{15}$	$1.0\ 10^{16}$
200	$4.4\ 10^{14}$	$2.7\ 10^{14}$	$7.1\ 10^{14}$	$2.5\ 10^{15}$	$3.2\ 10^{15}$

6 Calculational Case Study for Irradiation in the Petten High Flux Reactor

Model

In the framework of the EFTTRA cooperation [11], calculations have been done for a specific experimental facility containing four americium samples [6]. The experimental facility consists of an aluminium ring with provisions for nine samples and with a gas hole in the centre. The facility is supposed to be irradiated in the Petten high flux reactor for about three years. The sample with radius of 2.5 mm is supposed to be made of Am_2O_3 diluted in an inert matrix of Al_2O_3 encapsulated by a titanium steel cladding. The americium composition as present after an interim storage time of five years is used (see Table 2.2). The initial density of Am_2O_3 in the sample is 0.4255 g cm^{-3}.

The results given in this paper have been obtained with the SCALE code system [7] in combination with ROLAIDS-CPM [8] to treat radial burnup effects [9]. The one-dimensional model of the experimental facility has been verified by one- and two-dimensional WIMS-6 [10] calculations. The results of the one-dimensional calculations with both the SCALE and the WIMS-6 code systems, agree very well with the two-dimensional WIMS-6 calculations [6]. In both models the americium sample is divided in five radial zones with equal volumes

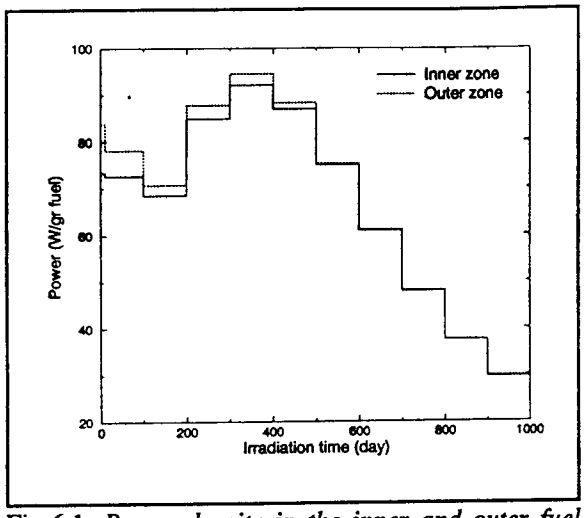

Fig 6.1: *Power density in the inner and outer fuel zones as function of irradiation time.*

to study radial burnup effects. All data used are based on JEF2.2 in 172 groups.

Results

The differences in burnup between the five zones of the samples are very small, because the differences in power history between the five zones are very small. This is seen in Figure 6.1, where the fission power in the americium sample for the inner and outer zones are given as a function of irradiation time. Because the fission power distribution over the sample is rather flat, it is expected that the radiation damage effects in the matrix due to fission product recoil and buildup will also be homogeneously distributed over the sample. The maximum power is reached after about one year of irradiation due to buildup of fissile Pu-239. The initial power is caused by neutron capture in Am-241 and fissioning of Am-242m. After irradiation of 1,000 days the actinide density in the sample has decreased from 0.3868 g cm^{-3} to 0.1096 g cm^{-3}, of which 0.0615 g cm^{-3} is Cm-244.

The radiotoxicity of the samples is given in Figure 6.2. It is seen that the radiotoxicity is considerably reduced compared to the radiotoxicity of the unirradiated samples. The remaining radiotoxicity is due to Cm-244 mainly, which is formed by neutron capture in Am-243, and which decays with half life of 18 years to Pu-240. The transmutation efficiency is therefore expected to decrease when the initial Am-243 content in the americium sample increases.

7 Conclusions

The radiotoxicity due to spent fuel is mainly determined by plutonium. After a time period of 100,000 years, also uranium gives an important contribution to the radiotoxicity. However, if uranium and plutonium are partitioned from spent fuel in order to be recycled again, and when the radiotoxicity of plutonium is considerably reduced, then the radiotoxicity due to HLW up to 50,000 years is mainly determined by americium.

Partitioning and transmutation of americium has two possible benefits: reduction of the possible dose to the population due to leakage of Np-237 from underground disposal sites, and reduction of the total inventory (radiotoxicity) of underground disposal sites (this can be either reduction of the inventory per disposal site or reduction of the total number of disposal sites).

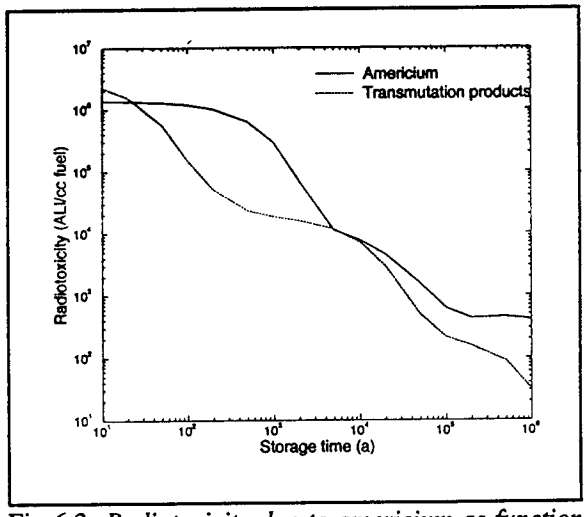

Fig 6.2: *Radiotoxicity due to americium as function of storage time after irradiation in the Petten High Flux Reactor for 1000 days.*

The initial amount of Am-241 in spent fuel contributes for about 50% to the total radiotoxicity of the daughter product Np-237 after 2 10^5 years of storage. This percentage increases when the interim storage time of spent fuel increases. Because all Am-241 can be fissioned or transmuted to other nuclides, transmutation of americium leads to a 50% reduction of the total radiotoxicity due to Np-237, which leads to an equally large reduction of the possible dose to the population due to leakage of Np-237. It is clear however that also Np-237 should be transmuted if the radiotoxicity due to Np-237 after one million years of storage is to be reduced considerably.

The long-term radiotoxicity due to americium can be reduced with a factor of 20 after about one hundred years of storage, when it has been irradiated with a thermal neutron fluence of about 2 10^{22} cm^{-2}. Irradiation of americium in a thermal neutron flux of 10^{14} $cm^{-2}s^{-1}$ (a characteristic value for the Petten High Flux Reactor), leads

to the above-mentioned reduction factor for the radiotoxicity. This was confirmed by calculations on an experimental facility containing an americium sample, which was supposed to be irradiated in the Petten HFR for about three years. Generally it can be concluded that reduction of the radiotoxicity due to americium in thermal reactors is feasible when long irradiation times are applied. This would only be meaningful, however, when the radiotoxicity due to plutonium would also be reduced considerably.

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TRANSMUTATION OF TC-99 IN FISSION REACTORS

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Abstract
Transmutation of Tc-99 in three different types of fission reactors is considered: a heavy water reactor, a fast reactor and a light water reactor. For the first type a CANDU reactor was chosen, for the second one the Superphénix reactor, and for the third one a PWR. The three most promising Tc-99 transmuters are the fast reactor with a moderated subassembly in the inner core, a fast reactor with a non-moderated subassembly in the inner core, and a heavy water reactor with Tc-99 target pins in the moderator between the fuel bundles. Transmutation half lives of 15 to 25 years can be achieved, with yearly transmuted Tc-99 masses of about 100 kg at a thermal reactor power of about 3000 MW.

1 Introduction

The radiotoxicity of fission products after a storage time of several thousands of years is mainly determined by only three isotopes: I-129, Tc-99 and Cs-135. Although the radiotoxicity of fission products is not significant compared with the radiotoxicity of actinides in spent fuel, the importance of fission products becomes apparent when the solubility of fission products is taken into account (sometimes called 'mobility'). Studies on storage of vitrified waste in salt layers [1] show that dose rates after about one to two millions of years are dominated by the neptunium decay chain and by fission products (Cs-135 and I-129). Studies on storage of spent fuel and vitrified waste in granite [2] also show that the dominating nuclides are Tc-99 and the neptunium decay chain.

This paper deals only with transmutation of Tc-99, which may be accomplished by a single neutron capture. Because no neutrons are produced in the transmutation process, the introduction of Tc-99 in a reactor will always lead to a reactivity decrease, which can be compensated by an increased fuel enrichment. The cross section of Tc-99 is rather low (see Figure 1.1), both in the thermal and in the fast energy range. This means that a high thermal or fast neutron flux is needed to achieve acceptably low transmutation half lives (see chapter 2). This report ranks fission reactors with respect to their Tc-99 transmutation capability.

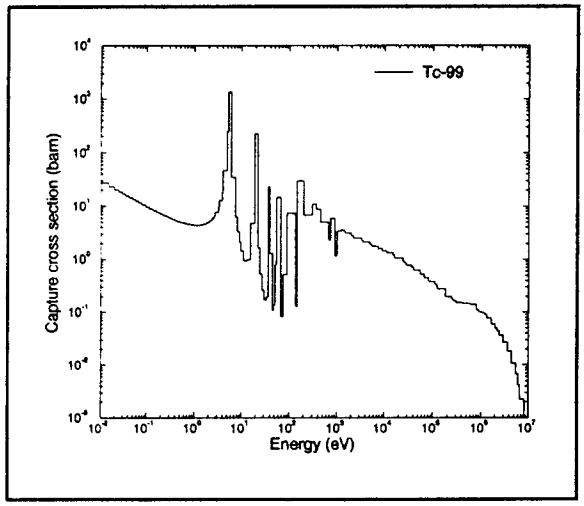

Fig 1.1: *Capture cross section of Tc-99 as function of energy.*

2 Transmutation in Heavy Water Reactors

Introduction

The transmutation rate of Tc-99 depends on both the neutron spectrum and the flux level. A reactor with a relatively high flux and a soft neutron spectrum could be a very good Tc-99 transmuter, as softening the neutron spectrum by a moderator will usually lead to higher spectrum-averaged cross sections. A high thermal neutron flux and a soft neutron spectrum are typical for a Heavy Water Reactor (HWR) like CANDU. The use of deuterium as moderator instead of hydrogen leads to very low neutron absorption in the moderator, while the use of natural uranium as fuel leads to low thermal absorption cross sections in the fuel and as a consequence a high thermal neutron flux at achievable power rating. Although the introduction of Tc-99 in the core of a HWR will most probably change these characteristics, it seems worthwile to investigate the possibilities of a HWR as Tc-99 transmuter.

Model

Data mentioned in this chapter have been taken from the CANDU type HWR as present in Darlington (Can) with a power of 935 MWe. The geometry of a standard fuel bundle is shown in Figure 2.1. Along the line from the centre fuel pin (the "first" ring of fuel pins) to the moderator, the second, third and fourth ring of fuel pins are shown with the pressure tube, the gas annulus, the calandria tube and the moderator. The gas annulus between the pressure tube and the calandria tube is filled with gas to limit heat transfer from the fuel bundle to the moderator. All fuel bundles for a given reactor design are equal.

Four cases were considered, all with equal amount of Tc-99 per fuel bundle. Case HWR-A corresponds with the centre fuel pin of each fuel bundle exchanged for a Tc-99 pin. This is shown in Figure 2.2, where the black pin in the centre stands for the Tc-99 pin. Case HWR-B corresponds with nine fuel pins in the outer ring of a fuel bundle exchanged for Tc-99 pins. This fuel bundle is then surrounded by eight fuel bundles without Tc-99. This is shown in Figure 2.3. Case HWR-C corresponds with Tc-99 pins positioned in the moderator between the fuel bundles. This is shown in Figure 2.4. Case HWR-D corresponds with Tc-99 homogeneously dissolved in the moderator between the fuel bundles. The four cases are summarized in Table 2.1.

Table 2.1: *Transmutation cases for the HWR. All cases contain an equal amount of Tc-99.*

Case	Description
HWR-A	Tc-99 target pin in the centre of each fuel bundle.
HWR-B	Nine Tc-99 target pins in the outer fuel ring of one fuel bundle surrounded by eight standard fuel bundles.
HWR-C	Tc-99 target pins in the moderator between fuel bundles.
HWR-D	Tc-99 homogeneously dissolved in the moderator.

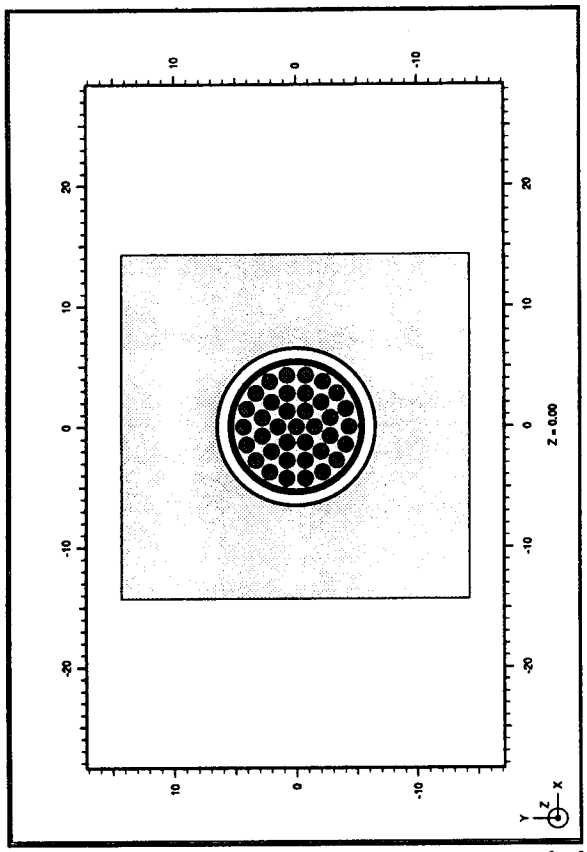

Fig 2.1: *Geometry of a standard CANDU fuel bundle.*

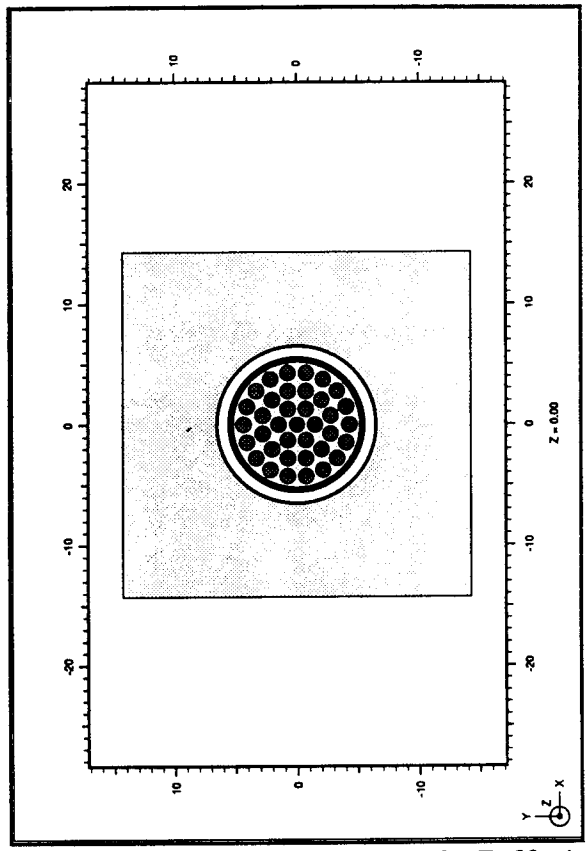

Fig 2.2: *Geometry of case HWR-A. The Tc-99 pin is located at the centre of the fuel bundle.*

All calculations were done with the KENO Monte Carlo code [3]. The transmutation half life was calculated from:

$$T_{1/2} = \frac{\ln 2}{\sigma \phi} \qquad (1)$$

where $T_{1/2}$ is the transmutation half life, σ is the one-group capture cross section of Tc-99 and ϕ is the neutron flux. All data libraries used were based on the JEF2.2 library.

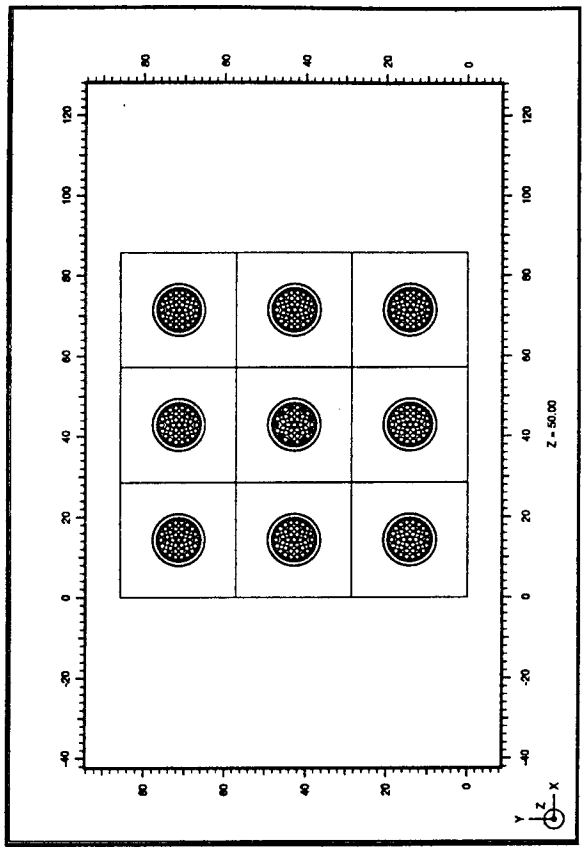

Fig 2.3: *Geometry of case HWR-B. Nine Tc-99 pins are located in the outer ring of the centre fuel bundle.*

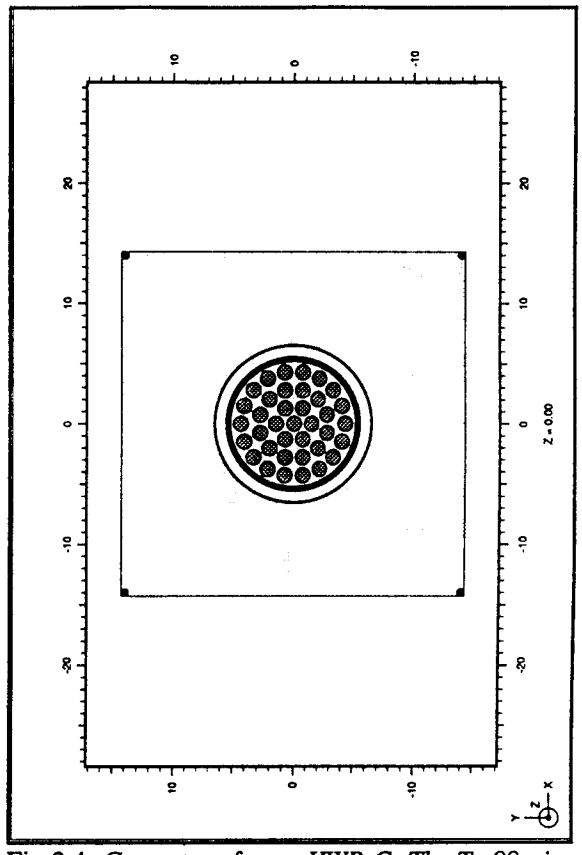

Fig 2.4: *Geometry of case HWR-C. The Tc-99 pins are located in the moderator between the fuel bundles (in the corners of the figure).*

Results

Results of the calculations are given in Table 2.2. It is seen that the transmutation half lives range from about 44 years for case HWR-A to 11 years for case HWR-D. This last case is however quite unrealistic, because it is assumed here that only the moderator between the fuel channels contains Tc-99. In reality the moderator volume is much larger, which implies that a large part of the Tc-99 is contained outside the core region. The neutron flux outside the core is very low. When the moderator regions inside and outside the core cannot be separated, the results of this case will be much worse. The loaded Tc-99 mass in all cases equals about 4.1 tonnes, which is quite high.

Table 2.2: *Results for transmutation of Tc-99 in HWRs. The Tc-99 loading in all cases is 4.1 tonnes. The HWR reference design uses natural uranium as fuel.*

Case	Enrichment (^{w}%)	Neutron flux ($cm^{-2}s^{-1}$)	Tc-99 cross section (barn)	Transm rate (kg/a)	Half life (a)
HWR-A	0.94	$1.51\ 10^{14}$	3.29	65	44.2
HWR-B	0.95	$9.07\ 10^{13}$	6.01	70	40.3
HWR-C	1.30	$9.07\ 10^{13}$	9.89	115	24.5
HWR-D	3.20	$1.36\ 10^{14}$	14.6	252	11.0

Conclusions for the HWR

The results of this chapter show that transmutation of Tc-99 in HWRs is feasible. When one fuel pin per fuel bundle is exchanged for a Tc-99 pin (cases HWR-A and HWR-B), a gross yearly transmutation rate of about 65 kg is obtained, which is the yearly production of about three 1000 MWe LWRs. More promising is case HWR-C, where Tc-99 pins are put in the moderator between the fuel bundles. This seems practically achievable. The yearly Tc-99 production in the HWR itself equals about 25 kg. This amount is not yet subtracted.

3 Transmutation in Fast Reactors

Introduction

Transmutation of Tc-99 may be accomplished in a Fast Reactor (FR) in three ways. In a special moderated subassembly (a subassembly with pins containing moderator) loaded at the periphery of the core, in a special moderated subassembly loaded in the inner core, and in a special non-moderated subassembly loaded in the inner core. The first option has been described in reference 4. Transmutation of Tc-99 in the periphery of a fast reactor core with thermal power of 2600 MW leads to transmutation half lives of 40 to 50 years with yearly transmuted Tc-99 masses of 60 to 70 kg. The Tc-99 loading in such cases equals about 4.7 tonnes of Tc-99. Reducing the Tc-99 inventory to about 500 kg leads to lower transmutation half lives of about 15 years due to reduced self shielding. The yearly transmuted Tc-99 mass is reduced then to about 20 to 25 kg, which is only just enough to compensate for the reactors own Tc-99 production. Generally transmutation efficiencies of this scheme are only moderate.

The second method for transmutation of Tc-99 in fast reactors has been considered in this chapter because of its promises of shorter half lives and larger amounts of Tc-99 yearly transmuted. Values for the transmutation half life of 17 years with yearly transmuted Tc-99 mass of 96 kg are quoted in reference 5. The thermal neutron flux in a moderated subassembly in the inner core of a fast reactor is expected to be quite high, of the order of 10^{14} to 10^{15} $cm^{-2}s^{-1}$. Moderation cannot be accomplished by water because of the presence of sodium in the core, therefore CaH_2 with density of 1.5 g cm^{-3} has been used. Besides the transmutation rate of Tc-99, the needed fuel enrichment and the power peaking in nearby fuel pins caused by moderation in the special subassembly are important.

Although fast neutron cross sections of Tc-99 are relatively low, transmutation of Tc-99 in a fast reactor without moderation could be advantageous because of its very high fast neutron flux and beause of the lack of power peaking and other side effects. This option has also been considered in this chapter.

Model

The Superhénix reactor with a thermal power of 3000 MW was used as a base design for the calculations. A geometric model was built consisting of fuel pins, Tc-99 pins and moderator pins (pins containing CaH_2), all with same diameter. The Tc-99 loading is characterized by the ratio of the number of fuel pins and Tc-99 pins. The void in the fuel pin was smeared with the fuel; structural materials were smeared with the coolant.

All calculations were done with the Monte Carlo code KENO [3] with data libraries based on JEF2.2. Because no burnup calculations can be done with the Monte Carlo code KENO, the transmutation half life was calculated according to equation 1.

Results for a moderated subassembly in the inner core

The Tc-99 transmutation half life was calculated for several cases with varying ratio of Tc-99 and moderator, accomplished by replacing moderator pins in the moderated subassembly by Tc-99 pins. The number of Tc-99 and moderator pins always summed to 271, which is the number of pins per assembly. The number of fuel pins for all cases in this paragraph was equal to 1098, which corresponds to slightly more than four fuel assemblies. Because the inner core of the Superphénix reactor contains 193 assemblies, which equals 52303 fuel pins, the modelled section represents 2.62% of the inner core, or 1.39% of the inner and outer core.

Case FR-A corresponds to the case where the outer three layers of pins in the moderated subassembly are occupied with Tc-99 pins, and all inner layers in the moderated subassembly are occupied with moderator pins. This configuration is shown in Figure 3.1. Cases FR-B to FR-H correspond to one more layer of Tc-99 pins each case. This means that for case FR-H all moderator pins are replaced by Tc-99 pins. This configuration is shown in Figure 3.2. Results for all these cases are given in Table 3.1. The enrichment shown in the table is that enrichment needed to get the k_∞ of standard fuel at BOC (about 1.17). The flux is normalised such that the average specific power is 114 W g^{-1} fuel. It should be noted that the fuel enrichment for cases FR-A to FR-F exceed 30^w%, which is considered as an upper limit. This means that cases FR-A to FR-F are most probably not practically achievable and that the number of moderated subassemblies in the core has to be decreased to get a lower Tc-99 loading and a lower fuel enrichment requirement.

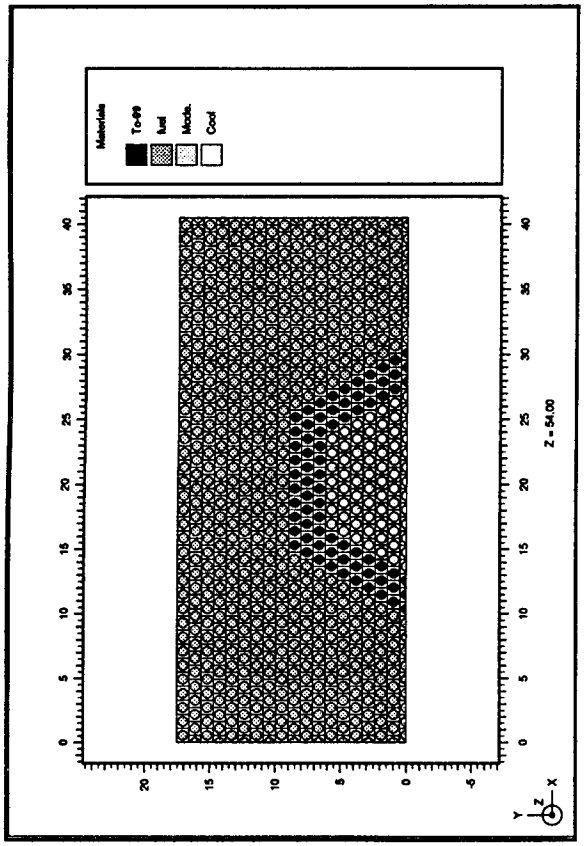

Fig 3.1: *Geometry for case FR-A. The fuel pins are located outside, the moderator pins inside the special subassembly.*

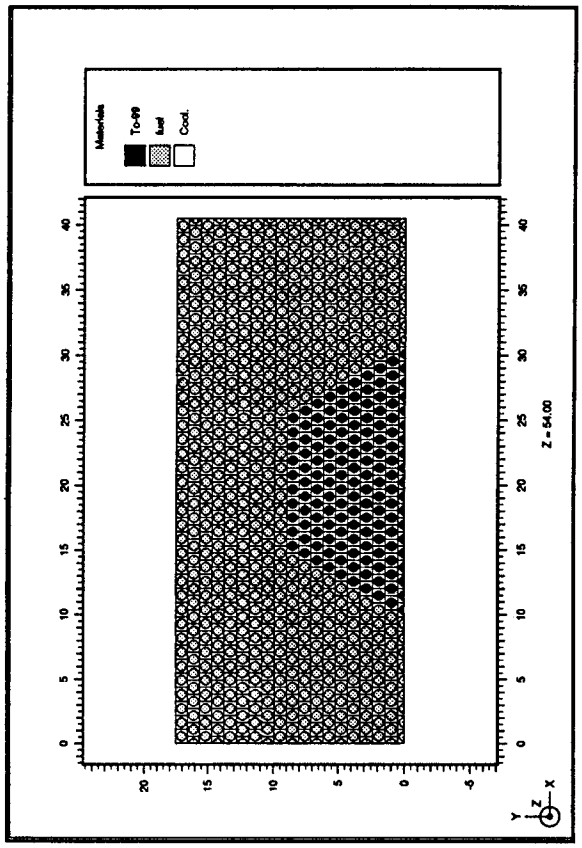

Fig 3.2: *Configuration for case FR-H. In this case no moderator pins are present.*

Due to the moderator in the special subassembly, the power in the fuel pins adjacent to the special subassembly increases with 30% for case FR-A, 12% for case FR-B and only 1% for case FR-C. With increasing number of Tc-99 pins in the special moderated subassembly, the influence of the moderator diminishes, and the influence of Tc-99 acting as an absorber increases, leading to a power decrease in the fuel pins adjacent to the special subassembly. This power increase and decrease could be compensated for by a variable plutonium enrichment around the special subassembly. This is not further investigated.
In Table 3.1, the Tc-99 inventories for cases FR-A to FR-H are given, assuming that the inner core is loaded according to the configuration of the case considered. Also the yearly transmuted Tc-99 masses are given. An interesting phenomenon can be noticed from this table. For cases FR-A to FR-D the yearly transmuted Tc-99 mass increases due to the increase of the Tc-99 inventory. However, the increase from case FR-C to FR-D is very small due to the decreasing number of moderator pins leading to much less moderation. This leads to lower Tc-99 cross sections. The transmuted mass for case FR-F is even lower than for case FR-D due to decreased moderation. For cases FR-G and FR-H the yearly transmuted masses do not change significantly anymore because of the very small increase of the Tc-99 inventory.

Results for a non-moderated subassembly in the inner core
Case FR-H of the previous paragraph contains only Tc-99 pins. The inventory in such case is very high, especially if that is compared with the yearly production of one 1000 MWe LWR, which is only about 20 kg.

It would make sense to reduce the Tc-99 inventory by replacing Tc-99 pins by fuel pins. Four cases with different amounts of Tc-99 loading are investigated. Case FR-I with the tenth layer of the special subassembly loaded with Tc-99 pins, case FR-J with the eighth layer of the special subassembly loaded with Tc-99 pins, case FR-K with the sixth layer of the special subassembly loaded with Tc-99 pins and case FR-L with the fifth layer of the special subassembly loaded with Tc-99 pins. All other positions in the special subassembly are filled with fuel pins in these cases. The geometry for cases FR-I and FR-L are shown in Figures 3.3 and 3.4, respectively.
Results of the calculations are given in Table 3.2. It is seen that the transmutation half lives for these cases are reasonably low, between 15 and 18 years. The yearly transmuted Tc-99 mass reaches about 100 kg, which is the yearly production of about five 1000 MWe LWRs.

Table 3.1: *Results for cases FR-A to FR-H. The enrichment of standard fuel is 19.5 ^{w}%.*

Case	Number of Tc-99 pins[a)]	Enrichment (^{w}%)	Power factor[b)]	Neutron flux ($cm^{-2}s^{-1}$)
FR-A	144	35.4	1.30	2.04E15
FR-B	180	34.1	1.12	2.15E15
FR-C	210	33.1	1.01	2.25E15
FR-D	234	32.1	0.95	2.36E15
FR-E[c)]	252			
FR-F	264	30.7	0.88	2.58E15
FR-G	270	29.8	0.87	2.69E15
FR-H	271	29.4	0.87	2.72E15
Case	Inventory[d)] Tc-99 (kg)	Transm rate Tc-99 (kg/a)	Tc-99 cross section (barn)	Half life (a)
FR-A	2743	122	0.706	15.3
FR-B	3429	137	0.602	17.0
FR-C	4000	144	0.516	18.9
FR-D	4458	145	0.444	21.0
FR-E[c)]	4800			
FR-F	5029	132	0.326	26.1
FR-G	5143	132	0.306	26.7
FR-H	5162	132	0.301	26.9

a) Number of Tc-99 pins per special subassembly.
b) Power in fuel pins adjacent to special subassembly relative to average power.
c) Not calculated.
d) Inventory when the inner core of 193 assemblies is loaded according to the configuration of the case considered.

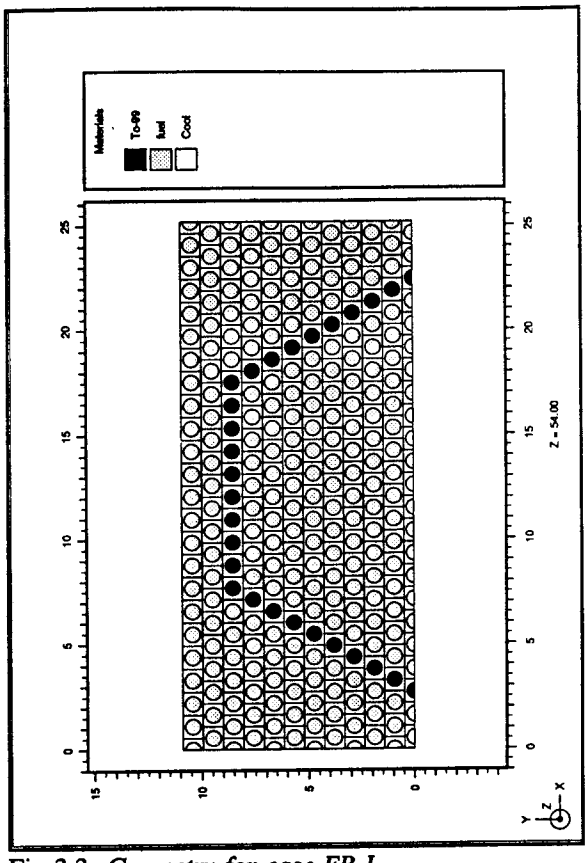

Fig 3.3: *Geometry for case FR-I.*

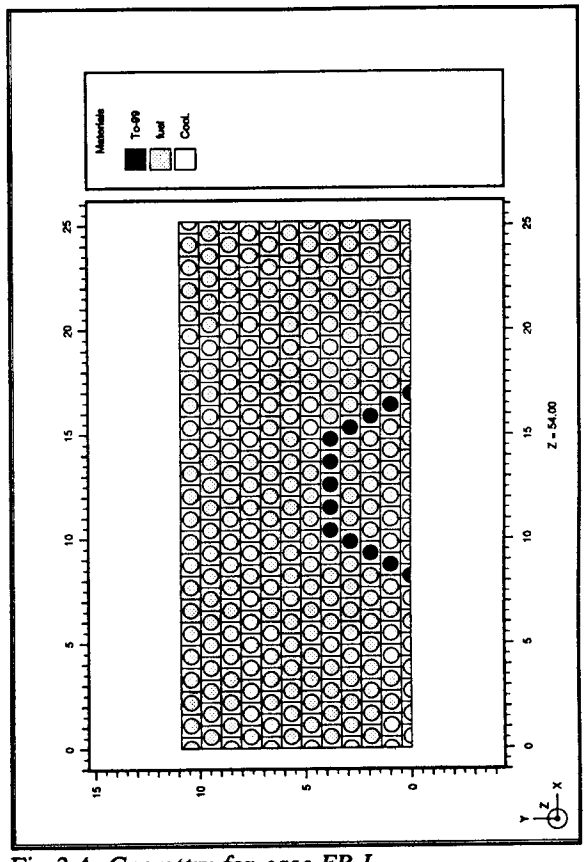

Fig 3.4: *Geometry for case FR-L.*

Table 3.2: *Results for cases FR-I to FR-L. The enrichment of standard fuel is 19.5 w%. For notes see Table 3.1.*

Case	Number of Tc-99 pins[a)]	Enrichment (w%)	Neutron flux ($cm^{-2}s^{-1}$)	
FR-I	54	25.7	3.71E15	
FR-J	42	24.3	3.94E15	
FR-K	30	22.8	4.04E15	
FR-L	24	22.0	4.13E15	
Case	**Inventory[d)] Tc-99 (kg)**	**Transm rate Tc-99 (kg/a)**	**Tc-99 cross section (barn)**	**Half life (a)**
FR-I	2662	101	0.331	17.9
FR-J	2071	86	0.340	16.4
FR-K	1479	64	0.345	15.8
FR-L	1183	53	0.350	15.2

Conclusions for the FR

Results on transmutation of Tc-99 in the inner core of a fast reactor are given in this chapter. Both transmutation in a moderated subassembly and in a non-moderated subassembly were considered.

Transmutation in a moderated subassembly has the largest influence on the power distribution in adjacent fuel pins, leading to a necessarily large Tc-99 layer between the moderator pins and adjacent fuel pins, or to a variable plutonium enrichment decreasing towards the periphery of the special subassembly. Most probably this will limit the practical applicability of moderated subassemblies in the inner core of a fast reactor. Nevertheless, when this power peaking is addressed at, the application of a moderated subassembly in the inner core of a fast reactor leads to low transmutation half lives with high yearly transmuted Tc-99 masses.

Transmutation of Tc-99 in a fast reactor seems also possible without moderation. Although the Tc-99 cross section in a non-moderated subassembly is about a factor of two lower than in a moderated one, the neutron flux may be higher, possibly leading to equal transmutation rates. This was actually confirmed by these calculations. Transmutation of Tc-99 in a non-moderated subassembly in the inner core of Superphénix can lead to a gross yearly transmuted Tc-99 mass of about 100 kg with a Tc-99 inventory of 2.7 tonnes. The yearly Tc-99 production in Superphénix equals about 20 kg. This amount is not yet subtracted.

4 Transmutation in Light Water Reactors

Introduction

Light Water Reactors (LWRs) are abundantly present in Western Europe and the US. Transmutation of Tc-99 in LWRs could have the advantage that no special Tc-99 burners are necessary and that each LWR consumes its own produced Tc-99. Then an equilibrium state is achieved and the net production of Tc-99 is zero.

Whether such situation can be achieved or not depends of course on the transmutation rate of Tc-99 in LWRs and on the inventory necessary to transmute the yearly production of one LWR. When this inventory becomes very high it must be concluded that equilibrium with respect to Tc-99 production cannot be achieved practically. The work presented in this chapter is performed in cooperation with Belgo-Nucléaire (see also reference 6).

Model

The modelled PWR corresponds with a Westinghouse type of PWR with a power of 900 MWe.

Calculations on transmutation of Tc-99 were done for both PWRs loaded with UO_2 fuel only (cases LWR-A to LWR-D, see Figure 4.1), and for PWRs loaded with UO_2 fuel for three quarters of the core and with MOX fuel for one quarter of the core (cases LWR-E to LWR-H, see Figure 4.2). In both cases Tc-99 pins with same diameter as fuel pins were modelled in the guide tubes of the core. In the first case the Tc-99 pins were modelled in all guide tubes of the core, in the latter case the Tc-99 pins were modelled in the guide tubes of the MOX fuel only. In both cases the enrichment of the UO_2 fuel was increased to achieve the same average k_∞ as for the corresponding core (full UO_2 or one quarter MOX fuel) without Tc-99 pins. This average k_∞ was obtained by the WIMS package and the accompanied 69-group cross section data library. The enrichment at BOC determined by the WIMS package was then used in KENO Monte Carlo calculations [3] with the 172-group cross section data library based on JEF2.2 to calculate the neutron flux averaged over the Tc-99 target pins in the guide tubes. Resonance shielding of the Tc-99 target pins could then properly be accounted for. Therefore, and also because the WIMS 69-group cross section library is not based on JEF2.2 but on an older evaluated file, the calculations have been performed as described above. The transmutation half lives were calculated according to equation 1.

Results for UO_2 fuel

These cases correspond with a PWR fully filled with UO_2 fuel, where Tc-99 pins with same diameter as fuel pins are inserted in the guide tubes. The different cases correspond with different Tc-99 densities in the target pins of 1, 2, 5 and 10.5 g cm^{-3}. It is assumed that the remaining space in the guide tubes is filled with some inert matrix with zero cross section. The geometry of these cases is given in Figure 4.1. Results are given in Table 4.1. It is seen that the cross section decreases with increasing Tc-99 density. The lowest transmutation half life is obtained for lowest Tc-99 density, and it is seen that in case LWR-A the yearly transmuted Tc-99 mass is still larger than the yearly production of Tc-99, which equals about 18 kg for a 900 MWe PWR.

Table 4.1: *Results for cases LWR-A to LWR-D (full UO_2 core). The enrichment of standard fuel is 3.7^w%.*

Case	Density Tc-99 (g cm^{-3})	Enrichment U-235 (%)	Neutron flux (cm^{-2}s^{-1})	
LWR-A	1.0	4.5	2.68E14	
LWR-B	2.0	5.1	2.61E14	
LWR-C	5.0	6.5	2.47E14	
LWR-D	10.5	8.6	2.33E14	
Case	**Inventory Tc-99 (kg)**	**Transm rate Tc-99 (kg/a)**	**Tc-99 cross section (barn)**	**Half life (a)**
LWR-A	726.5	23.7	3.93	20.9
LWR-B	1453	37.2	3.15	26.8
LWR-C	3633	63.8	2.27	39.2
LWR-D	7628	91.7	1.64	57.3

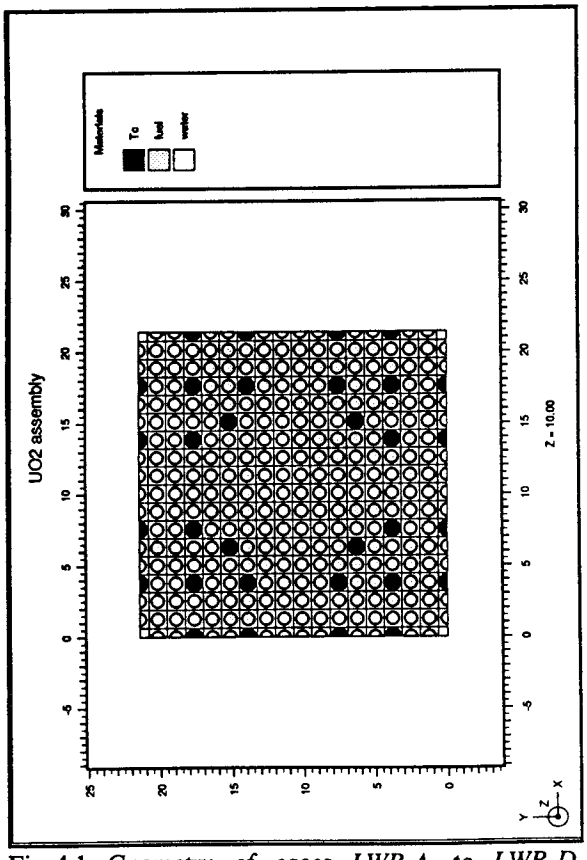

Fig 4.1: *Geometry of cases LWR-A to LWR-D. Four quarters of an assembly with UO_2 fuel are shown.*

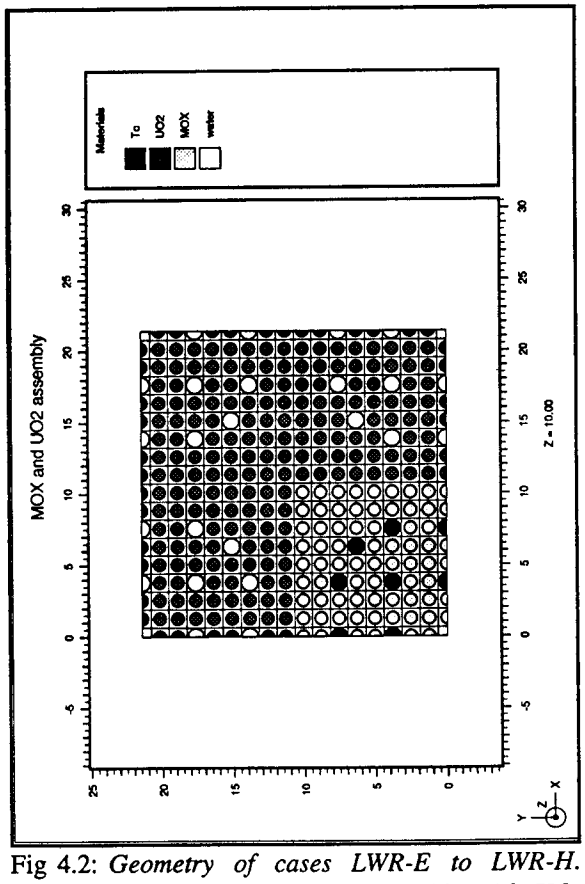

Fig 4.2: *Geometry of cases LWR-E to LWR-H. Three quarters of an assembly with UO_2 fuel and one quarter with MOX fuel are shown.*

Results for MOX fuel

These cases correspond with a PWR filled for three quarters of the core with UO_2 fuel and for one quarter of the core with MOX fuel. Tc-99 pins with same diameter as fuel pins inserted in the guide tubes of the MOX fuel. The different cases correspond again with different Tc-99 densities in the target pins of 1, 2, 5 and 10.5 g cm^{-3}. Again the remaining space in the guide tubes was assumed to be filled with some inert matrix with zero cross section. The geometry of these cases is given in Figure 4.2. Results are given in Table 4.2. It is seen that the transmutation half lives are about 50% larger than for the corresponding cases LWR-A to LWR-D. This is mainly because of the low capture cross sections of Tc-99 in a the harder neutron spectrum characterisitic for MOX fuel. Only for the case with Tc-99 density of 10.5 g cm^{-3}, the transmuted rate is just large enough to compensate for the reactors own yearly Tc-99 production (about 18 kg).

Table 4.2: *Results for cases LWR-E to LWR-H (1/4 MOX, 3/4 UO_2 core). The enrichment of standard fuel is 3.7w%.*

Case	Density Tc-99 (g cm^{-3})	Enrichment U-235 (%)	Neutron Flux ($cm^{-2}s^{-1}$)	
LWR-E	1.0	4.1	2.31E14	
LWR-F	2.0	4.3	2.26E14	
LWR-G	5.0	4.7	2.15E14	
LWR-H	10.5	5.1	2.03E14	
Case	Inventory Tc-99 (kg)	Transm rate Tc-99 (kg/a)	Tc-99 Cross Section (barn)	Half Life (a)
LWR-E	181.6	4.0	3.10	30.7
LWR-F	363.3	6.3	2.44	39.9
LWR-G	908.2	11.1	1.82	56.4
LWR-H	1907	17.0	1.40	77.4

The dependence of the yearly transmuted Tc-99 mass as function of density is clearly seen in Figure 4.3. Due to resonance and spatial self shielding, the yearly transmuted Tc-99 mass does not show a linear dependence with Tc-99 density. It could be very beneficial to transmute Tc-99 at the lowest possible density.

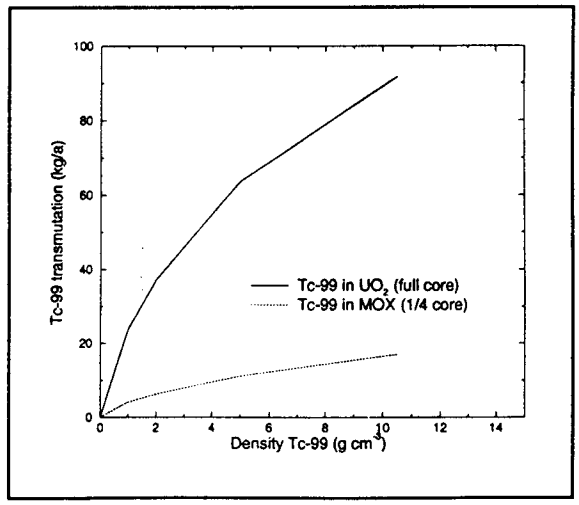

Fig 4.3: *Tc-99 transmutation rate as function of density.*

Conclusions for the LWR

Transmutation of Tc-99 in PWRs lead to rather large transmutation half lives and low yearly transmuted masses. Transmutation in a UO_2 fuelled core has preference above transmutation in MOX fuelled PWRs because of the softer neutron spectrum leading to higher Tc-99 cross sections. Transmutation should be performed at the lowest possible density. Even Tc-99 with density of 1 g cm^{-3} put in all guide tubes lead to yearly transmuted masses of about 24 kg with inventory of about 726 kg. Even for this case it is very

questionable whether breakeven (Tc-99 transmutation equal to Tc-99 production) can be achieved, because in practice not all guide tube are available for transmutation purposes.

5 General Conclusions

To give a ranking of fission reactors with respect to their Tc-99 transmutation capability, the ratio of the yearly transmuted Tc-99 mass and the transmutation half life seems quite reasonable to use as a ranking parameter. This ranking is given in Table 5.1. It must be noticed again that the first case (moderated special subassembly in the inner core of a fast reactor) needs a too high plutonium enrichment (>30^{w}%). Therefore the inventory and transmutation rate of Tc-99 given in the first row of Table 5.1 should be scaled down to reduce the enrichment need. Also a variable plutonium enrichment decreasing towards the periphery of the special subassembly is needed. Nevertheless this option is expected to yield the best performance for transmutation of Tc-99.

Table 5.1: *Ranking of reactors with respect to Tc-99 transmutation capability.*

React	Configuration	Inventory Tc-99 (kg)	Transm rate Tc-99 (kg/a)	Half Life (a)
FR	Moderated S/A in inner core	3429	137	17
FR	Non-moderated S/A in inner core	2662	101	18
HWR	Pin in moderator	4126	115	25
LWR	Pin in guide tube UO_2 fuel	3633	64	39
LWR	Pin in guide tube MOX fuel	1907	17	77

6 Acknowledgements

The authors acknowledge the Commission of the European Union for co-funding this work under contract number F12W-CT91-0104.

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A PHYSICAL ASSESSMENT OF THE EFFECTIVENESS OF DIFFERENT ACTINIDE TRANSMUTATION SYSTEMS

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ABSTRACT

A general method for investigating the effectiveness of actinide transmutation systems is proposed. The method allows to assess the impact of different long–term transmutation strategies on the actinide inventories of the systems, the composition of the actinide waste, and the radiological risk associated with the disposal of this waste. In a comparative study, the method is applied to systems with a wide range of characteristics including a PWR, a fast reactor, a high–flux superthermal system, and two accelerator–based systems with fast neutron spectra.

The results of the study emphasise the importance of a good overall neutron balance for completely burning the actinides. As to the radiological risk of the waste, it is found that most transuranic actinides can be recycled in fast reactors and PWRs with similar risks, and that high–flux superthermal systems burn actinides generally with a somewhat smaller risk than other systems. Interestingly, it appears that, in the long range, the radiological risk of the waste cannot be reduced by changing from the uranium–plutonium to the thorium–uranium fuel cycle.

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INTRODUCTION

Actinides contribute significantly to the long–term toxicity of radioactive wastes, since they are all unstable and decay through extended chains, involving long–lived α emmitters, to nuclides in the stable range. The decays can be by–passed by fissioning the actinides. For a long–term nuclear energy system, the goal must be therefore to fission the actinides with a high overall efficiency. A transmutation system can be termed to be "effective", if it contributes to achieve this goal. This means, for instance, that the fissionable nuclides, i.e. the even–neutron nuclides with a high–energy fission threshold, should not be treated as waste, but utilised more extensively for energy production.

The effectiveness of transmutation systems is usually assessed by means of detailed calculations of the mass flows for the specific concepts and fuel management schemes. This approach was adopted, for example, in the "Overview of Physics Aspects of Different Transmutation Concepts" prepared by a task force of the OECD–NEA Nuclear Science Committee [1]. While yielding all parameters of interest, the detailed analysis of a system usually requires a significant computational effort. This effort can become prohibitive, if many systems are to be compared using consistent assumptions and approximations.

For this reason and to improve the understanding of the basic phenomena, there is a need for simplified approaches which concentrate on specific aspects of the systems. Transmutation properties have been discussed, for instance, in terms of the fission–to–capture ratios of the nuclides to be burnt or the overall neutron balance of the systems. For the investigation of long–term transmutation strategies, the present paper proposes a method which allows to quantify the impact of the strategies on the actinide inventories of the systems, the composition of the actinide waste, and the radiological risk associated with the disposal of this waste. The method is used to compare the transmutation effectiveness of systems with different characteristics and different fuel cycles on a consistent basis, i.e. using a common data base.

COMPARED SYSTEMS

The comparison includes the following systems:

- A normal PWR
  The fuel cell characteristics are those of the 920 MWe plant of Gösgen, and the average neutron flux is assumed to be 10^{14} $cm^{-2}s^{-1}$.

- A MOX–fuelled fast reactor (FR)
  The composition of the fuel corresponds to that of the fresh core of Superphénix, and the average flux is assumed to be 10^{15} $cm^{-2}s^{-1}$.

- A "superthermal" system with a high flux (STS)
  A well–moderated thermal neutron spectrum, typical for the D_2O moderator of a continuous spallation neutron source, is assumed. The system simulates the base–case design of the Los Alamos ATW concept [2] with a neutron flux of 10^{16} $cm^{-2}s^{-1}$.

- An accelerator–driven "fast" molten salt system (FMS)
  This is a directly driven system with chloride molten salt fuel and a continuous reprocessing system for removing the fission products. The geometry and the composition are those of the "molten salt core system" proposed by JAERI [3], and the average neutron flux is 10^{15} $cm^{-2}s^{-1}$. The neutrons have a "harder" spectrum (i.e. a higher mean energy) than the neutrons in a MOX–fuelled fast reactor.
- An accelerator–driven "superfast" system using metal fuel (SFS)
  Here, the idea is to combine the favourable neutronics of an actinide target and a metal fuel cycle with low reprocessing losses. The system features a sodium–cooled target with actinide–zirconium fuel and full actinide recycling. The geometry conforms with that of the Phoenix concept [4], and the neutron flux is the same as that assumed for the other fast systems. Of the investigated systems, this system has the hardest neutron spectrum.

MODELLING OF THE SYSTEMS

The parameters used in this investigation are derived from a common set of spatially averaged one–group cross sections prepared as follows:

The cross sections for the PWR and the superthermal system were obtained from thermal cell calculations using the BOXER code [5] together with a cross section library derived from the JEF–1 evaluated nuclear data file.

The cross sections for the fast reactor and the hybrid systems were generated using two–dimensional cylindrical models of the systems. With the height of the cylinder fixed to represent the height of the reactor or the depth of the target, the radius was adjusted to give the appropriate beginning–of–life multiplication factor. For the subcritical cases, neutron sources were calculated using a version of the HETC code which includes high–energy fission [6]. Neutron spectrum calculations below 15 MeV are based on JEF–2.2 and the computational scheme described in Ref. 7.

The sensitivity of the parameters to the data and the modelling of the systems is discussed in Ref. 8.

FISSION–TO–CAPTURE RATIOS

In all fission–based systems, neutron–induced fission compete with capture reactions. In general, neutron captures are undesirable, since they lead to an increase rather than a reduction of the atomic mass number. It can therefore be speculated that the transmutation effectiveness of a system for a particular actinide increases with the fission–to–capture ratio of this actinide.

Table 1 shows that, for most actinides, fast neutron spectra yield a higher fission–to–capture ratio than thermal spectra. In accelerator–based fast systems, the mean neutron energy and thus the fission–to–capture ratios increase beyond the limits of normal fast reactors due to the subcriticality of the target and the effect of the evaporation neutrons which have higher energies than the fission neutrons. As expected, the fissionable nuclides show particularly strong spectrum hardening effects. There may therefore be an incentive for designing accelerator–based fast systems specifically with the aim of fissioning the fissionable nuclides.

OVERALL NEUTRON BALANCE

An important aspect is the overall neutron balance of a system. For a closed long–term system, this should be such as to allow the complete conversion of the actinides to fission products while maintaining criticality and taking account of losses due to neutrons leaking out of the system. In Ref. 9, Salvatores et al. have proposed to measure the overall neutron balance in terms of the "neutron production" parameter, –D. Unlike other neutron balance parameters, the "neutron production" depends on the ratio of neutron induced reactions (fissions, captures, n,2n reactions) to radioactive decays and therefore on the neutron flux.

Table 1: Fission–to–Capture Ratios for Actinides with $T_{1/2} > 10$ a

Nuclide	STS	PWR	FR	FMS	SFS
^{230}Th	0.00	0.00	0.20	0.43	1.87
^{232}Th	0.00	0.01	0.03	0.06	0.16
^{231}Pa	0.00	0.01	0.09	0.18	0.52
^{232}U	1.13	1.49	3.45	4.79	9.58
^{233}U	10.21	7.91	10.68	11.84	15.29
^{234}U	0.00	0.03	0.61	1.12	2.60
^{235}U	5.57	4.23	3.61	4.29	6.39
^{236}U	0.01	0.04	0.21	0.42	0.95
^{238}U	0.00	0.13	0.18	0.33	0.99
^{237}Np	0.00	0.02	0.23	0.45	1.32
^{238}Pu	0.03	0.09	2.16	3.39	7.93
^{239}Pu	2.27	1.76	3.69	5.27	11.35
^{240}Pu	0.00	0.00	0.74	1.38	3.96
^{241}Pu	2.91	2.95	4.67	4.90	5.34
^{242}Pu	0.00	0.02	0.61	1.26	3.82
^{244}Pu	0.02	0.16	1.10	2.60	12.87
^{241}Am	0.01	0.01	0.15	0.30	0.75
^{242m}Am	4.88	4.93	6.55	7.03	8.74
^{243}Am	0.00	0.01	0.14	0.29	0.81
^{243}Cm	4.99	5.88	7.42	9.97	39.48
^{244}Cm	0.04	0.06	0.84	1.51	3.83
^{245}Cm	6.58	6.87	6.24	7.79	16.54
^{246}Cm	0.07	0.22	1.27	2.69	7.62
^{247}Cm	1.47	1.56	6.40	9.01	17.26
^{248}Cm	0.05	0.12	1.40	2.74	8.30

Nuclide–dependent "neutron production" values for the different systems are shown in Table 2. For the most abundant minor actinides in LWR–discharged fuels (^{237}Np, ^{241}Am, ^{243}Am) the overall neutron balance in a PWR is negative, indicating that the chain of successive transmutations does not provide enough neutrons to support itself. In a superthermal system, the overall neutron balance improves but remains poor compared with that of a fast reactor. Superfast systems offer the possibility of enhancing the overall neutron balance beyond the limit of fast reactors. This can be desirable when the system is intended to transmute not only actinides, but also fission products.

ASYMPTOTIC ACTINIDE INVENTORIES

Nuclide inventories and fuel compositions play an important role in system studies and risk analyses: the former have an impact on the overall characteristics of the systems and the core accident risk; the latter influence the different types of risks arising from the leakage and migration of radiation and activity in the fuel cycle, including the long–term risk associated with waste repositories. From a safety viewpoint, small nuclide inventories per MW are advantageous, since they comply with the goal of a small overall risk.

Table 2: Overall Neutron Balance for Actinides with $T_{1/2} > 10$ a (–D in Ref. 9)

Nuclide	STS	PWR	FR	FMS	SFS
^{230}Th	-0.75	-0.78	-0.07	0.25	0.77
^{232}Th	-1.21	0.17	0.38	0.55	0.74
^{231}Pa	0.25	0.22	0.67	0.83	1.08
^{232}U	1.25	1.20	1.60	1.70	1.85
^{233}U	1.26	1.18	1.35	1.42	1.50
^{234}U	-0.17	-0.34	0.55	0.92	1.26
^{235}U	0.82	0.61	0.94	1.13	1.35
^{236}U	-1.62	-1.92	0.02	0.54	1.06
^{238}U	-0.14	-0.04	0.73	1.06	1.43
^{237}Np	0.20	-1.05	0.67	1.03	1.43
^{238}Pu	0.07	-0.10	1.41	1.65	1.88
^{239}Pu	1.01	0.72	1.53	1.74	1.93
^{240}Pu	0.04	-0.30	1.00	1.41	1.82
^{241}Pu	1.04	0.70	1.26	1.50	1.79
^{242}Pu	-0.56	-1.16	0.60	1.27	1.83
^{244}Pu	1.38	1.55	1.94	2.14	2.26
^{241}Am	-0.43	-0.94	0.68	1.13	1.68
^{242m}Am	1.73	1.63	1.89	2.00	2.15
^{243}Am	0.39	-0.22	0.71	1.19	1.75
^{243}Cm	2.06	1.90	2.12	2.23	2.38
^{244}Cm	1.39	0.76	1.47	1.80	2.13
^{245}Cm	2.36	2.43	2.63	2.76	2.95
^{246}Cm	0.33	0.75	2.23	2.58	2.79
^{247}Cm	1.18	1.31	2.41	2.59	2.74
^{248}Cm	0.11	0.31	1.68	2.18	2.64

Considering that the ultimate goal is to incorporate transmutation into closed long–term nuclear energy systems, it is useful to calculate "asymptotic actinide inventories", i.e. actinide inventories resulting from the continuous burning of individual nuclides in the systems of interest assuming that the nuclides are exposed to a fixed neutron spectrum and flux and burnt until all nuclear reactions are in equilibrium. The asymptotic inventory for burning a mixture of nuclides can be obtained easily by linearly combining the nuclide–specific inventories.

A direct method has been used to efficiently calculate such inventories. In brief, the asymptotic inventory of nuclide i resulting from burning nuclide j, $N_{i,j}$, is obtained from the equation

$$\sum_k P_{i,j,k} = \lambda_{i,eff} N_{i,j} \ ,$$

where P and λ are the production rate and the effective half–life for the given neutron spectrum and flux. The production rate is summed over the generations, k, of the chain of transmutations until the initial nuclei are fissioned (a small fraction of the initial nuclei is "lost", i.e. transmuted to heavy nuclei with atomic numbers outside the range of the calculation, $90 < Z < 96$, but for the important actinides this fraction is negligible). The method is much faster than a normal burnup calculation, allowing asymtotic inventories to be calculated accurately for a wide range of parameters.

For nuclides with $T_{1/2} > 10$ a, asymptotic inventories resulting from burning ^{237}Np and ^{239}Pu are given in Tables 3 and 4. The k_{inf}[1] values in the tables indicate to what extent the asymptotic actinide mixtures can themselves maintain criticality. Obviously, burning ^{237}Np in a PWR does not produce a critical inventory. In this case, fissile material has to be added to the mixture whereby the effective inventory is increased, or the system has to be driven using externally generated neutrons.

Comparing the PWR with the fast reactor, it can be seen that, for the more abundant actinides, the asymptotic inventories of the latter are higher. However, the fast reactor has lower inventories of the higher plutonium isotopes, ^{243}Am, and the curium isotopes.

Interestingly, it appears that increasing the mean neutron energy beyond that of a fast reactor further enhances the inventory of the nuclide being burnt. On the other hand, hardening the neutron spectrum has the expected effect of reducing americium and curium inventories as well as ^{238}Pu and ^{240}Pu build–up from ^{237}Np and ^{239}Pu, respectively.

Table 3: Asymptotic Inventories Resulting from Burning ^{237}Np
(No. of atoms normalised to 1 fission/s)

Nuclide	STS	PWR	FR	FMS	SFS
^{230}Th	1.17E-05	1.80E+03	5.24E+04	5.06E+04	3.95E+04
^{232}Th	7.22E-07	6.34E+01	5.42E+01	4.87E+01	3.11E+01
^{231}Pa	2.64E-06	8.42E+02	2.81E+03	3.49E+03	3.19E+03
^{232}U	2.97E-03	4.01E+03	2.90E+04	2.38E+05	3.22E+05
^{233}U	1.73E-03	1.87E+03	2.06E+04	1.32E+05	1.03E+05
^{234}U	4.21E+01	4.21E+07	1.17E+08	1.03E+08	5.84E+07
^{235}U	8.51E+00	2.04E+07	2.82E+07	2.24E+07	1.05E+07
^{236}U	4.34E+01	1.89E+07	2.30E+07	1.64E+07	7.13E+06
^{238}U	3.79E+01	4.43E+05	1.63E+04	5.81E+03	9.30E+02
^{237}Np	9.29E+05	3.30E+08	5.47E+08	6.59E+08	8.04E+08
^{238}Pu	1.08E+05	3.52E+08	4.35E+08	3.72E+08	2.27E+08
^{239}Pu	5.13E+04	6.03E+07	9.95E+07	6.63E+07	2.30E+07
^{240}Pu	2.74E+04	1.65E+07	5.29E+07	2.39E+07	3.55E+06
^{241}Pu	1.40E+04	2.45E+07	6.29E+06	2.23E+06	1.87E+05
^{242}Pu	6.95E+04	2.99E+07	6.16E+06	1.87E+06	1.12E+05
^{244}Pu	8.40E+03	8.10E+03	6.03E+01	7.86E+00	9.27E-02
^{241}Am	5.68E+00	3.70E+06	4.35E+06	1.83E+06	1.88E+05
^{242m}Am	7.63E-02	9.56E+04	3.50E+05	1.28E+05	9.98E+03
^{243}Am	3.02E+04	1.78E+07	1.61E+06	4.24E+05	1.74E+04
^{243}Cm	6.30E-01	2.18E+04	1.17E+04	3.19E+03	8.93E+01
^{244}Cm	1.04E+05	2.76E+07	1.21E+06	2.23E+05	4.68E+03
^{245}Cm	2.18E+03	3.78E+06	1.24E+05	2.58E+04	5.29E+02
^{246}Cm	9.14E+04	1.67E+07	1.71E+05	1.72E+04	8.30E+01
^{247}Cm	2.88E+03	1.18E+06	1.75E+04	1.25E+03	3.33E+00
^{248}Cm	1.54E+04	2.64E+06	9.42E+03	4.22E+02	4.91E-01
k_{inf}	1.077	0.735	1.299	1.535	1.958

[1] Neutron multiplication factor of an infinite system

When applying these results to practical systems, it has to be taken into account that the asymptotic inventories depend on the neutron flux. Typical fluxes in PWRs and fast systems are 2 to 3 times higher than those assumed in the present comparison. On the other hand, the fuel cycle is not explicitly modelled in the calculations. Assuming a batch–type processing, the fluxes were therefore appropriately scaled to simulate the out–of–pile time of the fuel.

Compared with the PWR, the superthermal system has lower inventories than one would expect from the flux ratio alone. This is due to the well–moderated neutron spectrum and the fact that short–lived capture products with a high fission cross section are fissioned before they decay. The spectrum effect is relatively important and reduces the inventories of the nuclides being burnt by additional factors of 3.6 and 4.5 for ^{237}Np and ^{239}Pu, respectively. In the case of ^{237}Np, the ^{238}Np fission effect mitigates considerably the build–up of ^{238}Pu (cf. ^{238}Pu / ^{237}Np ratios in Table 3).

Table 4: Asymptotic Inventories Resulting from Burning ^{239}Pu
(No. of atoms normalised to 1 fission/s)

Nuclide	STS	PWR	FR	FMS	SFS
^{230}Th	1.77E-10	5.45E+01	1.65E+03	1.10E+03	4.21E+02
^{232}Th	4.25E-08	2.22E+00	4.80E+00	4.30E+00	3.08E+00
^{231}Pa	4.12E-11	2.55E+01	8.82E+01	5.57E+01	1.19E+01
^{232}U	3.71E-10	4.39E+01	1.35E+02	3.44E+02	1.41E+02
^{233}U	7.23E-10	2.18E+01	4.28E+02	1.98E+03	7.00E+02
^{234}U	6.38E-04	1.28E+06	3.70E+06	2.24E+06	6.26E+05
^{235}U	4.65E-02	6.35E+05	1.06E+06	7.17E+05	4.49E+05
^{236}U	2.56E+00	6.62E+05	2.04E+06	1.45E+06	7.06E+05
^{238}U	2.37E+00	3.69E+04	6.24E+03	2.98E+03	7.17E+02
^{237}Np	9.41E-02	2.53E+05	1.12E+06	8.41E+05	3.38E+05
^{238}Pu	1.64E+00	1.07E+07	1.37E+07	8.11E+06	2.42E+06
^{239}Pu	1.62E+05	7.25E+07	4.39E+08	4.88E+08	5.51E+08
^{240}Pu	8.18E+04	1.98E+07	2.33E+08	1.76E+08	8.50E+07
^{241}Pu	4.18E+04	2.95E+07	2.77E+07	1.64E+07	4.47E+06
^{242}Pu	2.08E+05	3.60E+07	2.72E+07	1.37E+07	2.69E+06
^{244}Pu	2.51E+04	9.75E+03	2.66E+02	5.78E+01	2.22E+00
^{241}Am	1.70E+01	4.45E+06	1.92E+07	1.34E+07	4.51E+06
^{242m}Am	2.28E-01	1.15E+05	1.54E+06	9.44E+05	2.39E+05
^{243}Am	9.02E+04	2.15E+07	7.09E+06	3.12E+06	4.16E+05
^{243}Cm	1.88E+00	2.62E+04	5.14E+04	2.35E+04	2.14E+03
^{244}Cm	3.11E+05	3.33E+07	5.32E+06	1.64E+06	1.12E+05
^{245}Cm	6.53E+03	4.54E+06	5.46E+05	1.90E+05	1.27E+04
^{246}Cm	2.73E+05	2.01E+07	7.55E+05	1.27E+05	1.99E+03
^{247}Cm	8.60E+03	1.42E+06	7.71E+04	9.20E+03	7.98E+01
^{248}Cm	4.61E+04	3.17E+06	4.16E+04	3.10E+03	1.18E+01
k_{inf}	1.518	1.326	2.069	2.372	2.756

KEY NUCLIDES FOR RADIOLOGICAL RISK ASSESSMENT

Actinides which contribute to the radiological risk of radioactive waste repositories are actinides with lifetimes greater than about 10^4 a and their α active daughter products. Examples are ^{237}Np and its daughter product ^{229}Th, ^{226}Ra, a daughter product of ^{234}U, and ^{231}Pa, a daughter product of ^{235}U. Risk analyses have shown that, due to its low solubility and strong sorption, plutonium does not directly contribute to the risk. Risk contributions from extremely long–lived and therefore practically inactive nuclides as ^{238}U ($T_{1/2} = 4.5 \cdot 10^9$ a) and ^{232}Th ($T_{1/2} = 1.4 \cdot 10^{10}$ a) are small or negligible.

It is evident that the risk of waste repositories is closely related to the activity of one or two long–lived "key nuclides" – mainly uranium isotopes – from each of the four principal actinide decay chains. In general, the precursors of these nuclides have already decayed when the nuclides or their daughter products enter the biosphere. Assuming that all daugther nuclides are shorter–lived, the resulting dose or risk can be expected to be proportional to the activity of the nuclide and to an effective dose conversion factor including daughter contributions for equilibrium conditions.

A practical implementation of this concept is shown in Table 5. For each key nuclide, the table specifies the precursors contributing to its activity, an effective dose conversion factor, and nuclides which produce dominant dose contributions. The effective dose conversion factor is derived from ICRP–61 [10] assuming that all daughter products are in equilibrium and contribute equally to the dose (more realistic models for calculating effective dose conversion factors are being investigated).

Table 5: Key Nuclides for Radiological Risk Assessment

Principal decay chain	Key nuclide	$T_{1/2}$ [a]	Included precursors	Eff. dose factor [Sv/Bq]	Principal dose contributors
4n	^{236}U	$2.3 \cdot 10^7$	all	$3.0 \cdot 10^{-8}$	^{236}U
4n + 1	^{237}Np	$2.1 \cdot 10^6$	all	$1.2 \cdot 10^{-6}$	^{237}Np, ^{229}Th
4n + 1	^{233}U	$1.6 \cdot 10^5$	^{233}Pa	$5.4 \cdot 10^{-7}$	^{229}Th
4n + 2	^{234}U	$2.4 \cdot 10^5$	all from ^{238}Pu branch	$3.2 \cdot 10^{-7}$	^{226}Ra
4n + 2	^{238}U	$4.5 \cdot 10^9$	all	$3.4 \cdot 10^{-7}$	^{226}Ra
4n + 3	^{235}U	$7.8 \cdot 10^8$	all	$4.2 \cdot 10^{-6}$	^{231}Pa, ^{227}Ac[1]
4n + 3	^{231}Pa	$3.3 \cdot 10^4$	none	$4.2 \cdot 10^{-6}$	^{231}Pa, ^{227}Ac[1]

[1] Daughter products of ^{227}Ac

For the thorium chain (4n chain), ^{236}U is chosen as a key nuclide[1]. For systems based on the uranium–plutonium fuel cycle, the key nuclide of the neptunium chain (4n+1 chain) is ^{237}Np. ^{233}U is included, because it is important in thorium–based systems. With regard to its abundance in uranium–based fuels, ^{238}U is included as a second nuclide of the radium chain (4n+2 chain). Lastly, ^{231}Pa is included as a second nuclide of the actinium chain (4n+3 chain), since it is produced in significant amounts in thorium–based systems and its specific activity is much higher than that of ^{235}U.

APPLICATION OF SCHEME TO SYSTEMS WITH BATCH–TYPE PROCESSING

The actinide inventory of the waste repository per fission may be written as

$$F_i \tilde{N}_{i,j} \ ,$$

where F_i and $\tilde{N}_{i,j}$ are the number of heavy atoms in the waste stream relative to the number of fissions and the fuel composition, respectively. The latter can be obtained by normalising the asymptotic actinide inventory, viz.,

$$\tilde{N}_{i,j} = \frac{N_{i,j}}{\sum_i N_{i,j}} \ .$$

The quantity F_i – in the following to be termed "waste fraction" – depends on the burnup of the fuel and the reprocessing and fuel fabrication losses. For a batch–type processing, it is obtained from the expression

$$\frac{L_i(1-B)}{B} \ ,$$

where the burnup, B, and the nuclide–dependent losses, L_i, have to be given as fractions of the inventory.

For a waste fraction of 1.0, the scheme of the key nuclides yields the activities in Tables 6 and 7. The tables contain all activities greater than one hundredth of the dominating activity. By multiplying the activities with the appropriate effective dose conversion factors from Table 5, one obtains nuclide–specific risks. Depending on the characteristics of the waste repository, these risks are independent or cumulative. Nuclide–specific and cumulative (total) risks for burning ^{232}Th, ^{238}U, ^{237}Np and ^{239}Pu are compiled in Table 8. Burnup and losses can be accounted for by multiplying the values in the tables with the appropriate waste fractions.

ACTINIDE RECYCLING IN THERMAL AND FAST SYSTEMS

For LWRs and fast reactors, the achievable burnup, expressed in atom % of the fissile material, is about the same. If the systems use the same type of fuel cycle, it can be assumed that they also have similar reprocessing and fuel fabrication losses. In this case, the waste fractions are comparable and the numbers in Tables 6 to 8 can be used directly to assess relative activities and risks.

The suitability of LWRs and fast systems for utilising and burning plutonium, neptunium and americium may be discussed using the cumulative risk ratios in Table 9. The FR/PWR ratios show that the cumulative nuclide–specific risks for the fast reactor and the PWR lie within a factor of two, with two exceptions: ^{239}Pu burns cleaner in the fast, and ^{242m}Am burns significantly cleaner in the thermal neutron spectrum. It is noteworthy that this applies to both fissile and fissionable nuclides. As outlined before, thermal systems do, however, not provide an adequate overall neutron balance for completely burning fissionable nuclides, necessitating the use of additonal fuel which augments the risk, or a hybrid system.

[1] Due to the inclusion of the longer–lived ^{244}Pu ($T_{1/2} = 8.3 \cdot 10^7$ a) the ^{236}U activity is slightly overestimated.

Table 6: Activities of Key Nuclides, Including Decayed Precursors, from Burning Th, U, Np and Pu (Bq per fission assuming a waste fraction of 1.0)

Nucl. burnt	Key nuclide	STS	PWR	FR	FMS	SFS
^{232}Th	^{233}U	9.95E-15	6.96E-15	1.50E-14	1.31E-14	9.95E-15
	^{234}U	4.09E-15	1.52E-15	3.00E-15	2.03E-15	9.08E-16
	^{231}Pa	3.72E-18	7.24E-17	5.42E-16	3.73E-15	6.43E-15
^{233}U	^{233}U	6.28E-14	8.45E-14	9.59E-14	1.04E-13	1.17E-13
	^{234}U	1.68E-14	1.75E-14	1.86E-14	1.58E-14	1.06E-14
^{235}U	^{236}U	4.20E-16	3.39E-16	3.29E-16	3.05E-16	2.38E-16
	^{237}Np	1.82E-16	1.07E-15	1.04E-15	8.94E-16	5.60E-16
	^{234}U	1.67E-16	1.04E-14	9.23E-15	6.12E-15	2.33E-15
^{238}U	^{236}U	3.61E-17	4.25E-18	5.03E-17	3.32E-17	8.90E-18
	^{237}Np	4.33E-17	3.42E-17	1.21E-16	1.24E-16	9.94E-17
	^{234}U	5.40E-19	1.12E-16	4.81E-16	5.98E-16	3.14E-16
	^{238}U	4.50E-18	4.83E-18	4.08E-18	4.17E-18	4.51E-18
	^{235}U	1.25E-18	2.47E-19	3.12E-18	3.05E-18	1.90E-18
^{237}Np	^{236}U	8.98E-17	6.23E-17	5.47E-17	2.99E-17	8.84E-18
	^{237}Np	6.29E-15	3.83E-15	4.33E-15	5.36E-15	7.28E-15
	^{234}U	6.42E-15	3.73E-14	3.82E-14	3.43E-14	2.30E-14
^{238}Pu	^{236}U	2.47E-16	9.37E-17	9.21E-17	5.87E-17	2.17E-17
	^{237}Np	2.96E-16	5.93E-16	2.30E-16	1.38E-16	4.59E-17
	^{234}U	2.31E-14	5.61E-14	6.46E-14	7.10E-14	7.96E-14
^{239}Pu	^{236}U	3.30E-16	1.99E-16	2.88E-16	2.31E-16	1.23E-16
	^{237}Np	3.96E-16	1.53E-15	6.37E-16	4.36E-16	1.47E-16
	^{234}U	4.68E-18	4.45E-15	2.24E-15	1.43E-15	4.62E-16
	^{235}U	6.56E-18	1.16E-17	1.79E-17	2.12E-17	2.65E-17
^{240}Pu	^{236}U	3.78E-16	2.74E-16	6.50E-16	7.01E-16	7.99E-16
	^{237}Np	4.54E-16	2.10E-15	1.43E-15	1.32E-15	9.49E-16
	^{234}U	5.33E-18	6.10E-15	5.00E-15	3.96E-15	1.87E-15
^{241}Pu	^{236}U	3.33E-16	2.06E-16	1.13E-16	6.95E-17	2.34E-17
	^{237}Np	4.91E-16	2.31E-15	4.10E-15	4.87E-15	6.32E-15
	^{234}U	5.76E-18	6.72E-15	1.41E-14	1.44E-14	1.24E-14
^{242}Pu	^{236}U	3.48E-16	2.72E-16	2.23E-16	1.58E-16	6.36E-17
	^{237}Np	6.96E-17	6.43E-16	3.92E-16	2.76E-16	1.09E-16
	^{234}U	3.88E-20	9.13E-16	1.00E-15	6.02E-16	1.74E-16
	^{238}U	2.44E-18	2.18E-18	2.81E-18	3.24E-18	3.98E-18
	^{235}U	3.26E-18	5.71E-18	4.45E-18	4.37E-18	3.47E-18

Table 7: Activities of Key Nuclides, Including Decayed Precursors, from Burning Am and Cm (Bq per fission assuming a waste fraction of 1.0)

Nucl. burnt	Key nuclide	STS	PWR	FR	FMS	SFS
^{241}Am	^{236}U	3.39E-16	1.37E-16	9.77E-17	5.85E-17	1.79E-17
	^{237}Np	4.90E-16	1.82E-15	3.53E-15	4.35E-15	6.01E-15
	^{234}U	1.38E-14	3.31E-14	2.85E-14	2.78E-14	2.29E-14
^{242m}Am	^{236}U	4.23E-16	3.34E-16	2.18E-16	1.68E-16	8.45E-17
	^{237}Np	8.99E-17	7.92E-16	3.89E-16	3.00E-16	1.46E-16
	^{234}U	1.00E-15	7.66E-15	4.13E-14	4.63E-14	5.49E-14
^{243}Am	^{236}U	4.28E-16	3.66E-16	4.86E-16	4.49E-16	3.37E-16
	^{237}Np	9.11E-17	8.66E-16	8.49E-16	7.30E-16	4.08E-16
	^{234}U	5.14E-20	1.23E-15	2.17E-15	1.55E-15	5.86E-16
	^{235}U	4.23E-18	7.68E-18	9.71E-18	1.24E-17	1.84E-17
^{243}Cm	^{236}U	4.14E-16	2.35E-16	3.62E-16	2.56E-16	7.51E-17
	^{237}Np	8.82E-17	6.49E-16	6.60E-16	4.32E-16	9.01E-17
	^{234}U	1.91E-19	1.12E-15	1.82E-15	1.10E-15	3.14E-16
	^{235}U	5.12E-18	1.56E-17	1.51E-17	2.05E-17	2.84E-17
^{244}Cm	^{236}U	4.89E-16	4.60E-16	6.86E-16	7.33E-16	8.11E-16
	^{237}Np	1.04E-16	1.09E-15	1.20E-15	1.19E-15	9.82E-16
	^{234}U	1.12E-20	1.54E-15	3.05E-15	2.39E-15	1.07E-15
^{245}Cm	^{236}U	6.47E-17	5.20E-17	1.53E-17	7.12E-18	3.55E-18
	^{237}Np	2.00E-16	1.59E-15	3.93E-15	5.89E-15	8.78E-15
	^{238}U	4.02E-18	3.37E-18	2.61E-18	1.89E-18	6.64E-19

An examination of the SFS/FR cumulative risk ratios in Table 9 and the fission–to–capture ratios in Table 1 does not reveal a simple relation between these ratios. It can be seen that, in contrast to the respective fission–to–capture ratios, the cumulative risks for the fissionable nuclides ^{237}Np, ^{238}Pu and ^{241}Am (i.e. nuclides which are of primary interest in connection with the burning of LWR–discharged minor actinides) show only modest spectrum hardening effects. As regards the fissile nuclides, ^{239}Pu shows a strong positive, and ^{241}Pu and ^{242m}Am show a negative spectrum hardening effect. This means that, in connection with risk considerations, the fission–to–capture ratio is not a useful parameter.

ACTINIDE BURNING IN HIGH–FLUX SUPERTHERMAL SYSTEMS

Table 8 indicates that high–flux superthermal systems can burn ^{237}Np slightly cleaner than other systems. This applies also for other important actinides with the exception of ^{232}Th, which burns cleaner in PWRs, and ^{239}Pu, which burns slighly cleaner in systems with a superfast neutron spectrum. The basic potential of high–flux superthermal systems for burning actinides is thus confirmed.

Table 8: Nuclide–Specific Risks from Burning ^{232}Th, ^{238}U, ^{237}Np and ^{239}Pu
(μSv per fission assuming a waste fraction of 1.0)

Nucl. burnt	Key nuclide	STS	PWR	FR	FMS	SFS
^{232}Th	^{233}U	5.37E-15	3.76E-15	8.08E-15	7.08E-15	5.37E-15
	^{234}U	1.31E-15	4.86E-16	9.59E-16	6.49E-16	2.91E-16
	^{231}Pa	1.56E-17	3.04E-16	2.28E-15	1.57E-14	2.70E-14
	Total	6.72E-15	4.57E-15	1.13E-14	2.34E-14	3.27E-14
^{238}U	^{236}U	1.08E-18	1.28E-19	1.51E-18	9.97E-19	2.67E-19
	^{237}Np	5.20E-17	4.10E-17	1.45E-16	1.48E-16	1.19E-16
	^{234}U	1.73E-19	3.58E-17	1.54E-16	1.91E-16	1.00E-16
	^{238}U	1.53E-18	1.64E-18	1.39E-18	1.42E-18	1.53E-18
	^{235}U	5.26E-18	1.04E-18	1.31E-17	1.28E-17	7.98E-18
	Total	6.00E-17	7.97E-17	3.15E-16	3.55E-16	2.30E-16
^{237}Np	^{237}Np	7.55E-15	4.60E-15	5.20E-15	6.43E-15	8.74E-15
	^{234}U	2.06E-15	1.19E-14	1.22E-14	1.10E-14	7.36E-15
	Total	9.62E-15	1.66E-14	1.74E-14	1.74E-14	1.61E-14
^{239}Pu	^{236}U	9.89E-18	5.98E-18	8.65E-18	6.92E-18	3.70E-18
	^{237}Np	4.75E-16	1.83E-15	7.64E-16	5.23E-16	1.76E-16
	^{234}U	1.50E-18	1.42E-15	7.17E-16	4.59E-16	1.48E-16
	^{235}U	2.76E-17	4.86E-17	7.52E-17	8.92E-17	1.11E-16
	Total	5.14E-16	3.31E-15	1.57E-15	1.08E-15	4.39E-16

Table 9: Ratios of Cumulative Risks from Burning Different Actinides

Nuclide	Type	FR/PWR	SFS/FR
^{237}Np	fissionable	1.05	0.93
^{238}Pu	fissionable	1.12	1.22
^{239}Pu	fissile	0.47	0.28
^{240}Pu	fissionable	0.74	0.53
^{241}Pu	fissile	1.91	1.22
^{242}Pu	fissionable	0.74	0.25
^{241}Am	fissionable	1.05	1.09
^{242m}Am	fissile	3.98	1.30
^{243}Am	fissionable	1.20	0.43

On the other hand, due to their poor overall neutron balance, high–flux superthermal systems will not be able to play a dominant role as energy producers in a long–term strategy. They are suitable for burning existing actinide wastes which cannot be recycled in other systems, and remaining actinide wastes from closed conventional systems.

THE URANIUM–PLUTONIUM AND THE THORIUM–URANIUM FUEL CYCLE

The ultimate goal in nuclear energy is to extract the fission energy from all ^{238}U and ^{232}Th with a minimum long–term risk for the population. The numbers in Table 2 indicate that, from the viewpoint of the overall neutron balance, this goal can be achieved best with uranium and fast reactors. For thorium, the surplus of neutrons may not be sufficient to compensate for neutron losses. Therefore, the benefits from incorporating accelerators into the systems may be more significant for thorium–based than for uranium–based systems.

As to the goal of a small long–term risk for the population, Table 8 indicates that, in spite of the reduced build–up of higher actinides, thorium does not perform better than uranium: For ^{232}Th, the risk is dominated by ^{233}U and – in systems with a very fast neutron spectrum – by ^{231}Pa. ^{237}Np and ^{234}U contribute about equally to the risk from burning ^{238}U. It can be seen that, for all systems and even without the ^{231}Pa contribution (^{231}Pa could be separated and burnt in a thermal system), the comparison favours the uranium–plutonium fuel cycle.

SUMMARY AND CONCLUSIONS

Based on the concepts of the asymptotic actinide inventory and the key nuclides for radiological risk assessment, a general method for investigating the effectiveness of actinide transmutation systems has been developed. The method allows to assess the impact of different long–term transmutation strategies on the actinide inventories of the systems, the composition of the actinide waste, and the radiological risk associated with the disposal of this waste. In a comparative study, the method was applied to systems with a wide range of characteristics including a PWR, a fast reactor, a high–flux superthermal system, and two hybrid systems with fast neutron spectra. The principal results of the study can be summarised as follows:

- A complete burning of both fissile and fissionable actinides requires an adequate overall neutron balance. In general, this can only be achieved in systems with a fast neutron spectrum.
- The neutron spectrum can be hardened and the neutron balance can be improved by coupling an accelerator with the system. This may be particularly interesting for thorium–based systems which are less abundant in neutrons than fast systems with uranium–based fuels.
- For comparable fuel burnup and reprocessing losses, the recycling of the important transuranic actinides in fast reactors and PWRs results in similar radiological risks of the waste. Risk ratios of more than a factor of two arise in two cases: ^{239}Pu burns cleaner in a fast, and ^{242m}Am burns significantly cleaner in a thermal neutron spectrum.
- It appears that the radiological risk of the waste is not directly related to the mean neutron energy of the system or the type of nuclide being burnt. Burning fissionable nuclides in a thermal neutron spectrum is possible from the viewpoint of the risk, but undesirable, because it necessitates the use of additional fuel which indirectly augments the risk, or a hybrid system.
- With the exception of ^{232}Th and ^{239}Pu, actinides burn with the smallest risk in high–flux superthermal systems. In addition, these systems feature low actinide inventories and, consequently, also small core accident risks.
- The present analysis indicates that, in the long range, the radiological risk of the waste cannot be reduced by changing from the uranium–plutonium to the thorium–uranium fuel cycle.

ACKNOWLEDGMENT

The involvement of Dr. S. Pelloni and Mr. J.M. Paratte in the cross section preparation and useful comments by Dr. J. Hadermann concerning waste management aspects are greatfully acknowledged.

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SOME IDEAS ABOUT HYBRID SYSTEM CONCEPTS

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ABSTRACT

Hybrid systems were studied in the past for fissile material production. Subsequently these ideas were reconsidered for minor actinide and long-lived fission product destruction as alternative to the traditional final disposal of nuclear waste. Now there are attemps to extend the use of the concepts developed for minor actinide incineration to plutonium burning.

The most promising hybrid system concept considers fuel and target both as liquids.

From the results obtained, it is possible to discard solid and sodium targets. The solutions adopting composite targets seem, at the present stage, the most promising, but still there remains the problem of Pu production at a level not acceptable in a burning system.

The most suitable solvent is heavy water for minor actinide annihilation in the blanket of a hybrid system.

Due to the criticality conditions and the necessity of electric energy production, the blanket using plutonium dissolved in molten salts is the most convenient one.

1. INTRODUCTION

A hybrid system is composed of a particles accelerator, a target, for neutron production, a blanket, for transmutation or fissile material production, and their respective interfaces (i.e. the window between accelerator and target).

These systems were studied in the past for fissile material production. In the last years these ideas were reconsidered for minor actinide (MA) and long-lived fission product destruction as alternative to the traditional final disposal of nuclear waste.

As plutonium is the most important of the actinides (about 95% in mass), the incineration of minor actinides is nonsense if the plutonium problem is not solved .

Plutonium being an excellent nuclear fuel, the most convenient solution should be its burning in a nuclear reactor to produce electric energy. But in the case of very big stockpiles, it seems interesting to burn a part of it in less efficient, but less time consuming systems, such as thermal hybrid systems as demonstrated by the Academy of Science of the United States.

In the present paper different solvents, moderators and target materials are analyzed for MA and plutonium burning.

The most promising hybrid system concept considers both fuel and target as liquids.

2. MINOR ACTNIDES

^{237}Np, ^{238}Np, ^{241}Am, ^{242m}Am, ^{243}Am , ^{242}Cm, ^{243}Cm, ^{244}Cm, ^{245}Cm, ^{246}Cm and ^{247}Cm.are considered MA taking into account the concentration existing in PWR standard spent fuel reprocessed after 10 years cooling.

3. PLUTONIUM

In the present paper, plutonium means all the actinides present in the discharged PWR standard spent fuel except the uranium isotopes. Standard PWR spent fuel is defined as UO_2 fuel enriched at 3.3% irradiated at 33000 MWd/t

4. TARGET

The target can be solid or liquid. In the first case, it is possible to consider two accident scenarios: loss of coolant and loss of beam. In the first case if the beam is not switched off quite soon the target will melt: in the second one,the fast quenching can provoke a strong thermal shock.

The molecular dynamic calculations seem to demonstrate that long-range order disappears in the areas travelled through by very energetic particles; this means that in these zones the material of the target becomes similar to glass. This result should be confirmed by the behaviour of the tungsten target at the Paul Scherrer Institute (PSI) [1] after a thermal shock it broke into very small parts, practically like powder.

The previous considerations seem to indicate the convenience of liquid targets.

In the case of a heavy metal flowing target, the protons do not penetrate greatly and the majority of the neutrons are produced in the first 20 cm of the target. The most interesting manner to flatten the neutron flux shape is to use composite targets: a inner liquid low mass target, such as lithium or sodium (primary target), surrounded by a solid secondary target made of heavy metal (such as lead or uranium).

A set of calculations for pure materials was carried out considering uranium, tungsten, lead, tin, sodium and beryllium. Uranium targets produce some amounts of ^{234}U, ^{235}U and ^{236}U as spallation product and ^{239}Pu due to epithermal neutron absorption; lead produces the long-lived ^{206}Pb; low mass number targets have low neutron yields and a very high fraction of neutrons are in the high energy range (E > 20 MeV). Due to the low neutron yield, it is possible to discard sodium as spallation target and tungsten because it is strong neutron absorver.

A second set of calculations was performed for composite targets (outer dimensions 20 cm diameter and 60 cm high), U-Be and Pb-Be, with the central beryllium zone with different diameters. Low energy neutron (E < 20 MeV) distribution in the target is significantly improved. The neutron yield is drastically reduced and the production of U isotopes remain high in the case of uranium targets. The neutron yield for U-Be target with a 2 cm diameter for the beryllium zone (22.94) is quite similar to the yield corresponding to lead (23.35).

A third set of calculations was performed for a target containing thefuel. This is plutonium dissolved in ^{7}LiF. The same Pu concentrations of the fuel (10, 50, 100 g/l) were considered. The maximum neutron yield achieved is 2.32 with a high energy neutron fraction about of 0.33 at 100 g/l; this means that direct spallation on fuel is not convenient.

Taking into account all the parameters considered it is most convenient to use a lead target.

5. BLANKET

For a neutron flux greater than 1.0 10^{15} n/(cm^2 s) it is impossible to use the traditional system: solid fuel and cooling water. In fact some blanket proposals consider liquid fuel: actinide dissolved or dispersed in heavy water. The most recent blanket concepts are based on plutonium or minor actinides dissolved in molten salts, particularly ^{7}LiF - ^{9}BeF, used in the Molten Salt Breeder Reactor Experience at the Oak Ridge National Laboratory.

A series of ANISN cell calculations were carried out considering Pu and MA:

1 - dissolved in heavy water and moderated by heavy water;

2 - dissolved in ^{7}LiF and moderated by heavy water;

3 - dissolved in ^{7}LiF and moderated by graphite.

Pu is at the above mentioned concentrations and MA at 2.5, 5, 10, 50 and 100 g/l as initial concentrations, the maximum neutron multiplication factor (K_{inf}) were verified with MCNP:

5.1 K_{inf} results for MA

The highest K_{inf} values correspond to the case of MA moderated and dissolved in D_2O; the minimum one for MA dissolved in ^{7}LiF and moderated by graphite. In this last case to reach K_{inf} values close to 1.0 it is necessary to consider MA concentrations higher than 500 g/l while the same figure is achieved with a MA concentration about 10 g/l for heavy water. This means that burners using D_2O as solvent and moderator can run with much smaller MA inventory than the corresponding ones having ^{7}LiF as solvent and graphite as moderator.

Taking into account the potential danger of the MA it should be possible to use a less efficient thermal cycle but using D_2O improves the safety conditions of the blanket.

On the other hand for burning plutonium it should be necessary to obtain the maximum efficiency as regards neutron economy therefore burning simultaneously the long-lived fission products should be not convenient. Nevertheless heavy water has better absorption properties than the molten salts.

It seems more convenient to burn simultaneously MA and long-lived fission products using heavy water as solvent and moderator. The long-lived fission products can be placed in the reflector.

5.2 K_{inf} results for Plutonium

The highest K_{inf} values correspond to the case of Pu moderated and dissolved in D_2O; the minimum one for Pu dissolved in ^7LiF and moderated by graphite.

In any case the minimum K_{inf} value is 1.45550; this means that the three sets of solvent-moderator studied are equivalent.

The outlet temperature of the molten salts can reach values of about 640 °C without increasing the pressure, for D_2O is 300 °C at 10 MPa. The efficiency of the thermal cycle for molten salts can reach 42%, against 33% for D_2O.

The increase of efficiency means that the same amount of electric energy is produced reducing the mass of fission products, particularly the long-lived ones.

In [2] another important advantage of the molten salts blanket is analysed that concern its reprocessing.

Hybrid systems having lead target with an effective radius of the fissile zone of 250 cm and a height of 500 cm reach the criticality for a Pu concentration of 7.5 g/l and the corresponding neutron flux is 1.1 10^{15} n/(cm^2 s). With characteristics preliminary these calculations demonstrate that it is possible to burn about 552 kg of Pu per year.

6. CONCLUSION

From the results obtained, it is possible to discard solid and sodium targets. The most convenient solution for target material is, at the present stage, liquid lead, but it is necessary to consider research work concerning composite targets, because these still remains the problem of Pu production, which is not acceptable in MA or Pu burning systems.

It seems more convenient to burn simultaneously MA and long-lived fission products using heavy water as solvent and moderator .

For energy production, the blanket using plutonium dissolved in molten salt allows higher thermal efficiency.

The considered hybrid concept is suitable for plutonium burning.

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EXPERIMENTAL DETERMINATION OF THE ENERGY GENERATED IN NUCLEAR CASCADES BY A HIGH ENERGY BEAM

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Abstract

An already existing, sub-critical arrangement made of natural Uranium and water moderator has been exposed to a low intensity ($\approx 10^9$ ppp) proton beam from CERN-PS at several kinetic energies from 600 MeV to 2.75 GeV. The energy delivered by the hadronic cascade induced by the beam in the device has been measured by the temperature rise of small sampling blocks of Uranium located in several different positions inside the device and counting the fissions in thin probe foils of natural Uranium. We find typically $G \approx 30$ in reasonable agreement with calculations, where G is the ratio of the energy produced in the device to the energy delivered by the beam. This result opens the way to the realisation of the so-called Energy Amplifier, a practical device to produce energy from Thorium or depleted Uranium targets exposed to an intense high energy proton beam. Results show that the optimal kinetic energy is ≥ 1 GeV, below which G decreases but is still acceptable in the energy range explored.

Nuclear reactors produce today a significant fraction (≈ 6%) of the world's energy supply and they are likely to continue to do so in the foreseeable future. Notwithstanding, new approaches to energy extraction from nuclei are of interest, especially provided they could eliminate or at least greatly reduce (i) the environmental impact of the long-lived highly radioactive waste; (ii) the possibility of diversions toward military applications; (iii) the risks of an accidental divergence related to the critical operation of the chain reaction and (iv) make a more efficient use of a fuel which is less radio-toxic to extract and more abundant on Earth than Uranium.

Thermonuclear fusion may hold the expectation of satisfying all these requirements, but only in a very distant future and with very sophisticated technologies. Recently some of us [1] have elaborated a much simpler scheme energy extraction from nuclei, called "Energy Amplifier" (EA) allowing in particular the use of natural Thorium as a fuel. This is based on a proton initiated, high energy (≈ 1 GeV) nuclear cascade absorbed in a "calorimeter". A calorimeter is made of a large number of heavy material elements alternated with a sampling medium, usually scintillator or liquid Argon. Such instruments are widely used to measure the energy for instance of parton "jets" through observation of the energy depositions of fully contained nuclear cascades. The fraction of the energy lost by hadrons in the sampling material can be increased by some 30–50% by adding some fissionable material in the plates [2]. This method is used in order to equalise the response of calorimeters to hadronic and electromagnetic cascades, as required for instance in measurements of jets. In the present paper we demonstrate experimentally that it is possible to "amplify" the energy deposition of a high energy cascade to such an extent as to potentially recover the energy of the incident beam and to make in addition a considerable net energy gain. In the experimental conditions of this investigation, the energy produced by the cascade has been typically some thirty times the one delivered by the beam.

In a proton induced cascade one can distinguish two qualitative and successive physical regimes: (1) a spallation driven, high energy phase and (2) a neutron driven, fission dominated regime. When the energy of the cascade products falls below several MeV, ionisation losses bring particles rapidly to rest, with the exception of neutrons which continue to exhibit a rich phenomenology eventually down to thermal energies. Neutrons from the first phase are acting as "source" for the second phase. The first phase has been widely explored in calorimeters: since it is rather complicated and the relevant cross sections are often poorly known, it can be approximately parametrized with the help of phenomenological models and Monte Carlo calculations [3], generally sufficiently detailed to give a first order agreement with the experimental measurements [4].

In the second phase (for which an almost complete set of cross sections exists) the main physical process is a diffusive process of neutrons which gradually lose energy by collisions and are multiplied by fissions and (n,2n) reactions. This phenomenology is reminiscent of the one of Nuclear Reactors. However, because the EA is not critical, there are important differences : in a Reactor the flux distribution inside the volume is determined essentially by the boundaries [5], in the EA the location and geometry of the initial cascade acting as neutron source is dominant. A simple analytic theory based on diffusion has been developed [6]. The neutron source excites a superposition of ortho-normal modes of the "buckling" equation representing the neutron flux, of which only the fundamental mode is relevant in a Reactor Theory. In this description a Reactor is a limiting case in which the strength of the initiating source tends to zero and the criticality to one. Each of these modes has a different buckling parameter B_i and a different multiplication coefficient k_i. As a consequence, in the conditions of our experiment in which the microscopic multiplication coefficient k_∞, defined as the number of neutrons produced at each absorption in the fuel-moderator mixture is $k_\infty<1$, (i) the spatial neutron flux is expected to decay exponentially from the point of the source [7], rather than having the characteristic cosine distribution centred with respect to the volume as in a critical Reactor [5] and (ii) for a proton pulse sharp in time (δ-function), the neutron population decreases exponentially with a time constant which grows linearly with the effective multiplication coefficient $1/(1-k)$ [6].

The presence of such a second neutron driven phase is essential in order to achieve a large gain. However, it is not necessary to let neutrons reach full thermalization. Different types of EA are possible, according to the amount of moderation introduced for the neutrons before capture. In one instance one may use liquid Lead as a cooling medium and as high energy target, with consequent little or no moderation. In other schemes neutrons may be either partially or completely moderated. We refer to Ref. [8] for more details. In the present test neutrons were almost completely thermalized.

The experimental set-up is shown in Fig. 1 and its main parameters are summarised in Table 1. It consists of an already existing sub-critical assembly [9] made of natural Uranium rods, immersed in a stainless steel tank filled with ordinary, de-mineralised water, which acts as moderator. The beam from the CERN-PS hits a small target of depleted Uranium, which is located such as to approximately centre the source of spallation neutrons with respect to the device. A low density channel (Styrofoam) removes most of the material along the beam path in its way to the target. The beam position has been continuously monitored with wire chamber hodoscopes. The beam kinetic energy has been varied in the interval 600 MeV to 2.75 GeV. The proton beam intensity has been typically of the order of 10^9 ppp on a $\leq$ 1.0 cm radius focal spot, in the form of a sharp ($\leq$ 100 ns) fast extracted bunch. The beam intensity has been accurately measured by a beam transformer, periodically cross calibrated with the activation of Aluminium foils and it is known to better than 3%. The beam intensity is far too small to produce bulk transformations of the fuel and one relies entirely on the natural presence (0.71%) of ^{235}U.

The neutronic behaviour of the assembly has been calibrated with the help of a 1.4 Ci neutron source (Am-Be) inserted in the centre of the device. The neutron flux measured with a Boron loaded counter is shown in Fig. 2 and confirms the expected exponential behaviour as a function of the distance from the source. We note that the water extends somewhat around the uranium bar assembly, acting as a "reflector". Comparing the source results with measurements with only water, we find an effective multiplication coefficient for a point-like centred source of $k = 0.915 \pm 0.010$ [10]. This result is in good agreement with calculations (Monte Carlo) which give $k = 0.92$. We note that the k factor for the measurement with the beam is slightly different because (1) the source is not point-like (2) additional materials (styrofaoam, target etc.) have been introduced. The calculated reduction is $\Delta k = -0.02$, leading to a value for the beam configuration of $k = 0.895 \pm 0.010$.

The parameters of the detectors are listed in Table 2. The energy delivered by the beam in the device, typically ≈ 1 watt, has been measured by the temperature rise of small sampling blocks of Uranium [11] moved at different positions inside the device. Thermometers have been cross calibrated with a known energy deposition produced by a known current pulse in a resistor within each block [12]. This method has been chosen since it is the closest to their expected use in the EA test. In practice the temperature behaviour in a succession of beam-on beam-off cycles (typically one hour each) has been measured and fitted with a simple phenomenological model in which all relevant effects, like heat leaks etc. have been taken into correct account. A "zero" measurement has been performed with a lead block which is known to be largely transparent to low energy neutrons. The major contribution to the gain is expected from fission. This has been demonstrated counting the fissions in thin (≈ 1 mg/cm^2) Uranium foils near the thermometers with the help of Lexan foils, in which fission fragments produce a characteristic hole after etching [13]. The mass of the foils has been determined by alpha counting of the alpha activity of natural Uranium.

To build up a fine grid of measurements inside the device, a large number of counters sensitive to fissions mounted at regular distances on supporting rods have been inserted between the Uranium bars and moved in different, successive positions during the data taking. These detectors (see Table 2) detect fission fragments from a thin ($\approx$ 1 mg/cm^2) Uranium foil with the help of solar cells operated as semiconductor detectors [14] and Argon proportional counter gaps at 3 ata [15]. Pulse-height and time information of each count are individually recorded over a window of 800 μs after the beam pulse. The Uranium foils can be displaced away periodically in situ during the data taking in order to determine the (negligible) background level due to interactions other than fissions in the foils. Typically one records some 20–100 fission/pulse, of course with rates widely dependent on the position of the counters. The proton beam intensity has been varied in order to ensure a data rate free of pile-ups.

Electronic counters are insensitive to the prompt neutrons and charged particles ($\approx$ 5%) because of saturation around $t = 0$ and to delayed neutrons ($\approx$ 7%), but they allow to observe exponential decay of the activity. A typical distribution is shown in Fig. 3 and shows good agreement with the (asymptotic) exponential behaviour predicted by the theory. The value of the multiplication constant determined with this method is $k = 0.89 \pm 0.03$, in agreement with expectations. A value of k has also been determined using the delayed neutron signal. The result, $k = 0.915 \pm 0.010$ is in good agreement with the other methods.

In order to calculate the energetic gain G of the device, defined as the ratio between the energy produced in the device divided by the energy delivered by the beam, we must integrate the energy depositions sampled by the thermometers over the full volume. A correction (typically $\leq$ 20%) must also be applied for the different opacities of the Uranium in the bars and in the thermometers and for the fact that the insertion of detectors depletes some of the moderating medium nearby. We have parametrized the spatial distribution of the energy deposition with a formula of the type $\phi(x,y,z) = const \times \exp(-d/\lambda)$, where $d = \sqrt{(\alpha x + \delta)^2 + y^2 + z^2}$ (x-axis along the beam line, z-axis vertical) with the (fudge) parameters δ and α taking into account respectively the average effective longitudinal displacement and the first moment of the longitudinal extent of the hadronic high energy cascade (neutron source) and λ the common exponential decay length. Such a paramatrization is in

agreement with the information of the electronic counters and the Monte Carlo simulation of the device. The fitted values for the parameters based on some 10^3 counter position measurements and the thermometer measurements show excellent agreement with the parametrization and give (1) $\alpha \approx 0.85$ slightly increasing with energy, (2) an universal exponential slope, as expected by the behaviour of the subcritical device and (3) a progressive movement of the centroid of the beam source δ as shown in Fig. 4a. The experimental dependence of the fission rate as a function of the distance d is shown in Fig. 4b for the different measurement methods (in excellent agreement). The same quantity, normalised to the expectation of the parametrization is given as function of the cosine of the angle with respect to the beam direction in Fig. 4c. Agreement is good, especially if one take into account that only an integral over the distribution is needed in order to determine G .

The integrated values of G from (1) the thermometers (2) from the fissions measured with the Lexan foils and (3) the electronic counters after the indicated corrections as a function of the proton kinetic energy are in excellent agreement. Combined results are shown in Fig. 5. The gain G is essentially constant above a proton kinetic energy of 1 GeV and drops somewhat for lower values. A practical EA can therefore be operated conveniently with proton beam energies of the order of 800 MeV to 1.2 GeV. A sector focused cyclotron scaled up from the PSI machine appears to be the most adequate device to produce currents of the order of 10 mA in this energy range [16].

Results (Fig. 5) are in satisfactory agreement with Monte Carlo calculations based on FLUKA [3] for the energy cascade complemented by a home made Monte Carlo based on the ENDF-6 cross sections [17] for the neutronic behaviour. Typically we find agreements of the order of 10% for the gain, the spatial distribution of the fissions and the time decay of the time dependence of the neutron activity. This confirms the validity of our previous predictions on the EA [1],[8], then based only on the Monte Carlo.

A simple expression can be used to calculate the energy gain of a general EA, $G = G_0 / (1-k)$, where G_0 relates to the efficiency of the spallation regime and the well known *(1-k)* factor with k as the effective multiplication coefficient, relates to the neutron driven part of the cascade. Using $k = 0.895$ from the source measurement and the values for 1 GeV proton energy, we find $G_0 = 3.1 \pm 0.4$. A gain $G = 62 \pm 8$ is then expected with a somewhat larger device and $k \approx 0.95$, which is safely away from criticality. The energy produced by the EA (E_{ea}) has to be spent in part to run the accelerator. We define the "commercial" gain G_C as E_{ea}/E_{acc} where E_{acc} is the energy output of the accelerator. Assuming a realistic accelerator efficiency of *0.4* [16] and an efficiency of transformation from heat into electricity of *1/3*: $E_{ea} = E_{acc} \times G - E_{acc}/(0.4 \times 1/3) = E_{acc} \times (G - 7.5)$; $G_C = G - 7.5$. For $k = 0.95$ we obtain a comfortable "commercial" gain $G_C = 54.5 \pm 8$. With a 10 mA, 1 GeV accelerator [16], a suitable EA should deliver a net power of 545±80 MW thermal or about 181±30 MW electrical.

Although the possibility of a high gain has been demonstrated, the set-up of Fig. 1 cannot be used immediately for an extended power production. Some substantial modifications are required [1][8]. In a practical, full scale EA the rate of interactions will be much larger than in the present test. As a consequence, nuclear transmutations of elements are sizeable and modify the evolution of the subsequent cascades. This different regime is quite useful since it permits to "breed" fissionable nuclei from the bulk material of the target and thus (1) achieve useful gains from targets which otherwise would not be suitable, like for instance natural Thorium (Th^{232}) or depleted Uranium (U^{238}); (2) the fissionable fuel being continuously regenerated from the bulk material rather than only supplied initially with the fuel, a much longer burn-up is possible. Previous experience with reactors indicate that most of these procedures are basically feasible.

According to the EA proposal [8], after an extended exposure of the target to the beam (corresponding to the burning of some 3 – 15 % of the mass of the target, depending on the specific conditions) the used fuel is extracted and after an adequate cool-down period is (1) topped with fresh Th^{232} or U^{238} ; (2) many reaction products which are either stable or environmentally acceptable ($T_{1/2} \leq 32$ years) elements are extracted and stored in a "secular" repository and (3) new regenerated fuel is constituted with the rest. Over the lifetime of the plant, the composition of the fuel will change and reach an equilibrium condition, as a balance between production and "neutron incineration". Similar schemes of transmutation and/or energy generation by nuclear cascades have been proposed, in particular the (ADWT) one from Los Alamos based on molten salt, thermal neutrons, high flux capability and continuous recirculation of the fuel [18] and others [19]. The experimental results reported here have of course equally positive relevance for these proposals.

The conclusions of the analysis of a full scale device [1][8] are also that (1)Thorium is a far cleaner fuel than U^{238} and it is used very efficiently (750 kg of natural Th^{232} deliver the same useful energy (800 MWatt × year) as 167 tons of natural Uranium with the ordinary Pressurised Water Reactors (PWR) operating with isotopically enriched Uranium) (2) the fuel properties are such to permit indefinite recycling (3) the long term total radio-toxicity spilled in the environment due to mineral mining and chemical handling of the fuel is at least four to five order of magnitudes smaller than the one for PWR's for an equivalent power production [20] and (4) diversion of the spent fuel to military applications can be made practically impossible or extraordinarily difficult [8][21].

We believe that in most respects the EA (or equivalent scenarios) is comparable in performance to Thermonuclear Fusion. Both approaches offer practically unlimited fuel resources: the energetic content of Lithium on the Earth's crust needed by Fusion is estimated to be seven times the one of Thorium and they are both adequate for millions of years of very intensive utilisation. However the EA can be built economically [22], in a variety of sizes and it offers a much greater flexibility of utilisation. Moreover it presents no major technological barriers and it is far more suited because of its simplicity as an alternative to fossil fuels and to respond to the growing energy demands of the developing countries.

We are grateful to UPM and ENRESA for their support in making available the assembly, to the Spanish and Swiss Office of Energy for their special care in providing the necessary authorizations, to the CERN ECP/DS and ECP/ESS/OS Groups for having provided and supported the CASCADE data acquisition system, and to the Commissariat à l'Energie Atomique-Marcoule for providing the neutron source used for calibration. In particular, we would like to mention the outstanding dedication of R. Cappi, L. Durieu and J.P. Riunaud of the CERN PS Division who succeeded in stretching the performance of the accelerator to accommodate the needs of the experiment. This experiment would not have been possible without the enthusiastic contribution of many members of all the collaborating institutions.

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3. OECD has organised an "Intercomparison of Data and Codes for Intermediate Energy". The result of this exercise for thin targets shows that proton and neutron double differential cross-sections can be considered reliable to better than a factor of 2 and single differential cross sections are coherent to within 30%. The thick target results - which incorporate inter alia transport properties - are more relevant to Accelerator Driven Systems. They are still being anlysed by the OECD task force. We have therefore chosen the FLUKA code which has been very successful at CERN in calculating shielding and the properties of calorimeters, (see ref. 4). FLUKA is largely adequate for the low energy neutron part of these and other High Energy Physics applications, but has not been designed for reactor type calculations. We have stopped, somewhat arbitrarily, the FLUKA calculation when the energy of the neutron falls below 1 MeV and a special transport code developed by one of us (C. Rubbia) and based on pointwise cross sections has been used. Concerning the FLUKA code, we refer in particular to A. Fassó et al, in "Intermediate Energy Nuclear Data: Models and Codes", Proceedings of a Specialists' Meeting, Issy les Moulineaux (France) 30 May-1-June 1994, p. 271, published by OECD, 1994, and references therein.
4. A. Fassó, A. Ferrari, J. Ranft, P.R. Sala, G.R. Stevenson and J.M. Zazula, Nuclear Instruments and Methods A, 332, 459 (1993), also, CERN Divisional Report CERN/TIS-RP/93-2/PP (1993).
5. See for example S. Glasstone, "Principles of Nuclear Reactor Engineering", D. Van Nostrand Inc. (1955).
6. C. Rubbia, "An Analytic Approach to the Energy Amplifier", Internal Note, CERN/AT/ET/ 94-036. Also C. López, Private Communication.
7. C. Rubbia, "Why So Many Exponentials?", Internal Note, CERN/AT/ET/ 94-032.

8. C. Rubbia, "A High Gain Energy Amplifier Operated with Fast Neutrons", Contribution to the Las Vegas Conference on Accelerator Driven Transmutation Technologies and Applications 25-29 July 1994, and C. Rubbia et al, to be published.
9. F. Carminati, P. Cennini, J. Galvez, C. Geles, R. Klapisch, P. Mandrillon, J. Oropesa, J.P. Revol, Ch. Roche, C. Rubbia, J.A. Rubio and F. Saldana "A Test of the Basic Concepts of the Energy Amplifier", Internal Note, CERN /AT/ET/ 94-007 and references therein.
10. Calero et al., "Measurements of Relative Neutron Flux in the Subcritical Assembly used for the First Energy Amplifier Test", Internal Note, CERN/AT/ET/94-041.
11. Calero et al., " Experimental Temperature Measurements for the Energy Amplifier Test", Internal Note, CERN /AT/ET/ 94-038.
12. C. López, S. Vieira et al., "Report on the Energy Output Measurement System for the Energy Amplifier Experiment", Internal Report Universidad Autónoma de Madrid-Instituto de Ciencia de Materiales 'Nicolas Cabrera', September 1994.
13. Angelopoulos et al., "Experimental Determination of the Gain in the Energy Amplifier", Internal Note, CERN/AT/ET/ 94-043 and paper subm. to NIM.
14. J. M. Loiseaux and H. Nifenecker,"Proposal for the Determination of the Physical Parameters of an Energy Amplifier", Internal Note, CERN/AT/ET 94-018, F. Attale et al., "Calibration of the Grenoble Detectors", Internal Note, CERN/AT/ET 94-035 and S. Andriamonje "Caractéristiques des Detecteurs Photovoltaïques pour la détection des fragments de fisssion à l'intérieur des barreaux d'Uranium" Internal Note, CERN/AT/ET 94-044.
15. F. Casagrande and P. Cennini, "Fission Fragment Argon Gas Detector for the Energy Amplifier Test", Internal Note, CERN/AT/ET 94-042.
16. C. Rubbia, P. Mandrillon and F. Fietier, "A High Intensity Accelerator for Driving the Energy Amplifier for Nuclear Energy Production" in Proceedings of EPAC94, London, June 1994, to be published; Th. Staimbach et al., "Potential of Cyclotron Based Accelerators for Energy Production and Transmutation", Contribution to the Las Vegas Conference on Accelerator Driven Transmutation Technologies and Applications, 25-29 July 1994.
17. The ENDF formats and libraries are coordinated by the National Nuclear Data Center (NNDC) and decided by the Cross Section Evaluation Working Group, (CSEWG), a cooperative effort of national laboratories, industry and universities in the US, and AECL/ Chalk River, Canada.

18. C. Bowman et al., Nucl. Instr. Methods, A320, 336 (1992). More recent developments are to be found in the Proceedings of the International Conference on Accelerator-Driven Transmutation Technologies and Applications, Las Vegas, 25-29 July 1994.
19. The first proposal for large scale use of spallation to cause transmutation (in that case to breed nuclear fuel from a fertile element) came from Chalk River (W. Lewis, AECL report 969, 1953). It is only recently that this has surfaced again as a practical proposition. In addition to the work by the Los Alamos group quoted in reference [16], several different proposals have been made. We quote in particular the PHOENIX concept at Brookhaven (G. Van Tuyle et al. BNL report 52279 (1991) and H. Takahashi, Fusion Technology, 20 657, 1991; the OMEGA project at JAERI (T. Takizuka et al, Specialist Mtg. on Accelerator-driven Transmutation for Radwaste and other Applications, Stockholm 24-28 June 1991) and the project at ITEP, Moscow (V.D. Kazaritsky at al. in "Accelerators Applied to Nuclear Waste", 8th Journées Saturne, Report LNS/Ph/94-12).
20. C. Rubbia et al. (to be published).
21. J. Magill, C. O'Caroll, P. Gerontopoulos, K. Richter, J. Van Geel, "Advantages and Limitations of Thorium Fuelled Energy Amplifiers", Contribution to the Nuclear Society 5th Annual Conference on Nuclear Power and Industry, Obninsk, 7-11 November 1994.
22. C. Geles, G. Lindecker, P. Mandrillon, C. Roche, C. Rubbia, J.A. Rubio, "Energy Amplifier: Notes on Cost Analysis", Internal Notes, CERN/AT/ET 94-002,94-011, and 94-011 addendum; D. Finon and P. Menanteau, "Premiers Eléments d'Analyse de l'Evaluation Economique de l'Amplificateur d'Energie", Institut d'Economie et Politique de l'Energie du CNRS, Grenoble 1994, Internal Report and to be published.

FIGURE CAPTIONS

Fig. 1 Top view of the subcritical assembly. Distances are in mm.

Fig. 2 Neutron counting as a function of the distance to the source. The triangles correspond to the Monte Carlo simulation data.

Fig. 3 Time dependence of fission rate. The line corresponds to the MonteCarlo simulation.

Fig. 4a x-Displacement of the cascade centre as a function of the beam kinetic energy. The line is the result of an empirical exponential fit to the data.

Fig. 4b Number of fissions/mg/pulse (10^9 protons) as a function of the distance to the centre of the cascade. The circles, triangles and points correspond to the Thermometer, Lexan and Semiconductor counters measurements respectively. The line is the result of an exponential fit to the semiconductor counters data. Small deviations of the fit at small distances are due to the finite dimensions of the source.

Fig. 4c Residuals (ratio between experimental data and fit) as a function of $\cos(\theta)$, θ being the polar angle of the detector position respect to the beam direction. It shows no forward/backward asymmetries in the fission rate.

Fig. 5 Average energy gain from the different detectors as a function of the beam kinetic energy. The continuous line is the result of an empirical fit to the data. The dashed line corresponds to the modified FLUKA [3] simulation.

Table 1: Parameters of the sub-critical assembly.

Materials:		
Fuel	*Natural Uranium*	
Moderator	*Light Water*	
Reflector	*Light Water*	
Tubes holding the fuel (cladding)	*Aluminium*	
Tank	*Stainless Steel*	
Total mass:		
Fuel	3.62	T
Moderator	0.34	T
Tubes, tank, Reflector	0.35	T
General dimensions: Fuel		
General shape	*Hexagonal prism*	
Lattice cell	*Hexagonal*	
Bar infra-centre distances	5.08	cm
Fuel height	107.00	cm
Fuel diameter	89.00	cm
Volume moderator/Volume fuel	1.77	
General dimensions: Reflector		
Lower, upper thickness	16.0	cm
Lateral thickness, max.	15.5	cm
Lateral thickness, min.	12.5	cm
General dimensions: Tank		
Height	152.4	cm
Diameter	122.0	cm

Table 2.- Summary of detector parameters.

Detector	Active Element	Uranium converter	Clustering	Location	Number of units
Gas Ionisation Chamber 4 ata Argon	Circular window, Φ=15 mm vertical plane	1 mg/cm^2 deposit	1 array of 6, spaced 10 cm vertically . 2 arrays of 8, spaced 12.3 cm vertically	in water, between U bars	22
Amorphous Si Diode thickness 300μm	Rectangular. window 9.6 x 11.6 mm^2 vertical plane	1 mg/cm^2 deposit	10 arrays of 16 counters, spaced 6.4 cm vertically	in water, between U bars	160
Amorphous Si Diode thickness 300μm	90° sector r= 14 mm horizontal plane	1 mg/cm^2 deposit	4 clusters of 6 counters spaced 21.3 cm vertically	in fuel bar, between U cartridges	24
Thermistances in metallic probes	U Cylinders Φ=8 mm Pb Cylinder Φ=10 mm	~ 55 g U	3 thermometers with two U probes	in water, between U bars	6
Lexan foils track detectors	Equilateral triangles r=37mm Circles Φ=32 mm Rectangle 10x25 mm^2 Vertical plane	~ 1 mg/cm^2	5 vertical sets of 2 detectors	in water, between U bars	10

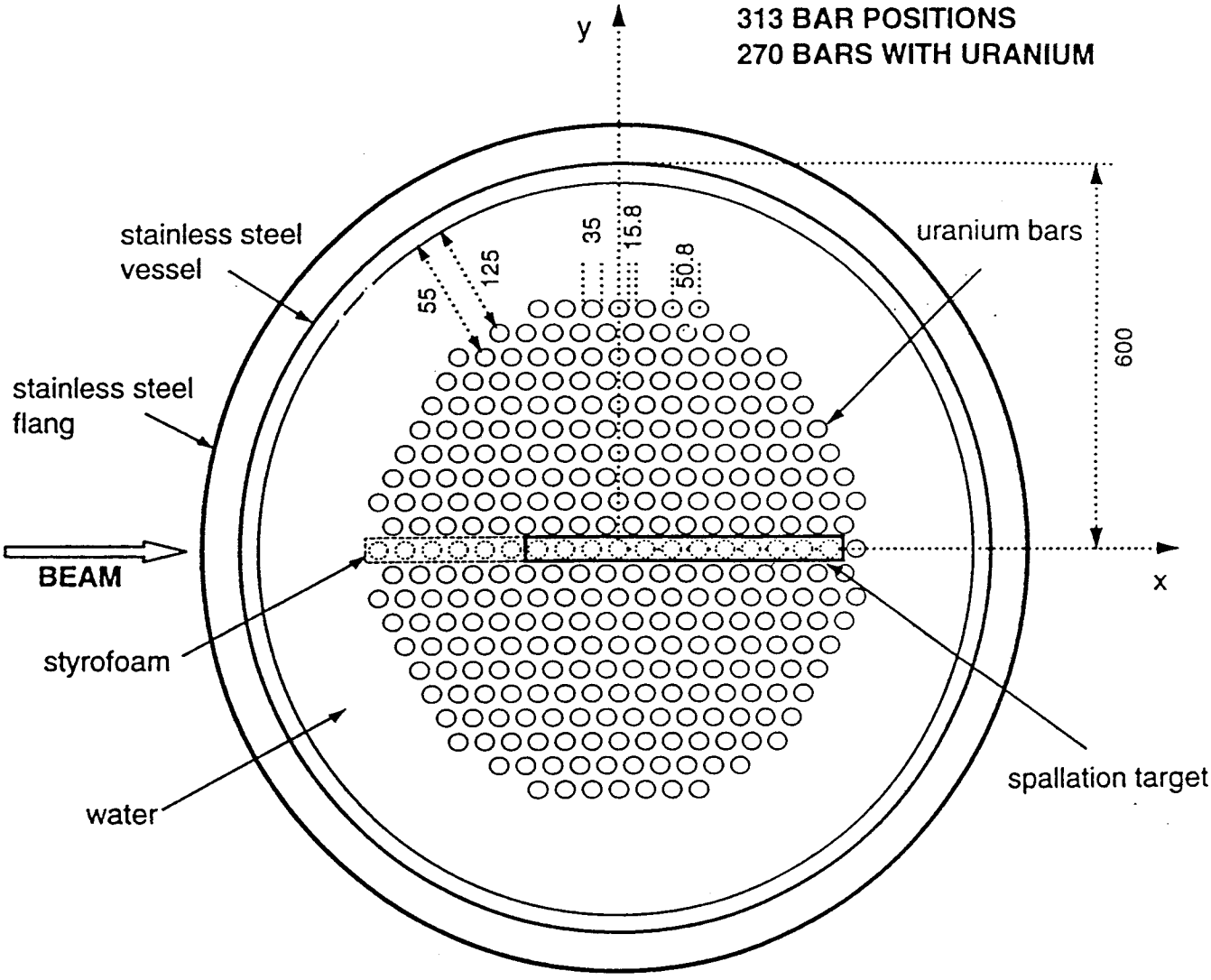

313 BAR POSITIONS
270 BARS WITH URANIUM
y
x
stainless steel vessel
stainless steel flang
BEAM
styrofoam
water
uranium bars
spallation target
55
125
35
15.8
50.8
600

Figure 1

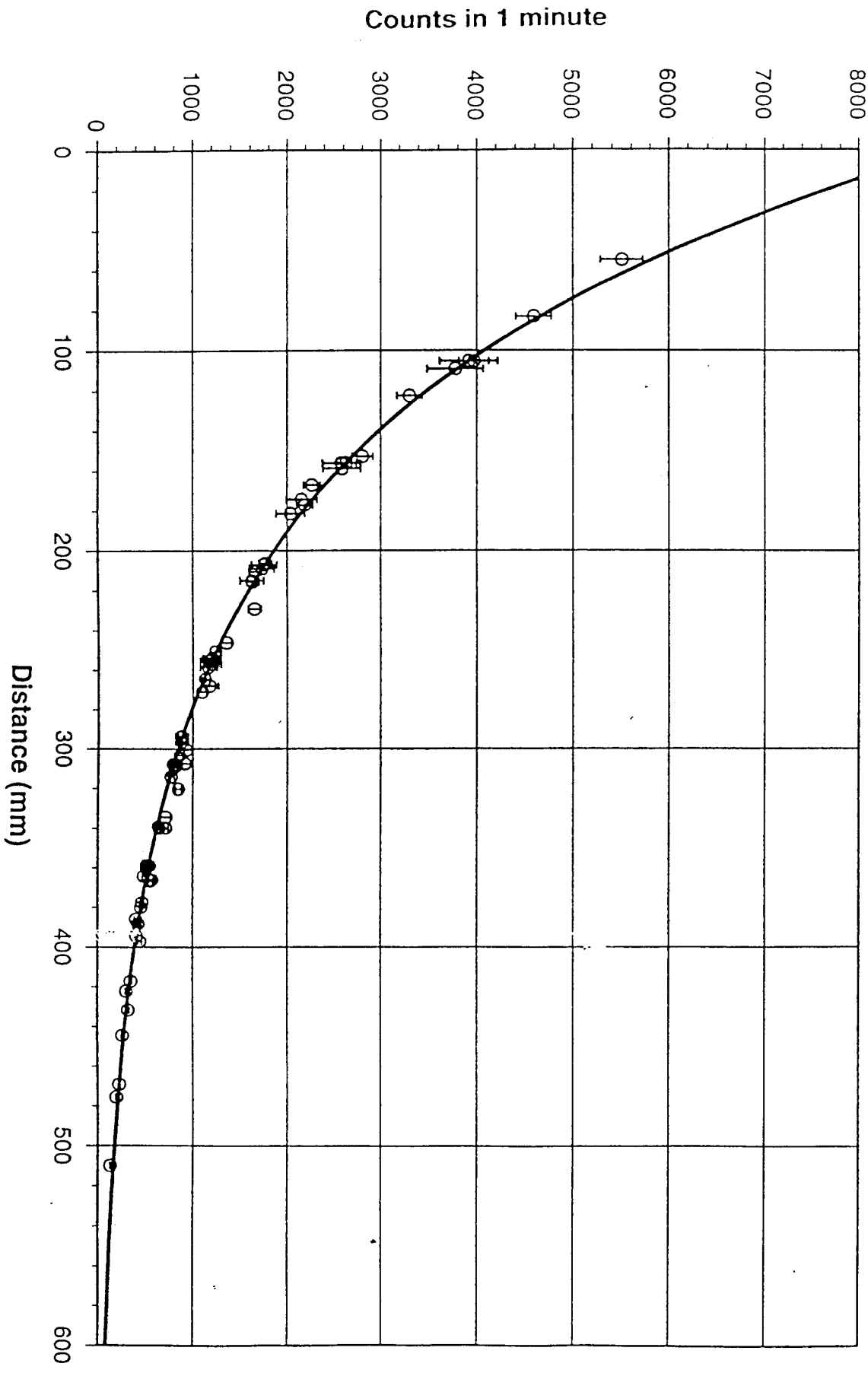

Counts in 1 minute
0
1000
2000
3000
4000
5000
6000
7000
8000
Distance (mm)
0
100
200
300
400
500
600

Figure 2

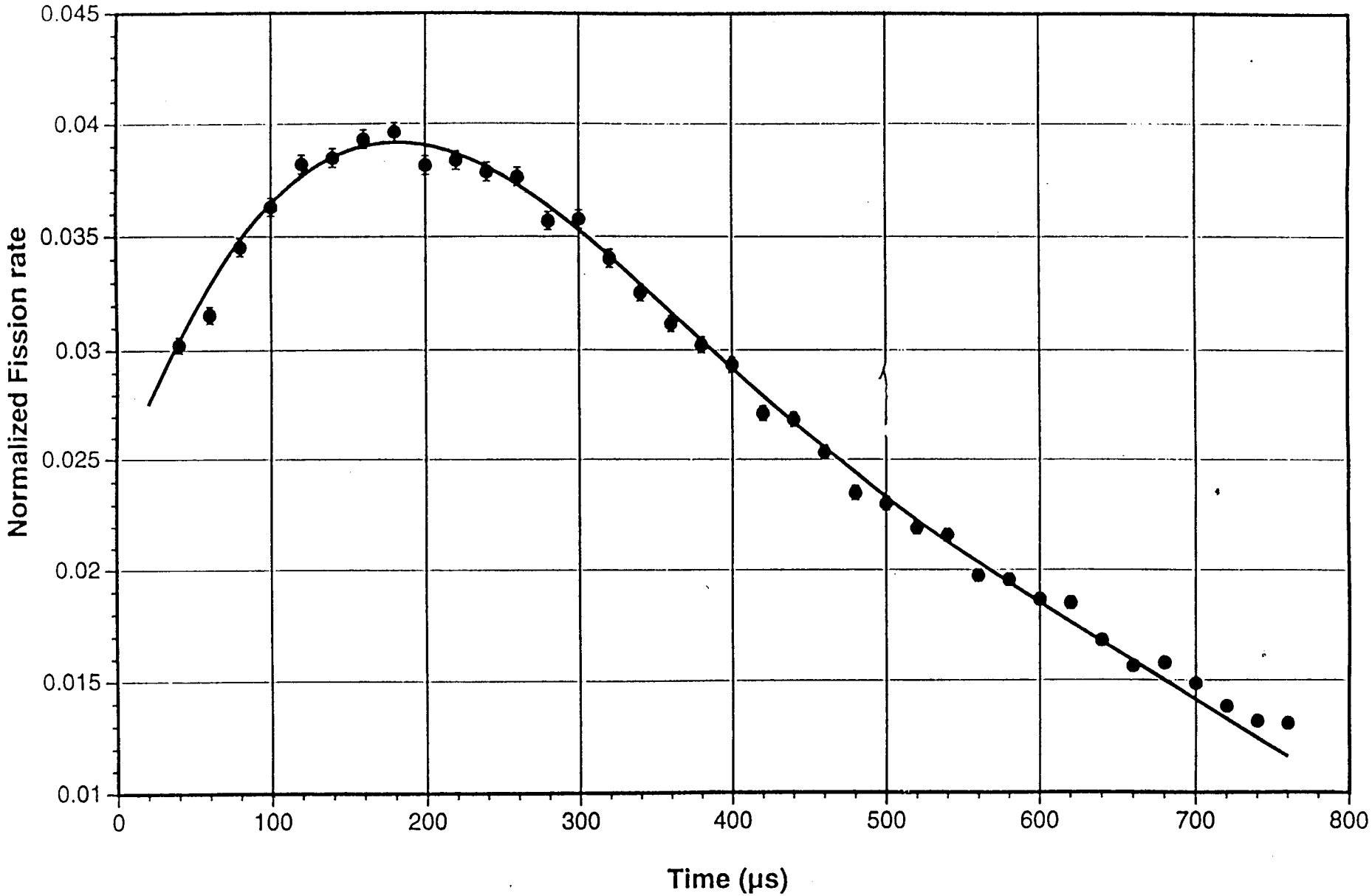

Normalized Fission rate
0.045
0.04
0.035
0.03
0.025
0.02
0.015
0.01
0
100
200
300
400
500
600
700
800
Time (μs)

Figure 3

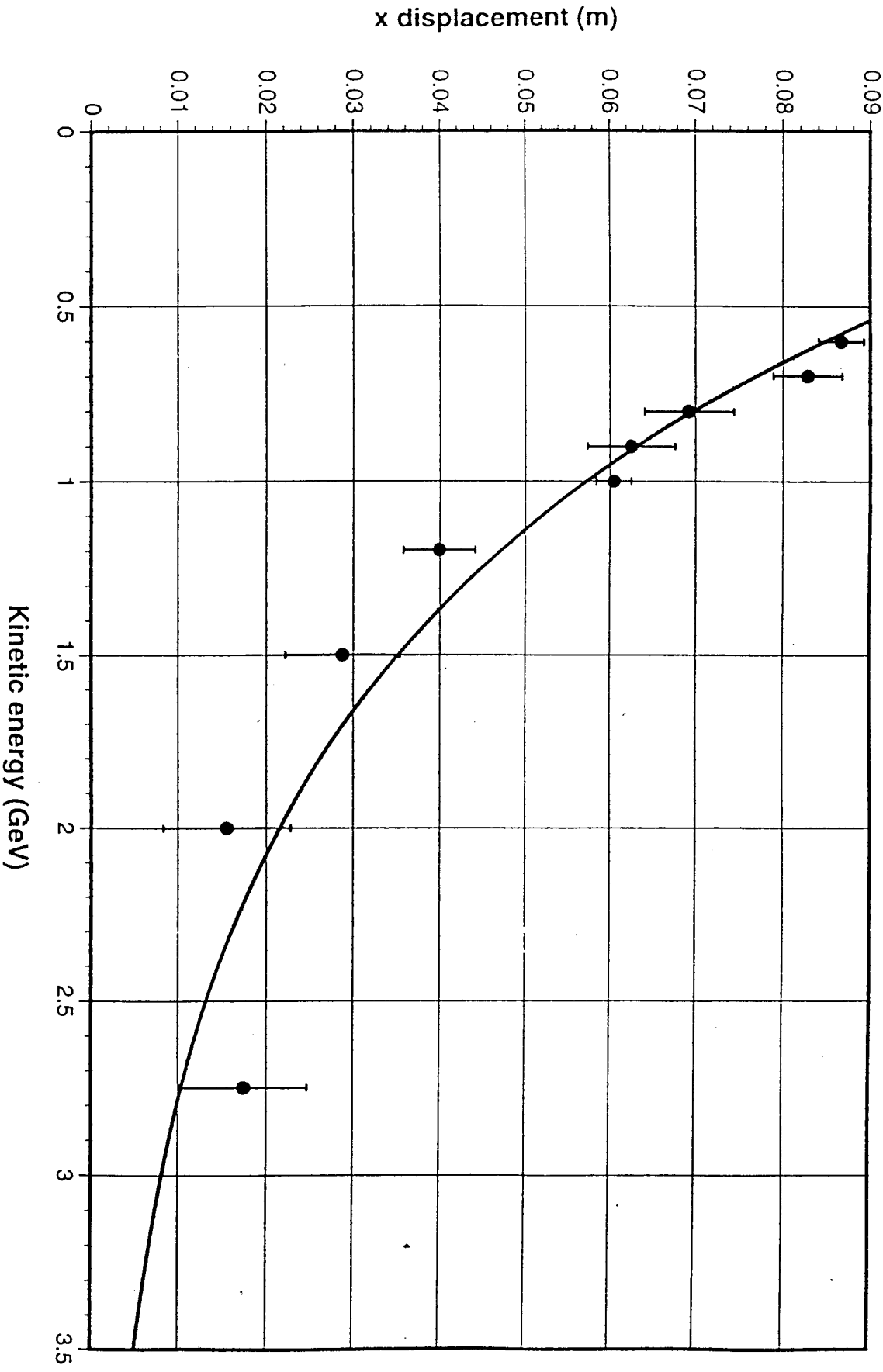

x displacement (m)
0
0.01
0.02
0.03
0.04
0.05
0.06
0.07
0.08
0.09
Kinetic energy (GeV)
0
0.5
1
1.5
2
2.5
3
3.5

Figure 4a

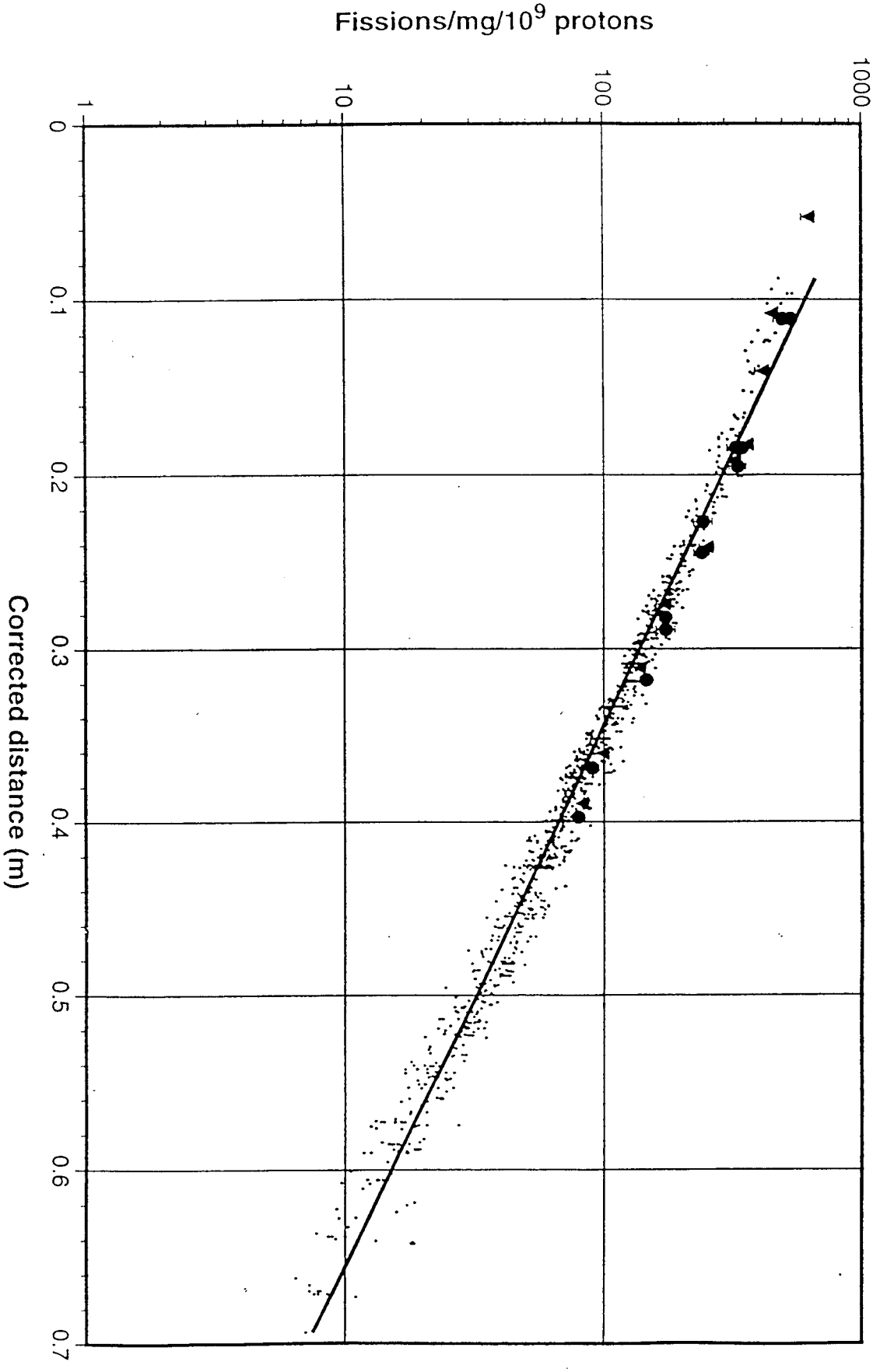

Fissions/mg/10^9 protons
1
10
100
1000
Corrected distance (m)
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7

Figure 4b

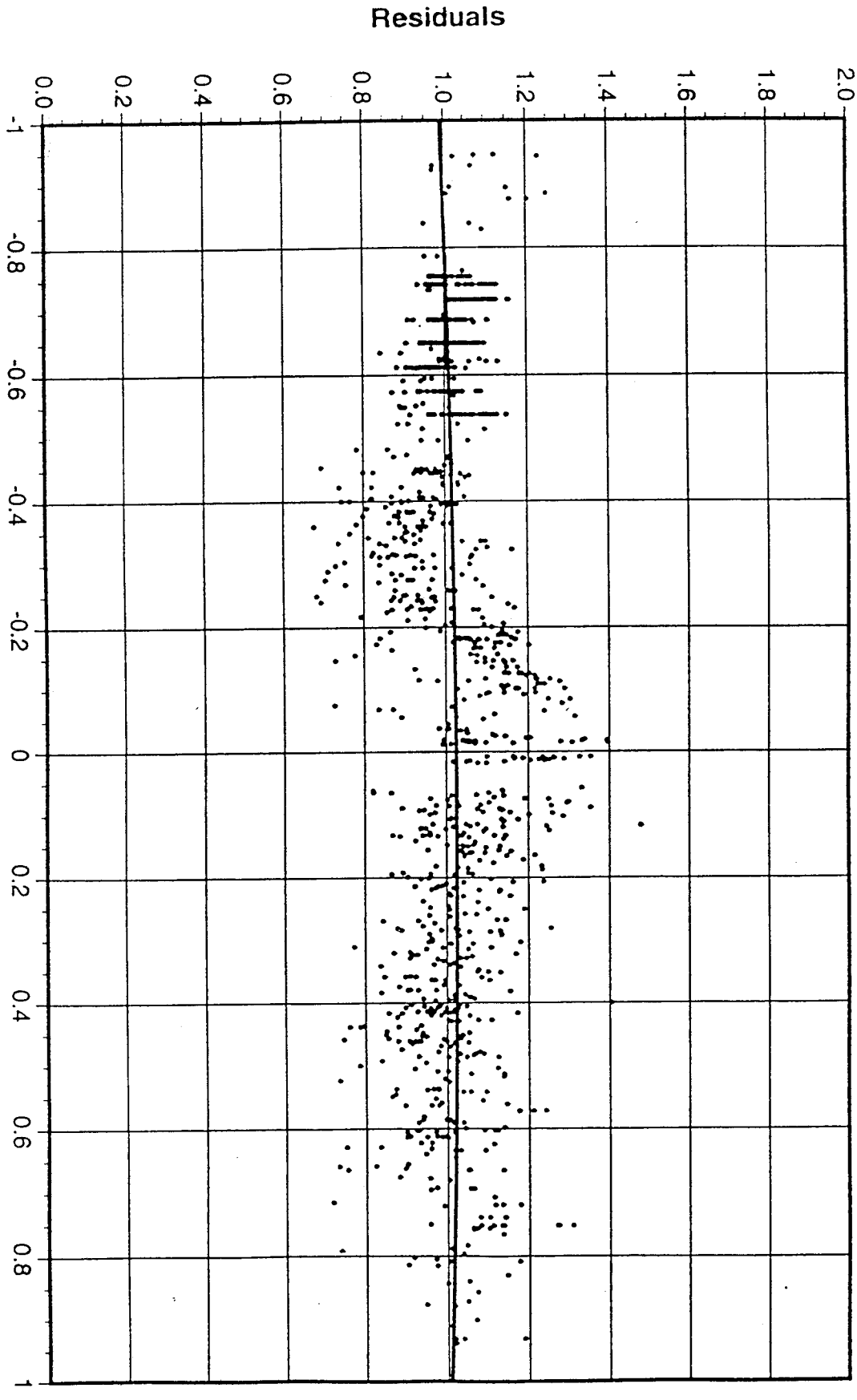

Residuals
2.0
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
-1
-0.8
-0.6
-0.4
-0.2
0
0.2
0.4
0.6
0.8
1
Cos(θ)

Figure 4c

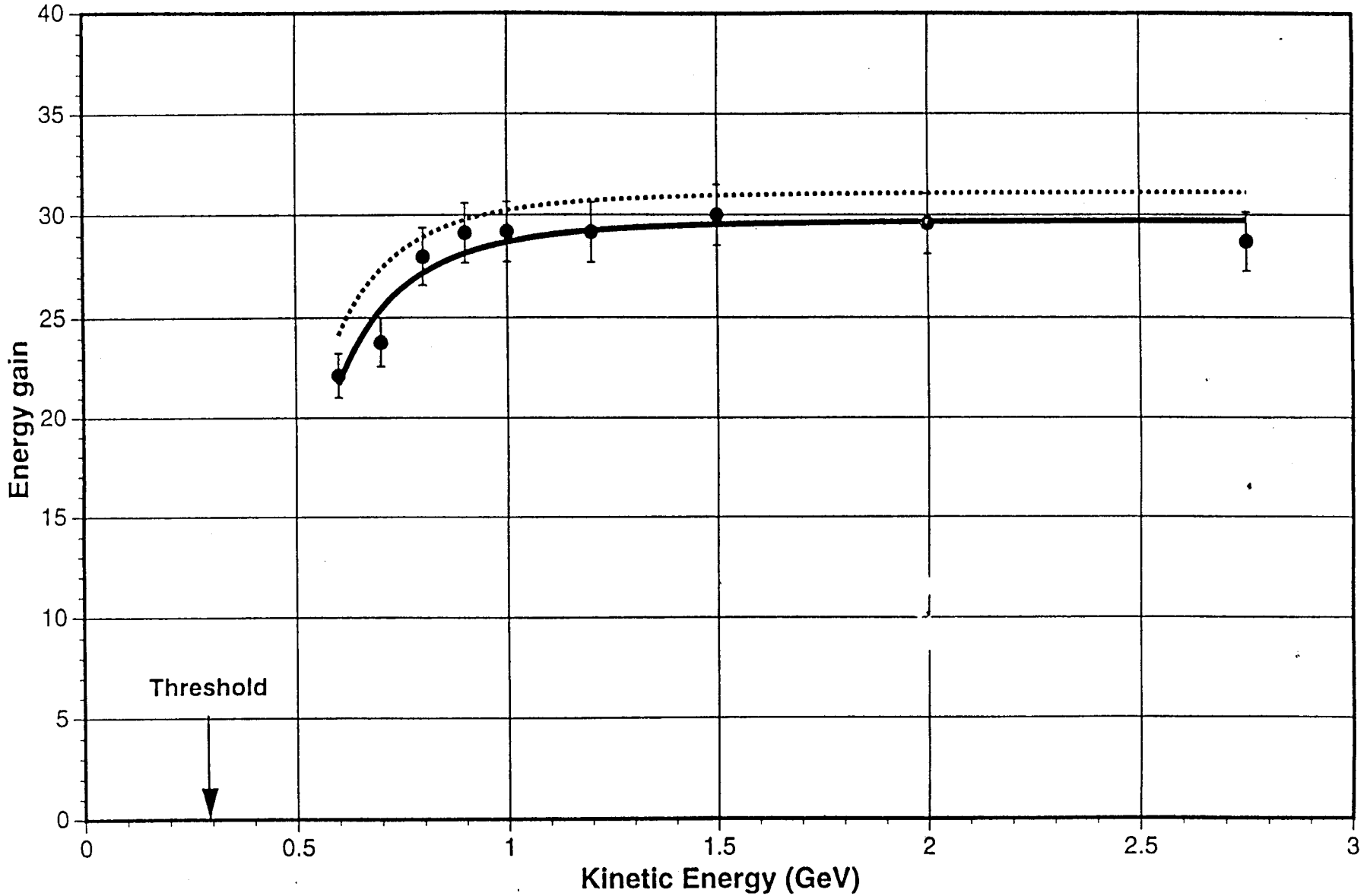

Energy gain
Kinetic Energy (GeV)
Threshold
0
5
10
15
20
25
30
35
40
0
0.5
1
1.5
2
2.5
3

Figure 5

Swedish Perspective on the Accelerator-Driven Nuclear System

Waclaw Gudowski and Henri Condé
Sweden

1. Introduction

The nuclear power programme of Sweden consists of 12 nuclear reactors (Tab. I) located at four different sites and with a combined capacity of 10 000 MW net electric power. The nuclear power plants generated about 42% of the total Swedish electric power produced in 1993. These nuclear power plants are owned by four companies which has formed the Swedish Nuclear Fuel and Waste Management Company, SKB (SKB - Svensk Kärnbränslehantering AB). SKB duty is to develop, plan, construct and operate facilities and systems for the management and disposal of spent nuclear fuel and radioactive wastes from the Swedish nuclear power plants. On the behalf of its owners SKB is responsible for all handling, transport and storage of the nuclear wastes outside of the nuclear power productions facilities (1). SKB is also in charge of the comprehensive research programme in the radwaste field.

Tab. I Swedish nuclear reactors

Reaktor name and type	Capacity MW(el)	Commerc. Operation	Licensed to
Barsebäck 1 (BWR)	600	1975	Indefinit.
Barsebäck 2 (BWR)	600	1977	2010
Oskarshamn 1 (BWR)	442	1972	Indefinit.
Oskarshamn 2 (BWR)	605	1975	Indefinit.
Oskarshamn 3 (BWR)	1160	1985	2010
Ringhals 1 (BWR)	750	1976	Indefinit.
Ringhals 2 (PWR)	800	1975	1995
Ringhals 3 (PWR)	915	1981	2010
Ringhals 4 (PWR)	915	1983	2010
Forsmark 1 (BWR)	970	1980	2010
Forsmark 2 (BWR)	970	1981	2010
Forsmark 3 (BWR)	1090	1985	2010

A complete system has been planned for the management of all radioactive residues from the 12 nuclear reactors and from the research facilities. The system is based on the projected generation of waste up to the year 2010. For spent fuel a central interim storage facility, CLAB, was taken into operation in July 1985. This facility has a current facility of 5 000 tonnes of spent fuel. The spent fuel will be stored in CLAB for about 40 years. It will then be encapsulated in a corrosion-resistant canister and deposited at depth in the Swedish bedrock. The construction of the deep repository will be made in steps. A first stage of the repository, for 5 - 10% of the fuel, is planned to be put into operation in 2008. The next stage for the full repository will only be built after a through evaluation of the experiences of the first stage and a renewed licensing. The site for the deep repository has not yet been chosen.

The estimated costs for the Swedish deep rock repository amounts to about 40 billion SEK. One attractive option to reduce these costs would be to transmute the most cumbersome components of the spent fuel using accelerator-driven transmutation technology.

2. Accelerators enter nuclear energy field

In recent years new concepts of treating spent fuel from fission reactors have been developed (2). This has initiated rapid growing international research activities in which a number of Swedish, mostly university-linked, groups participate. The technique is in general named accelerator-driven transmutation technology (ADTT) and is aiming to convert the long-lived radioactivity in the burned nuclear fuel, to short lived one and by that reducing the need for geological depositions. The accelerator-driven transmutation technology offers in the long-term a possibility to produce clean fission energy over an indefinite time, a concept which would fit Swedish needs and technological skill. However, due to the environmental requirements the Swedish parliament decided that the two dominating energy sources - the nuclear power and fossil burning - will be either shut down (nuclear power) or its use will be limited (fossil fuels), the Swedish energy supply in the long run is a problem area. The parliament decisions are based on the perceived risks which in the first case are linked to a possible release of radioactivity at a large reactor accident and/or the handling of the highly radioactive spent fuel and also to the proliferation concerns. In the second case - the risk for a global environmental catastrophe through the "green house effect". Although research of alternative renewable energy sources is in progress, no large scale solutions which can meet the future energy demands have come out so far. At the same time the research problems connected with the utilization of fusion energy are still numerous. Today it is difficult to predict when the basic problems in this field will be solved. Finally, import of energy effects negatively the balance of trade and makes Sweden depending on political and economical decisions in foreign countries.

With the twofold aim to find *methods for treating the high level nuclear wastes* which could be more easily accepted by the public than a direct geological deposition, and at the same time *recruit students to the nuclear energy field*, a national collaboration has been initiated on the research of accelerator driven transmutation technologies, particularly accelerator based nuclear waste transmutation systems (ADTW). The ambition to start research in this field was positively influenced by the Specialists' Meeting on "Accelerator-Driven Transmutation Technology for Radwaste and other Applications" which was held at Saltsjöbaden, Sweden on 24-28 June 1991.

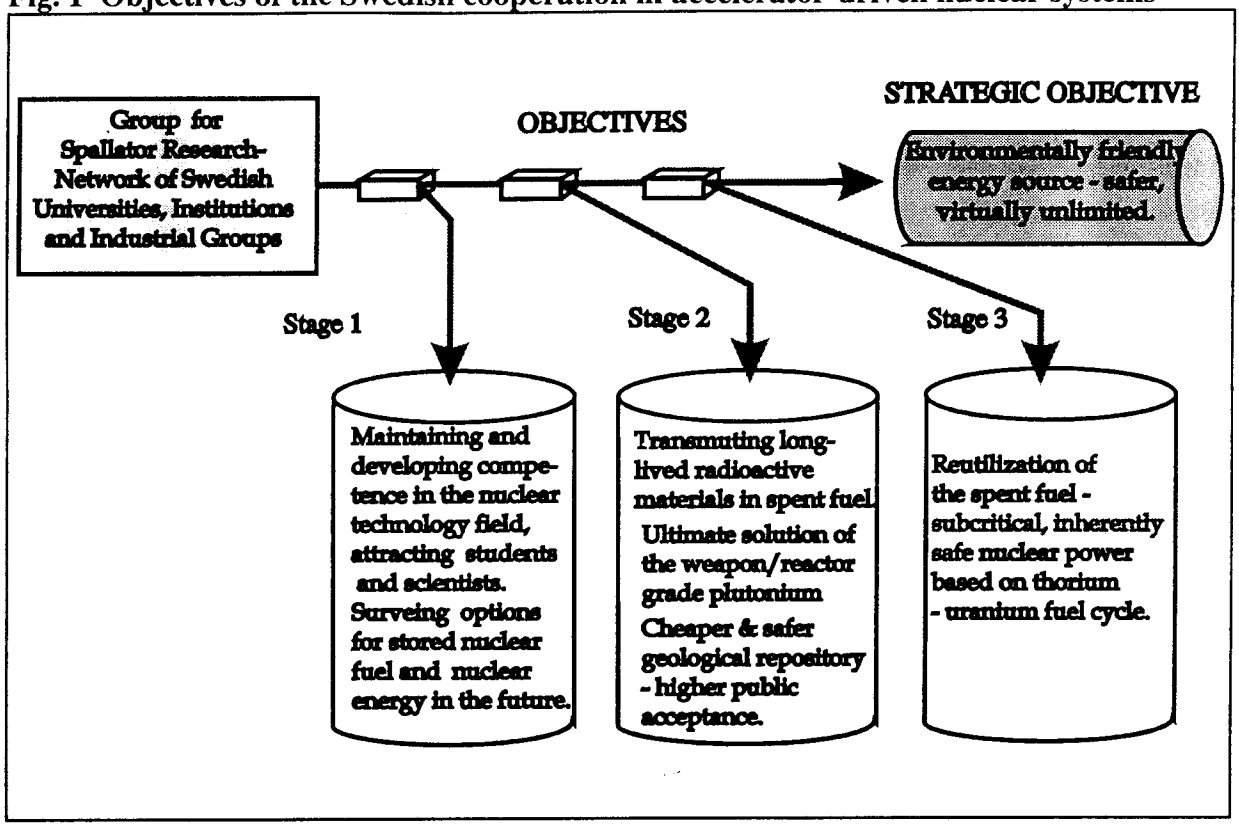
The "Group for Spallator Research" conducts - so far - the concerted research at CTH, KTH, the Manne Siegbahn Laboratory-Stockholm University and the Uppsala University.
The main task of this Group is to stimulate and to coordinate research and development projects in the accelerator driven transmutation technologies. These projects as - shown on Fig. 1 - are aimed to :

1) Practical solutions for accelerator driven transmutation of longlived radioactive material (e.g. plutonium, minor actinides, fission products) into shortlife or stable elements. It may result in cheaper and safer geological repository;

2) Investigation of new options for nuclear energy production with inherently safe systems, either with uranium or thorium fuel and with reduced longlived radioactive waste production. If successful, it will result in a new, environmental friendly, safe, cheap and virtually unlimited source of energy. The proposed systems for transmutation of spent fuel and production of energy are subcritical and inherently safe.

3) Opening new, exciting research and occupation possibilities for students and young specialists, which will ensure the proper level of competence needed for our nuclear power utilities, governmental agencies etc. The existing nuclear power facilities will, namely, need qualified personnel for at least two generations, even in the case of shutting-down of all the Swedish nuclear power plants by year 2010.

Fig. 1 Objectives of the Swedish cooperation in accelerator-driven nuclear systems



3. Research within a collaboration on accelerator based transmutation technologies in Sweden

3.1 Department of Nuclear Chemistry Chalmers University of Technology (CTH)

A research project was initiated at the Department of Nuclear Chemistry at CTH in 1976 to develop a process for separation of actinides from high level radioactive waste from the PUREX process. The project run for about 10 years time and resulted in the so called "CTH-process".

When the US and Japan separation and transmutation programs were presented around 1990 a new interest come into the field. This resulted in the creation of a new separation and transmutation project - SKB financed - at the Department of Nuclear Chemistry at CTH which started in 1991. Initially, a comprehensive survey was made of national and international activities in the field (3). Since 1993, the Department has started a new research project linked to the activities in other countries within the same research area.

The aim of the project is to study separation processes proposed for use in connection with nuclear transmutation. The project conducted in cooperation with research groups in US, Japan and EU contains both experimental investigations as well as modelling of different separation systems. The project is mostly directed towards fundamental research and aim to help judging the realism of different proposed separation and transmutation processes. In particular, the proposed separation systems by LANL and within the OMEGA project in Japan will be screened.

A very important issue in order to get a reasonable reduction of radionuclides through transmutation is a high separation efficiency. In turn, this means that one has to have a good control over what happens in the separation process and the chemistry must tolerate reasonable deviations in operating conditions. The separation process has to be optimized in this concept which requires separation data for a number of different elements and a sensitive analysing technique. The most important elements are the actinides, which are long-lived and highly radiotoxic. The development of analysis techniques for the actinides is, for that reason, an important research area.

The department operates the only large α-box laboratory in Sweden and has facilities to handle substantial activities of β,γ emitting nuclides in shielded cells with master-slave manipulators.

3.2 Department of Neutron and Reactor Physics and Centre for Safety Research, Royal Institute of Technology (KTH)

Alternative nuclear fuel handling has been discussed within a working-group at KTH through a number of years. The group consists of about 25 experts of different disciplines as reactor physics, nuclear chemistry, physics, material sciences, hydrology, geology and risk analysis. The group has worked actively to broaden the interest for transmutation after the Conference in Saltsjöbaden in 1991 and stimulated creation of the small research team fully devoted to study accelerator driven systems (4). The research program conducted by this team is focused on conceptual studies based on computer simulations (computer codes: MCNP, SYSDANT, ORIGEN and CASMO/SIMULATE) aimed to find the answer for some important questions like:

1. Impact of the ADTT on the costs and performance of the geological repository

2. Safety analysis of transmutation systems
    a) Criticality margins and criticality budget
    b) Minimizing the amount of actinides in the system
    c) Neutronics for the ADTW/ADEP systems

3. Safety and system analysis of the synergetic system: nuclear reactors and accelerator-driven waste transmutation system

Very extensive investigations - in collaboration with so called Swedish Industrial Group - were performed on the system proposed by Carminati et al. (5) because of its potential attractiveness: thorium breeding cycle and light water moderation. The conclusions, however, pointed at some very difficult problems with this idea:

1. The reactivity budget analysis of such system shows that it is virtually impossible to construct this device without external reactivity control systems. The idea of the subcritical system with k_{eff} inherently lower than 1 seems to be irreconcilable with the physics of this system and economical requirements

2. Solid fuel and light water moderator require multiple target due to the power density peaked around the targets. It makes the construction much more difficult - if not impossible - and more expensive (multiple cyclotrones, target window problems etc.).

3. To make the construction of such system economically possible it seems to be necessary to reevaluate the requirements of the inherent subcriticality. Instead, better safety mechanism could be invented (or re-invented) and applied to ensure safe operation. Very high k_{eff} of the level of .99 should be reconsider if one wants to proceed with light water and the solid fuel (6).

The research of the group at KTH is performed in very active and close cooperation with the Los Alamos National Laboratory. The common project in calculation of the neutronics of the accelerator-driven plutonium burner and thorium-based energy producer is now expanding into Russia and a common experimental programme becomes realistic.

3.3 Department of Neutron Research and Department of Radiation Science, Uppsala University

Basic and applied experimental neutron and nuclear research at the Uppsala University is centred around two laboratories administrated by the University namely the The Svedberg Laboratory (TSL), Uppsala and the Neutron Research Laboratory (NFL), Studsvik.

The main facilities of TSL is a cyclotron, a storage ring (CELSIUS) on-line the cyclotron and a tandem van de Graff-accelerator. The cyclotron can operate in an isochronous mode allowing acceleration of protons up to 100 MeV and heavy ions to 196 Q^2/A MeV and in a synchrocyclotron mode which allows acceleration of protons to 180 MeV. The storage ring CELSIUS is equipped with acceleration and electron cooling capacities. It takes both light and heavy ion beams. The maximum proton energy is 1.36 GeV. The tandem is a High Voltage 6 MV EN-tandem mostly used for applied research as C-14 dating, PIXE, ion-surface studies etc.

The NFL is located at the research reactors R2 and R2-0 at Studsvik. The main facility aside of the neutron diffractometers at the R2 reactor is an isotope separator on-line the R2-0 reactor (OSIRIS) for studies of nuclear structure and decay data for fission products and fission yields.

Measurements in progress or planned by different Uppsala groups of relevance for the research on the ATW concept at the TSL and NFL facilities are:

A comprehensive study has been carried out of the yield pattern of fission products formed in the thermal neutron induced fission of U-235 using the OSIRIS facility at NFL. Independent and/or cumulative yields have been obtained for 195 nuclear species, among them 83 isomeric states. Similar studies of the yield pattern have been carried out for the fast fission of U-238, is in progress for the thermal fission of U-233 and is planned for the fast fission of Th-232. At the same laboratory, continuous spectra of beta particles and gamma rays emitted in the decay of short lived fission products have also been measured. The mass range 79-98 and 130-147 was covered by a measurement at OSIRIS and 98-108 by a measurement at the on-line separator LOHENGRIN at ILL. Accurate average beta and gamma energies, obtained from the spectra, are essential input data for summation calculations of the heat developed in nuclear fuel by the decaying fission products.

Cross section measurements for residual nuclide production by proton and neutron induced reactions relevant for Accelerator Driven Transmutation Technologies are in progress within a collaboration between the Universities in Hannover and Cologne, KFA Jülich, ETH Honggerberg, Kossuth University Debrecen, University of Bourdeaux-Gradignan and Uppsala University. It is the aim to measure at TSL thin-target cross sections for the production of residual nuclei from about twenty medium and heavy target elements by proton-induced reactions for energies between 70 and 180 MeV. Radioactive and stable residual nuclei will be measured via gamma-spectrometry ($T_{1/2}$> 15 h) counting techniques (H-3), accelerator mass spectrometry (B-10, Al-26, Cl-36, Ca-41, Mn-53 and I-129) and conventional mass spectrometry (stable rare gas isotopes). The results will be used to test nuclear reaction models of spallation, fragmentation and medium energy fission. Furthermore, feasibility studies are underway of activation experiments with fast neutrons up to 100 MeV.

Model calculations in the intermediate energy region are tested against measurements at TSL of double differential cross section measurements at 100 MeV for (n,p)-reactions in C-12, Fe-54,56, Zr-90 and Pb-208 and of the n-p differential scattering cross section. A reasonable agreement was obtained between the experimental (n,p)-reaction data and the same data from DWA/RPA calculations adding contributions from multistep reactions. On the other hand a disagreement of about 5-10 % was observed between the experimental n-p scattering data at about 180 degrees and recent phase shift calculations. Accurate n-p scattering measurements over a wide angle range is underway to further explore the observed discrepancy.

Measurements of neutron induced fission cross sections for Bi-209 and U-238 have been performed for 100, 130 and 160 MeV at the neutron beam facility of the TSL in a collaboration between the Khlopin Radium Institute, S:t Petersburg and the Uppsala University. The fission fragments were detected by thin film break down counters. The neutron flux was measured relative to the n-p scattering cross section. A reasonable agreement was obtained with calculations using the LAHET code.

Research and development of measuring procedures and instrumentations for isotopic analysis of burned nuclear fuels are underway.
The Uppsala group is also participating in an experiment at SATURNE, Saclay to study the neutron production in thin targets of several elements (Pb, Bi, W, Fe etc) between 0.6 and 2 GeV.

The study is a collaboration between Centre d'Etudes de Bruyeres-le-Chatel, Laboratoire National SATURNE, I.P.N. Orsay, Centre d'Etudes de Saclay, College de France Paris and the Uppsala University.

3.4 Manne Siegbahn Laboratory (MSL) - Stockholm University

The Manne Siegbahn Laboratory has a long tradition in accelerator design and operation. The present main accelerator facility at MSL is a 52-m circumference synchrotron storage ring called CRYRING [4]. It includes both an injection line for light ions, the MINIS separator, and a line for highly-charged heavy-ions, the INIS- CRYSIS electron-beam ion-source system. Intermediate acceleration is provided by a radiofrequency quadrupole structure, RFQ, and the quality of stored ions in the ring is improved by an electron cooler.

Within the collaboration the group at MSL has proposed studies of critical issues connected to the construction of high current accelerators. The issues involve studies of space-charge current-limitation in the low energy part of the accelerator and a minimization of the particle losses due to rest gas collisions in the vacuum system. Furthermore, the injection lines feeding CRYRING allow and can hence be used to investigate how to merge two beams into the RFQ as has been proposed for the high current ATW-facility at Los Alamos. Another research area of concern for the construction of a high power accelerator is ultra high vacuum technology. Experienced scientists in this area are available at both MSL and TSL.

MSL has the competence and experimental resources to measure nuclear reaction data of importance for the spallation concepts. A small van de Graff accelerator (2 MeV) is available near the CRYRING facility for this purpose.

3.5 The Industrial Group

A more industrially oriented group of reactor physicists, formerly at ABB Atom, and neutron physicists from Uppsala University and KTH have investigated the technical and economical possibilities to implement a nuclear reactor construction based on the concept which recently has been presented by C. Rubbia et al (5). As the first goal, this group of reactor and neutron physicists has investigated the technical and economical possibilities to realize a nuclear energy production facility based on **the light-water spallator driven by a smaller scale accelerator**. The strategy chosen by the group was to start with a LWR-construction and to marginally modify the construction, with state-of-the-art technology, and if necessary supplement it with an autogenous rescue cooling system of the PIUS-type or similar.

4. International perspective

The research activities of the Group for Spallator Research has been primarily devoted to system and feasibility studies together with participating in the number of international efforts mainly in US (Los Alamos), Russia (Troitsk), France (Saturne) and in future possibly Japan (PNC-JAERI), CERN and Switzerland (PSI).
Collaborative projects are already in progress with leading international laboratories on ADTT research as described above. In particular, a test experiment of burning plutonium from the present nuclear power plants but also the weapon grade plutonium from warheads of destructed nuclear weapons at the linear accelerator of the Institute for Nuclear Research at Troitsk outside Moscow has been proposed as a collaboration between U.S.A., Russia and Sweden. It is anticipated that the Swedish groups (coordinated and financed through the Centre for Spallator Research) - in similarity to e.g. CERN collaboration - will prepare some part of the experimental equipment and take responsibility for its installation and performance.

The next international conference on Accelerator-Driven Transmutation Technologies and Applications 1996 will be arranged in Sweden, 3 - 7 June in Kalmar.

Group for Spallator Research is also going to work actively to create an international organisation or centre for advancement of ADTT.

5. Final remarks

Accelerator-driven nuclear systems can become an important complement for nuclear reactors opening new options for the nuclear fuel cycle and furthermore, in the countries like Sweden, where the future of the conventional nuclear power has no prospects, these systems can make the nuclear energy again an attractive source of environmental friendly energy. Also the idea of burning weapon grade plutonium in accelerator driven systems has a lot of advantages and should be thoroughly exploited. The best way to achieve these goals is an intensive international cooperation and the common efforts to build the first demonstration facility.

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Transmutation of Fission Products through Accelerator

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Abstract

The transmutation of fission products through particle accelerators has been studied under the OMEGA program. The photonuclear reaction has also been investigated to be applied to transmuting long-lived fission products, such as Cesium and Strontium, which have difficulties on reaction with neutrons due to its so small cross section. It is applicable for the transmutation if the energy balance can be improved with a monochromatic gamma rays in the range of the Giant Dipole Resonance generated through an excellent high current electron linear accelerator. The feasibility studies are being conducted on the transmutation system using it through an electron accelerator.

1. Introduction

The high level radioactive waste (HLW) generated from the reprocessing of spent fuel is one of issues in the backend of nuclear fuel cycle. In Japan, a present national policy for the HLW is to vitrify and dispose it in a deep geological repository after 30-to-50-year storage for cooling down the decay heat. The researches and developments have been devoted to the technologies for waste disposal and the methodologies for safety assessment. In parallel, the long-term R&D program, "OMEGA," on nuclides for partitioning and transmutation was initiated in 1988 by the Japanese Atomic Energy Commission in order to explore the new way utilizing the HLW as useful resources and reduce the burden to the geological disposal. The program led by the Science and Technology Agency has been conducted with the collaboration of three major research organizations; the Japan Atomic Energy Research Institute, the Power Reactor and Nuclear Fuel Development Corporation(PNC) and the Central Research Institute of Electric Power Industry.

The program is composed of major R&D, as shown in Fig.1. The nuclides are partitioned from HLW according to half-life and potential value for utilization. The minor actinides (MA; ^{237}Np, ^{241}Am, ^{243}Am, ^{244}Cm, etc.) and long-lived fission products (FP; ^{90}Sr, ^{99}Tc, ^{129}I, ^{137}Cs, etc.) are transmuted into short-lived or stable nuclides.

2. Transmutation of Fission Products

The methods of transmutation of the long-lived nuclides in the HLW are roughly classified into those using neutron and those using protons or photons. The neutron methods are available to transmute actinides having comparatively large fission cross sections for fast and thermal neutrons. The fission reactors are expected as neutron sources for the transmutation of long-lived minor actinides. The studies in OMEGA program have indicated that the fast reactors have an advantage over the thermal reactors in transmuting actinides efficiently and in generating less secondary waste. Thus, the concept of utilization of the electric power plants of fast reactors has been developed into the acitinide recycle as an advanced nuclear fuel cycle.

2.1 Methodologies of Transmutation of Fission Products

For the transmutation of fission products, Tc and I, having comparatively large cross sections for thermal neutron, the neutron methods could be conceptually applied to transmutation by the thermal or fast reactor if the core was modified to have a specified region. However, Cs and Sr as typical fission products having so small cross sections for thermal and fast neutrons, demands extremely high neutron fluxes for transmutation up to 10^{17} to 10^{18} n/cm^2s in fission reactors. It may be hard to obtain this high neutron fluxes at the present days. The fission reactors are limited to use in the transmutation of fission products. Thus, as another alternative of incineration, the utilization of high energy particles through accelerators has been considered, referring the recent technological advances and backgrounds of accelerators.

2. 2 Alternatives for Transmutation of Fission Products

The feasibility of the transmutation by the particle accelerators using spallation and spallation-neutron and photonuclear reaction has been evaluated, focusing on the energy balance in the transmutation.

Transmutation of Tc and Cs as target nuclides are referenced in this study. Tc as representative of nuclides has large cross sections for thermal neutron. Therefore the spallation-neutron method and the muon catalyzed fusion method are relatively effective for the transmutation, as shown in Table 1.

As for Cs, the spallation-neutron method is relatively potential to the transmutation with high flux neutrons supplied through the spallation reaction. In the method, the high energy protons impinge on heavy metal such as lead placed in the center of the target. The neutrons generated by spallation reaction are thermalized and supplied to the transmuting reaction (n,γ) of fission products. The muon-catalyzed fusion method is effective for transmutation of Cs. In this method ^{137}Cs is transmuted into ^{136}Ba through the (n,2n) reaction of neutrons generated by the muon and D-T fusion in the target. These methods are relatively more advantageous than the proton spallation method and photonuclear reaction method, as shown in Table 1.

The evaluation of these methods indicates that the spallation-neutron method is relatively feasible. Therefore a hybrid system as accelerator-driven reactor can be a candidate of the transmutation of fission products. However more technology developments are required for the system to be used for the transmutation of fission products, from the point of energy balance. The muon-catalyzed fusion is still future technology. Therefore it is needed to research other alternatives.

In this context, the photonuclear reaction method would be one of alternatives, with the advantage of its simpleness of transmutation, less secondary-waste production, and the experienced accelerator technology.

But it still has the energy balance problem. In order to improve the energy balance, the transmutation by the photonuclear reaction is re-evaluated through monochromatic γ rays generated from a high power electron linear accelerator.

3. Photonuclear Reaction for Transmutation

The photonuclear reaction is caused by γ rays: a nuclide is radiated and excited resonantly with γ rays of 10 - 20 MeV in the range of the Giant Dipole Resonance, and then a neutron is released from a nuclei and the nuclei is transmuted. The photonuclear reaction has uniform cross sections for the actinides and fission products. The cross sections of Sr and Cs have the threshold at around 8 MeV and the maximum peak at 300 or 200 mb as shown in Fig. 2. There exist competitive reactions as well as photonuclear reaction; the pair electron creation, photoelectric effect and the Compton effect. Among them, the largest is the pair electron creation that has cross section of around 8000 mb. With competitive reactions, the photonuclear reaction is expected to be caused by 3 to 5% of monochromatic γ rays in the target. In the reaction, a nuclei of ^{137}Cs is transmuted by the monochromatic γ rays around 15 MeV and the transmutation requires 400 MeV energy, as shown in Fig. 3. On the contrary, with the Bremsstrahlung γ ray it requires up to 4700 MeV energy. Therefore the photonuclear reaction method can be improved, if monochromatic γ ray is provided to the target. It is assumed that the criteria of the energy balance for transmutation is 10% of the usable energy that is 1100MeV with 200 MeV released energy through fission reaction, with 6% of fission yield of ^{137}Cs, and with 33% of the conversion of thermal power to electricity.

3.1 Accelerator for Photonuclear Reaction

In the large amount of transmutation by the photonuclear reaction, well-qualified and high-current electron beam is required. For example, with the transmutation of 40 kg of ^{137}Cs produced in a 1000 MWe LWR in a year, a few amperes of beam current and beam energy around some hundreds of MeV will be needed, being taken into the efficiency of conversion from electron beam to γ rays in the range of 10 to 20 MeV. This requires to develop an accelerator for the high quality electron beam with the energy range of 100 - 1GeV and high current. However, as shown in Fig. 4, the electron linacs in the world have been so far developed in order to elevate the beam energy for the high energy particle physics, and even in the medium beam energy, there is little experience for developing high current accelerator. As for the high current beam, it has to be well controlled to avoid the beam-break-up phenomenon which causes damage to accelerator guides. The heat removal from the tubes and other components has to be deliberately designed to avoid the thermal structural deformation disturbing beam stability. To get monochromatic γ rays with high intensity, the conversion of the electron beam into monochromatic γ rays, such as the laser Compton scattering et. al., is the key technology. Therefore it is needed to research these technical feasibilities by using an experimental accelerator.

3.2 The Research on High Current Electron Accelerator

It is technically and also financially quite difficult to achieve some hundreds MeV energy and the current of a few amperes at one step. Thus, the experimental high power electron linear accelerator, with 10 MeV beam energy and the maximum / average current of 100mA / 20mA is under development. The main specification and the basic structure of the accelerator are shown in Table 2 and Fig. 5, respectively.

The experimental accelerator basically consists of electron gun, chopper, pre-buncher,buncher,accelerator guides with traveling wave resonant rings and beam dump. It is designed to facilitate the study of key technologies and high intensity beam. The beam is delivered with a current of 100 mA, 4 mA pulse and 50 Hz pulse repetition. The accelerator will be completed in March 1997 but an injector experiment is planned to be conducted in 1995.

The components of the accelerator are deliberately designed through experiments and analyses. The injector system is designed to consist of a 200kv DC gun, magnetic lens, a RF chopper, chopper slits, a prebuncher,and a buncher. The electric gun is designed to be available on a pulse-to-pulse repetition rates from single pulse to 50 pulses/sec. In the chopper consisted of a RF chopping cavity and a slit, three magnetic fields are mixed together for adjusting RF field amplitude and phase, and the chopped beam can be led to the beam center line. Bunching in the injector is conducted by the prebuncher. The traveling-wave resonant ring accelerates or decelerates electrons depending on their phase with respect to the buncher RF field.

The accelerator tubes are characterized by the traveling-wave-ring and are excited with microwave power at a frequency of 1.25 GHz. The accelerating tube has a cylindrical, disk-loaded shape made by oxygen free high-purity copper. The structures maintain a constant axial electric field over its length. The length of accelerating section is 1.2 m with 13 of cavities and two coupling cavities. In the high power linac operation, considerable amount of heat is expected to be generated in accelerator structure. The dimension of the accelerating section is determined by the analyses of heat transfer and thermal stress.

A beam dump of the high power and low energy beam(200kw of 10 MeV electron beam) is a challenging technology, in which electrons with 10MeV energy, concentrates within a range of only a few centimeters. The energy deposition causes high thermal power densities in a beam dump. The klystron used in the accelerator were developed specially to operate in continuous waves and pulse with good efficiency(over 60%). The output windows were designed and tested with pill-box type windows. Based on the test with collaboration of KEK, it has been verified that the klystron can generate more than 1.2MW RF.

3.3 The Studies on the Transmutation System

The studies have been conducted to conceptualize a system for transmuting waste targets with design principles of simpleness, reliability and safety. The targets, solid or liquid, and its effective cooling are being investigated. The handling of the target with high radioactivity has to be easy and safe in the operation with the remote system. The estimation of transmutation shows, as shown in Fig. 6, that Sr or Cs targets can be incinerated during 300 days and the created nuclides can be decayed by 400 days' cooling. In this estimation, the long-lived nuclides would be transmuted in short time with less residual waste produced in the target. This means that the photonuclear method is simple in the transmutation. The system should be co-located with the reprocessing plant in order to reduce the risk with transmutation. For continuous and long duration of operation in transmutation, the accelerator as gamma ray source has to be reliable. Thus, the electron accelerator may be suitable for transmutation of fission products due to its simpleness in operation and maintenance.

4. Conclusion

The R&D activities for the transmutation of fission products are in progress in PNC under the OMEGA program. The transmutation methodology is evaluated on the energy balance through the comparison of neutron, proton and photon methods. It is indicated that neutron methods including spallation-neutron method would be available for the transmutation of fission products having large cross sections for neutron such as Tc, and that the photonuclear method would be advantageous for the transmutation of Cs and Sr having so small cross sections for neutron. More studies and experiments are required to evaluate the feasibilities on the transmutation system.

It should be reminded that the OMEGA program is not only intended to seek short-term alternatives for present back-end strategies for nuclear energy system, but also to pursue benefits for future generations through long-term basic R&Ds.

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Table 1 The Comparison of the Energy for Transmutation with Methods

(MeV)

Methods	Cs-137	Tc-99
Spallation(Proton)	570	330
Spallation-Neutron (Proton)	510	35
Muon Catalyzed Fusion	195	40
Photonuclear Reaction (Bremsstrahlung)	4700	-
Photonuclear Reaction (Monochromatic)	400	-

Table 2 Main Specification of PNC Electron LINAC

Max. Beam Energy	10MeV
Max. / Ave. Current	100mA / 20mA
Pulse Length	4ms
Beam Repitition	50Hz
Duty	20%
Average Beam Power	200kW
RF Frequency	1.249135GHz
Length of Microwave	24cm
Mode of Accerelation	$2\pi / 3$
Klystron Power	1.2MW
Total Length of Linac	16m

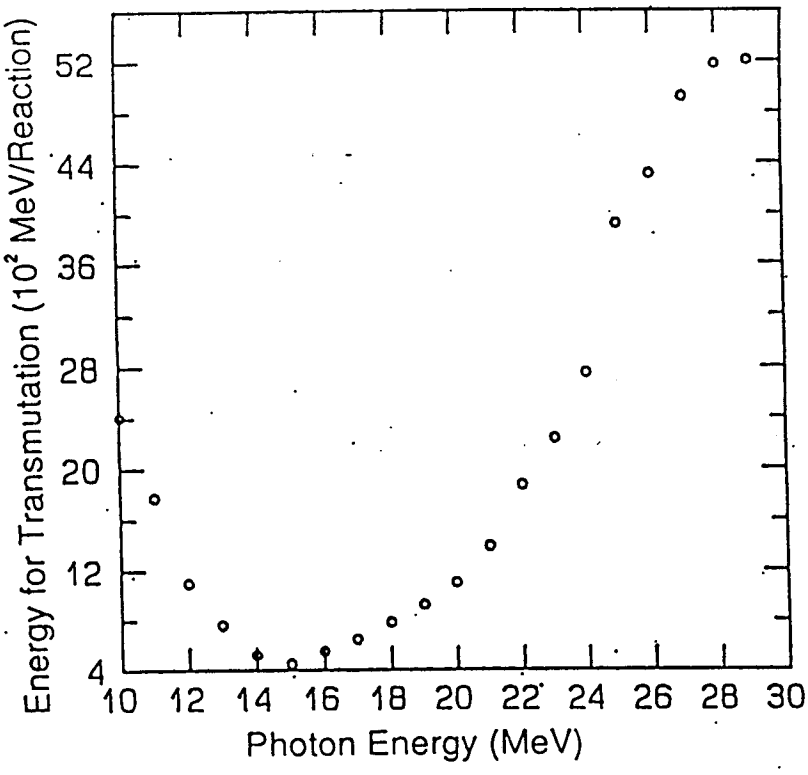


Fig. 3 The Energy for Transmutation of Cs-137 with the Monochromatic γ ray

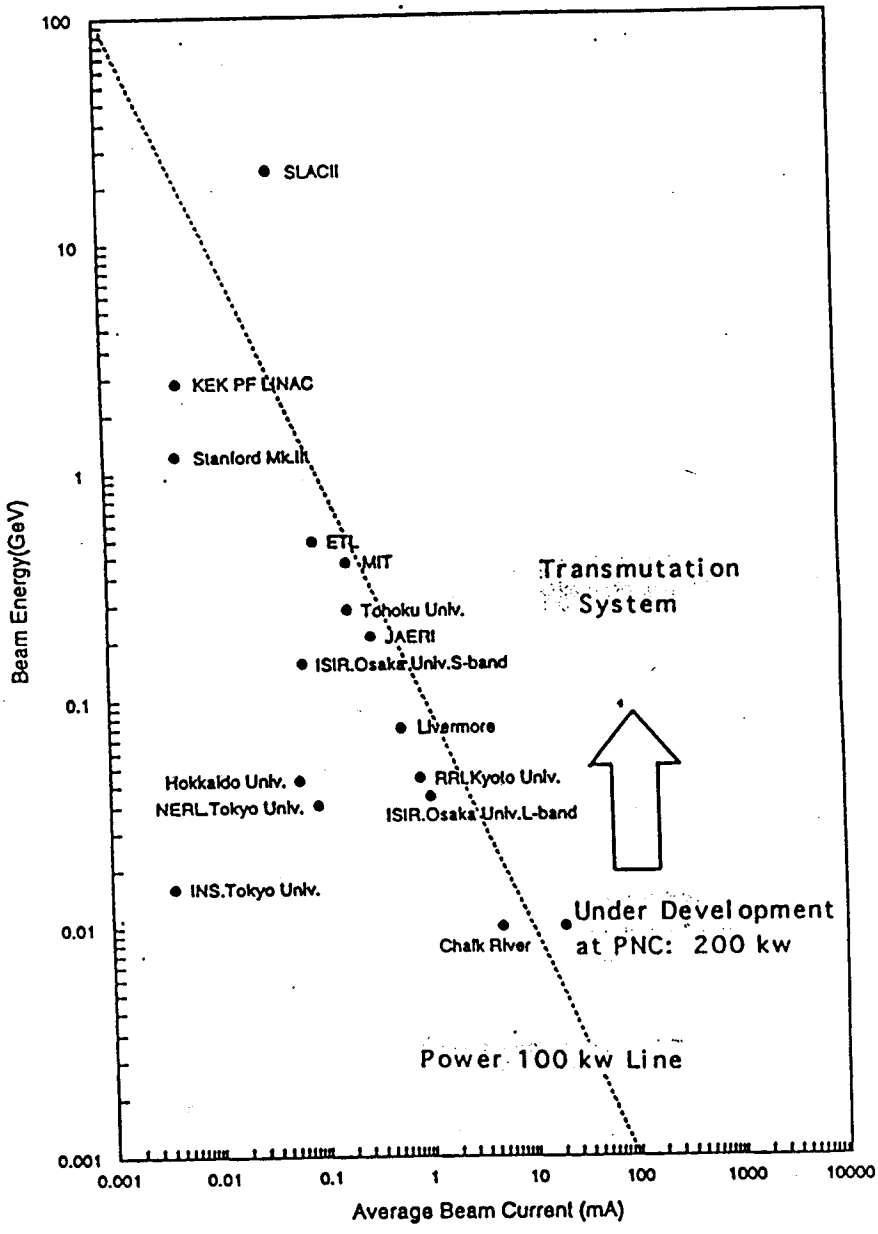


Fig. 4 Electron Linear Accelerator in the World

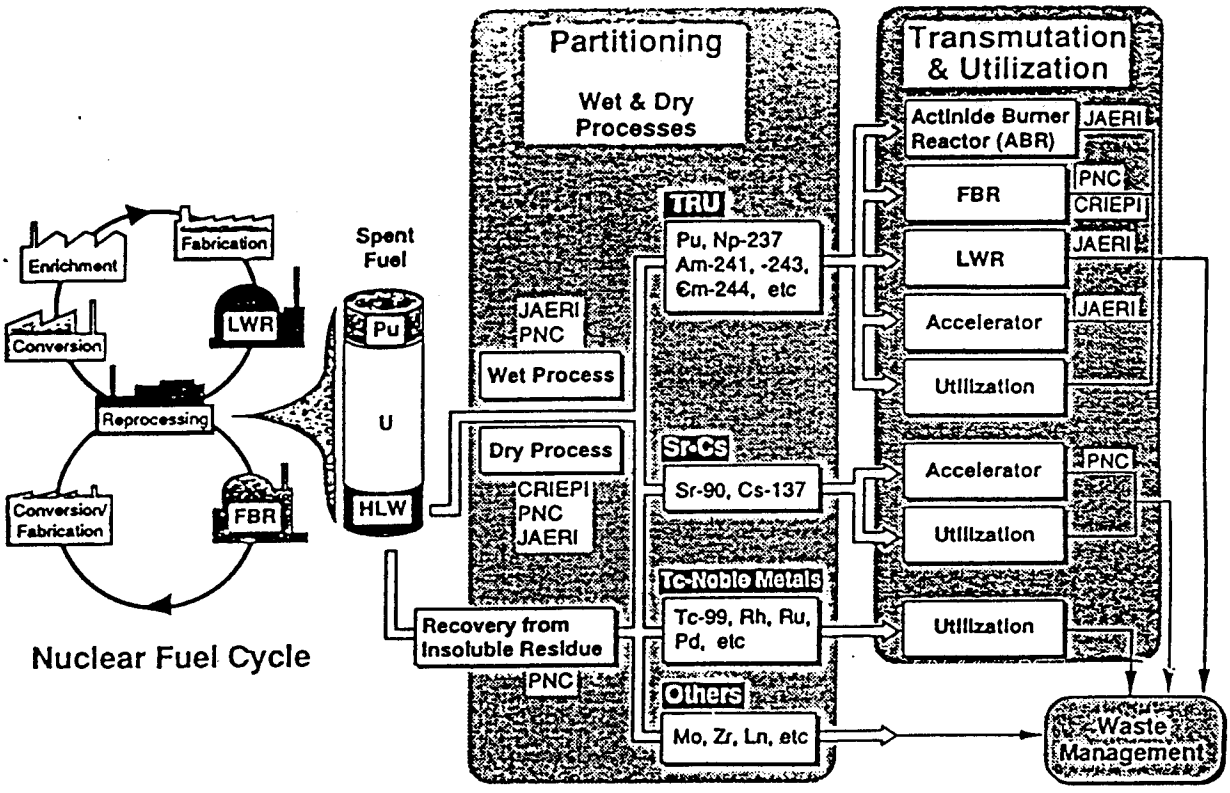


Fig. 1 R&D Activities under OMEGA Program

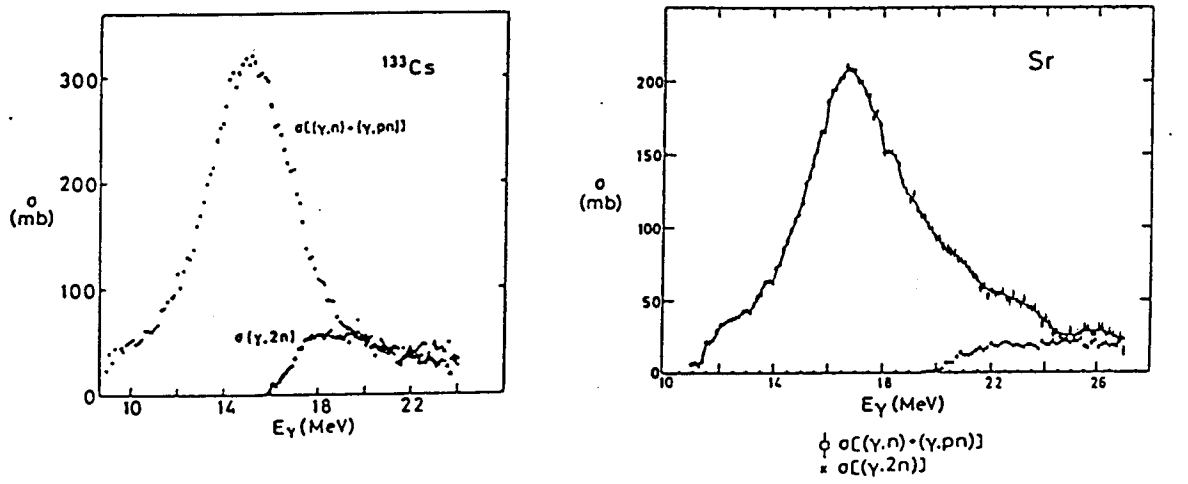


Fig. 2 The Cross Section of Photonuclear Reaction for Cs and Sr [10]

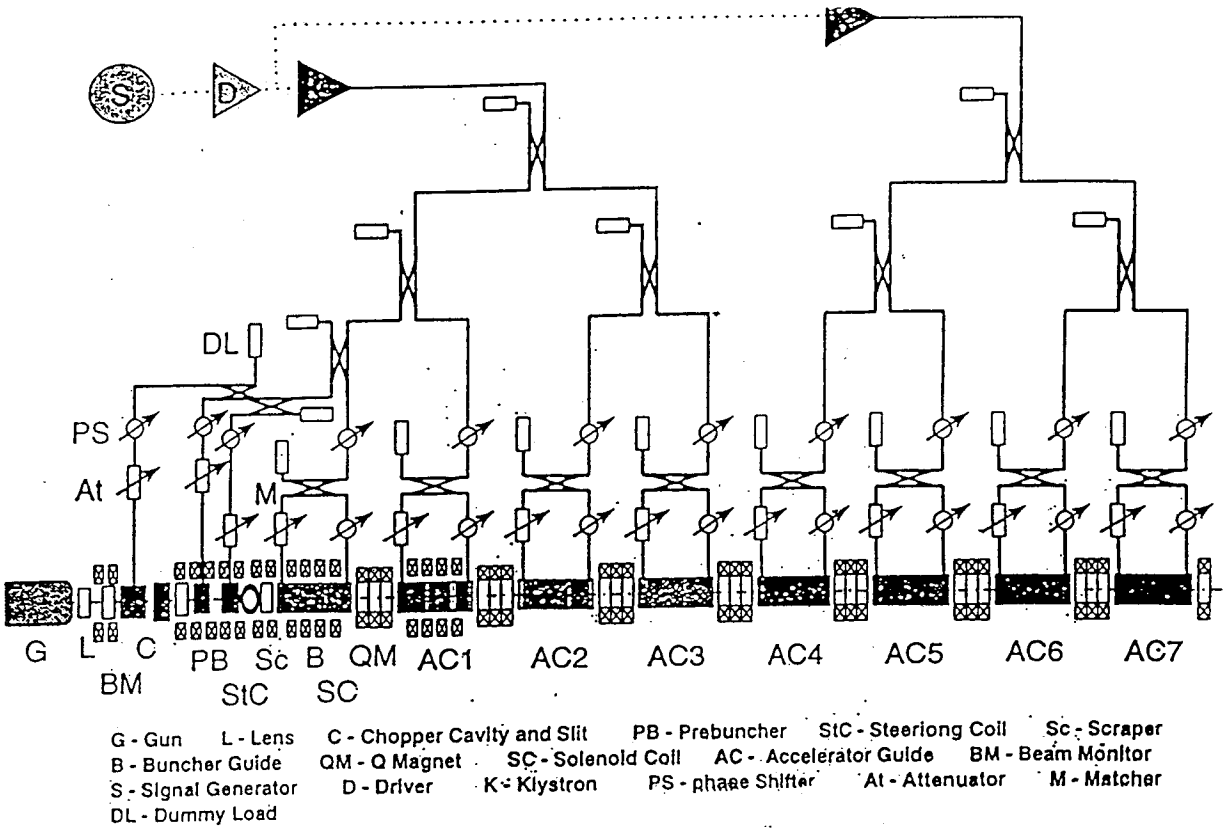
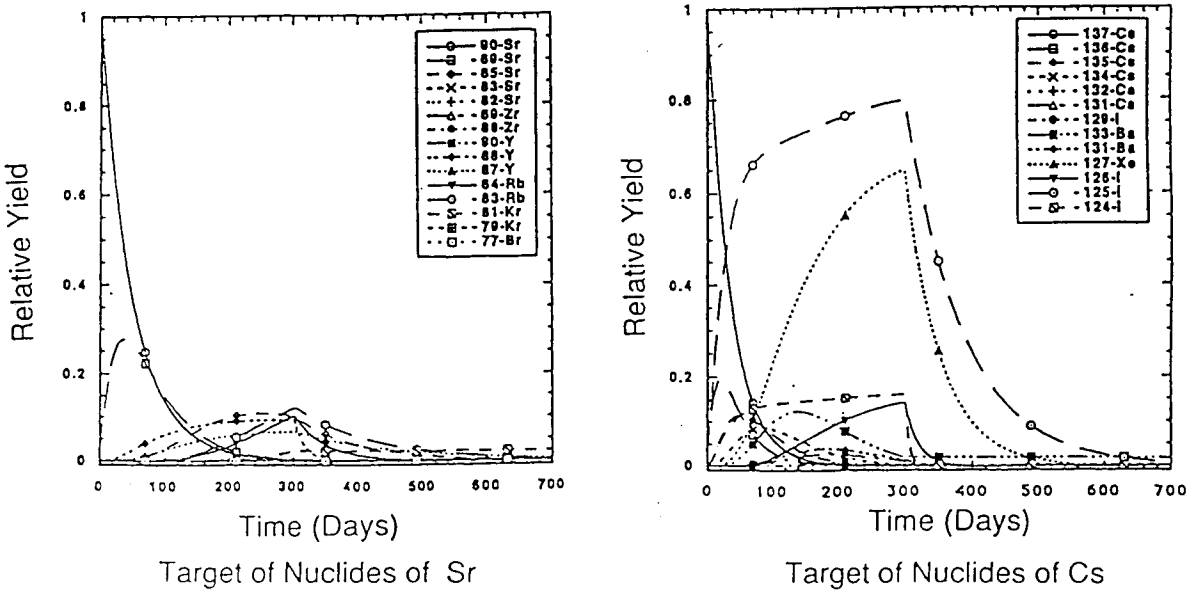


Fig. 5 Scheme of CW Electron LINAC of PNC



Target of Nuclides of Sr

Target of Nuclides of Cs

Fig. 6 Transmutation of Cs and Sr Targets by Photonuclear Reaction

USE OF ACCELERATOR TO NUCLEAR ENERGY

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Abstract.
Military Pu and the accumulated Pu from the operation of LWRs provide a great opportunity to start up a proliferation-resistance thorium fuel cycle, which has a high conversion factor in the thermal reactor, coupled with the Pu fuel cycle which has a high neutron economy in the fast reactor. The accelerator can play an important role in the safety of the fast breeder giving high breeding gain, yet separating energy generation and the processing and production of fuel. I will discuss a new approach to disposing of long-lived fission products (LLFPs) type II, such as Tc-99 and I-129, into outer solar space by providing an escape velocity from solar system of 42 Km/ sec from a parking orbit or from moon's surface, using a electro-static or RFQ accelerator and neutralizing the charged ions.

1. INTRODUCTION

Reactor safety, the disposal of high-level nuclear waste (HLW), and non-proliferation of nuclear material for military purposes are the problems of the greatest concern for nuclear energy. Technologies for accelerators developed in the field of high-energy physics can contribute to solving these problems [1]. For reactor safety, especially for that of an Na-cooled, Pu-fueled fast reactor, employing an accelerator, even a small one, can enhance the safety of using a slightly sub-critical reactor [2].

There is growing concern about how we can deal with weapons-grade Pu, and about the large amount of Pu accumulating from the operation of commercial reactors. It has been suggested that this Pu could be incinerated, using the reactor and a proton accelerator. However, because Pu is a very valuable material with the future potential of generating energy, we should consider transforming it into a proliferation-resistant material rather than simply eliminating it.

Although transmutation of minor actinides by fission processes produces energy, so that, in this reason, an energy balance easily can be achieved, to transmute the fission products themselves requires a substantial amount of energy[3].

It has been proposed to isolate HLW in outer space using a rocket[4]. However, this approach might disturb astronomical observations because the waste package would be a local source of radiation. Injection of HLW into the sun requires a rocket with more than about 30 km/sec velocity; this is too expensive even if the best chemical rocket-fuel was used, because the mass ratio (MR) of the total mass (mass of payload + mass of fuel) to the mass of the payload becomes very high, up to 1,235. By using a high-current electro-static or RFQ accelerator, we can disperse the LLFP at a smaller cost than with a rocket.

2. ^{233}U PRODUCTION AND TRANSMUTATION OF MINOR ACTINIDES

The volume of the spent fuel can be reduced by burning it for a long time in the reactor, which, in turn, can reduce the area needed for temporal or permanent storage for spent fuel. To do this, a fuel with a higher enrichment of U-235, which is cheap at present due to the small demand for electricity, has been used. The initial reactivity is suppressed with a burnable poison such as Gadolinium isotope, that captures the neutrons. However, this worsens the neutron economy. At present, LWR has a small conversion factor, only about 0.6, so that we are not only wasting the very valuable uranium resource, but also increasing radiation hazard due to mining the uranium source. A recent study carried out at Oak Ridge National Laboratory warned that the disposal of spent fuel containing minor actinides without processing them will create geological hazards.

At the last international fuel-cycle evaluation (INFCE), several options for producing ^{233}U were proposed[5]; however, this study was carried out before the collapse of Soviet Union, and the use of was not considered. We now have plenty of military Pu accumulated over the last 40 years of Cold War era, and also quantities of Pu operation of LWR. To incinerate completely the military Pu, it was suggested that an accelerator could be used as well as LWR. But Pu fuel is a very valuable fuel for future generations, and we should consider using this surplus Pu to start up ^{233}U and thorium fuel cycle in addition to a Pu fuel cycle.

It is well known that the ^{233}U thorium cycle is superior to the Pu fuel cycle for thermal reactors because of its high eta value. The use of Pu fueled fast breeder has been promoted because of high eta value for fast neutron. And this eta value of Pu increases as the neutron energy increases, thus creating a positive coolant-void coefficient even though the resulting hard spectrum can increase the breeding gain. To reduce this positive reactivity, a flat core or a small core has been recommended although they worsen the neutron economy. But using thorium in the blanket region instead of a uranium blanket also can reduce the positive reactivity, and the ^{233}U produced can be used in the thermal reactor with a high conversion factor.

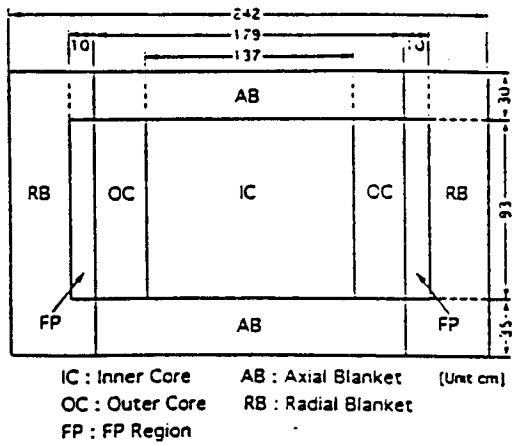

Fig. 1 Geometry of the Fast Neutron ^{233}U Producer and Transmutor of MA and LLFP when it is operated by subcritical condition, the lead target is installed in the center of the core

Table I. Multiplication Factor, Neutron Life-Time, Production of ^{239}Pu & ^{233}U, Transmutation of MA and LLFP, and Initial Breeding Ratio of Various Configuration of Reactor

	*Multiplication Factor Neutron Life time**	*Production of ^{239}Pu & ^{233}U transmutation MA or LLFP***	*Initial Breeding Ratio*
a) Uranium Blanket	1.0343+-.0043 129.4	^{239}Pu 243.8Kg	0.96026
b) Thorium Blanket	1.0275+-0.0045 134.56	^{239}Pu 123.8 ^{233}U 122.4	0.9765
c) Thorium Blanket Core	.9503+-.0046 123.94	^{233}U 265.5	1.0528
d) Thorium Blanket 5w/0 MA in core	.9808+-.0042 123.49	^{239}Pu 122.2 ^{233}U 106.2 Ma(cap) 41. Ma(fis) 10.9	1.02716
e) Thorium Blanket ^{99}Tc	.9920+-.0012 353.77	^{239}Pu 112.7 ^{233}U 60.1 ^{99}Tc 34.7	0.698
f) Thorim Blanket ^{129}I (30%)	1.002+-.006 462.18	^{239}Pu 173.6 ^{129}I 34.4	0.7014

a) Uranium-oxide(UO2) in core and blanket(BL), b) Thorium-oxide(ThO2) in BL, c) ThO2 in core and BL, d) ThO2 in BL & 5w% MA in core, e) ThO2 in BL & 99Tc with YH1.7, f) UO2 in BL &129I(30%) with YH1.7. * unit of 10^{-8}sec, ** in unit of Kg

We propose the use of a thorium blanket in which the neutrons are moderated by a moderator such as graphite, which can enhance neutron capture in the thorium blanket region without sacrificing breeding gain. This neutron modulation can introduce a negative coolant-density coefficient, and also a large doppler coefficient under some conditions, so that the safety-related neutronic behavior is improved together with a gain in the neutron life-time.

Figure 1 shows our geometry for a fast neutron ^{233}U producer and transmutor of MA which uses conventional mixed-oxide fuel, blended with MAs; thorium oxide is placed in the blanket region in the Prototype 700 MWt fast reactor [6]. Table I shows the initial production rate of ^{233}U and ^{239}Pu, the multiplication factors, neutron life time and initial breeding ratios for various blanket materials. When ^{232}Th is put in the blanket region, the rate of production of the total amount of ^{233}U created by the ^{232}Th blanket plus the ^{239}Pu created in core region is not very different from the total production of ^{239}Pu in a ^{238}U system. When ^{232}Th is put in the core region, ^{233}U production increases, but the multiplication factor, k, is reduced substantially; thus, a larger core volume is required.

When MA is added into the fuel, the production of ^{239}Pu increases, but the k value is slightly reduced.

^{233}U is produced with ^{232}U, which emits strong gamma radiation making it a more theft-resistant fissile material. However, until a few years ago the use of the thorium and ^{233}U fuel-cycle

system was opposed because of this high energy gamma-ray radiation generated by ^{232}U. Now, with the endorsement for mixing minor actinides into fuel cycle, the prospects of using this fuel cycle are far better. The large, heavy-shielded chemical processing facility might then be replaced by a compact facility, such as the facility for pyrolitic fuel developed by Argonne National Laboratory.

Improvements in breeding gain can be made by using a metal fuel in a fast reactor that makes a much harder neutron spectrum, and also in increasing the neutron economy to produce ^{233}U (or ^{239}Pu). Also, by running the reactor in a subcritical condition, the safety problem associated with criticality can be avoided. The produced ^{233}U fuel can be used in the LWR with ^{238}U, which produces ^{239}Pu for the fast reactor.

To prevent the removal of pure ^{233}U, which is more effective than ^{239}Pu as bomb material, can be obtaining from separated ^{233}Pa, the ^{232}Th should be mixed with a small amount of ^{238}U.

3. SEPARATION OF POWER PRODUCTION AND FUEL PROCESSING

From the point of view of non-proliferation, it is desirable to separate the power production and fuel-processing facilities; a small number of fuel-processing facilities which are internationally controlled is beneficial for inspection. The liquid fuel reactor with fuel-processing on site has a disadvantage in this regard, although there are several advantages over a solid fuel reactor.

If and when the world needs a large amount fissile material in a short time due to a rapid growth in energy demand, then the accelerator fuel producer can generate fissile material only from fertile material and electricity, without having a fissile material like breeder. The cost of the producing fissile material in this way is higher than that with a breeder. This high cost can be reduced somewhat by using a subcritical assembly; furthermore, the high cost of the fuel can be recovered by running the reactor with a high conversion factor.

When fuel-processing or fuel production facilities are located in a remote area where there is a fossil- fueled burning power plant or hydraulic power generator, the power can be used for fuel production using the accelerator without any loss during transmition from power generation to the consumer. In this case , a placement of many breeders is not suitable, because the fuel can be bred only by generating electricity which should be consumed; also, it requires a large inventory of fissile fuel.

When a critical reactor is operated, the multiplication factor should be 1, but in operation of a subcritical target, then k can be less than one, and the neutrons which are needed to maintain the critical condition can be diverted into producing fuel; hence the rate of fuel production can be increased by running the operation at a low k value. The initial requirement for fissile material will depend on the reactivity; we can adjust the reactivity by the amount of the initial inventory of the fuel material. To run the accelerator, electricity is needed. If we can produce the electricity from the target, the system can be self-sufficient not consume any power. Using a subcritical target which has multiplication factor k, then the accelerator current can be reduced proportionally to the subcriticality of (1-k).

We can deploy one power reactor close to the critical level and produce mostly power, while, the other target can have only a low level of power generation and can concentrate on production of fuel. In this system, the fuel production target has such low power density and the problem of thermo-hydrodynamics can be more easily solved. We might have another option, and instead of running rather many reactors with large subcriticality, use one power reactor and run the accelerator. I proposed using a small segmented cyclotron accelerator to run a slightly subcritical

reactor before, but if we can solve the problem of radiation damage and of non-uniformity of the spallation source by adopting the geometry configuration used for such as our old design of a regenerator for LWR fuel, and successful operation of large power accelerator with large subcritical condition to concentrate the fuel production. Accelerators can provide a lot of flexibility for fuel production than the fast breeder.

To be successful with nuclear power operation, we should have anyhow a reactor with a high conversion ratio or hopefully, a high breeding gain.

4. DISPOSAL OF LLFP INTO OUTER SPACE BY A ELECTRO-STATIC OR RADIO-FREQUENCY QUADRUPOLE (RFQ) ACCELERATOR[7].

From the point of view of neutron economy, it is not difficult to transmute MA by fission process which create neutrons. However, to transmute LLFP which consumes neutrons in the (n,gamma) reaction , the transmuting reactor has to have a high neutron economy. When the LLFP is transmuted to into nuclei which have a large neutron capture cross-section, the neutron economy worsens due to additional neutron capture.[3] Some of the LLFP has a small neutron cross-section; therefore, a high neutron flux is required to transmute it rapidly.

The MOX-typed fast reactor with a thermal power of 700 MWt[6,8], which is described in the above, can transmute a substantial amount of LLFP, but the breeding ratio falls from about 0.96 to 0.7 because of the many neutrons that are consumed in transmutation. The economy of such transmutation should be studied more thoroughly.

It has been suggested that HLW might be disposed of in outer space using a rocket. The National Aeronautical and Space Administration (NASA) studied[4-1] the possibility of using the space shuttle for this. Several possible options for the destination in space have been evaluated, such as Earth orbits, solar orbits, solar-system escape, and solar impact after first launching the HLWs into a parking orbit by space shuttle. Although disposition to Earth orbits and the solar orbit do not require a large propulsion energy even, using a chemical fuel.

However, these methods have several disadvantages; future planetary spacecraft would regularly penetrate this orbit belt; there is no assurance that the waste package would not encounter Earth after a few thousand years; and, the isolation of solid HLW in outer space might disturb astronomical observations because it would be a local source of radiation.

The disposition of the HLW into an orbit escaping the solar system is ideal but requires a large amount of energy; with mass ratios (MRs) of (mass of payload + mass of fuel)/(mass of payload) the MR becomes very high, upto 21,883, when a chemical rocket fuel is used. Such a method of disposal requires a nuclear propulsion rocket which has a large I_{sp} =1000 sec, or 6000 sec for a solid-core nuclear fuel or gaseous-core nuclear fuel.

Instead of using a rocket to generate such high escape velocity, by ejecting LLFPs using an accelerator, we can reduce substantially the energy needed for their disposal.

To uniformly disperse the LLFPs into outer solar space from a parking orbit, requires an earth escape velocity (V_{EE}) = 42.07 Km/sec. Column 2 of Table 2 shows the accelerating energy, which is in the order of 1 KeV, for each LLFP isotope. The third column shows the power required to accelerate the total amount of LLFP isotopes created by one 1 GWe LWR.

Instead of dispersing the LLFPs uniformly in solar outer space, if we disperse them uniformly in the plane of the earth's orbit, then the velocity required to eject them can be reduced from 42.07km/sec to $\{\sqrt{2}-1\}V_{EC}$=12.32 km /sec. The fourth column in Table 2 shows the acceleration energy for each LLFP isotopes, and the fifth column shows the power for

accelerating all the isotopes produced by one 1 GWe LWR.

	Disposing uniformly in the outer solar system.		Disposing uniformly in the earth orbit plane of outer solar system.	
Species	Accelerating Energy (eV)	Accelerator Power (W)	Accelerating Energy (eV)	Accelerator Power (W)
^{79}Se	722.2	5.523	61.9	0.473
^{93}Zr	850.2	669.8	72.9	57.43
^{99}Tc	905.0	718.5	77.6	61.6
^{107}Pd	978.2	978.2	83.9	17.4
^{126}Sn	1151.9	25.374	98.8	2.17
^{129}I	1179.3	166.7	101.1	14.3
^{135}Cs	1234.2	279.8	105.8	24.0
Total		2843.9		177.4

TABLE 2.
The energy required to accelerate the isotropic species of LLFPs which will be disposed uniformly in the outer solar system and in an Earth orbiting plane. The corresponding accelerator power used for ejecting the LLFPs generated by a 1 GWe light water reactor

	Disposing uniformly in the outer solar system.		Disposing uniformly in the earth orbit plane of outer solar system.	
Species	Accelerating Energy (eV)	Accelerator Power (W)	Accelerating Energy (eV)	Accelerator Power (W)
Se	722.2	525.8	61.9	45.1
Zr	850.2	3374.4	72.9	289.3
Tc	905.0	718.5	77.6	61.6
Pd	978.2	1278.8	83.9	109.7
Sn	1151.9	162.1	98.8	13.9
I	1179.3	218.3	101.1	18.71
Cs	1234.2	2220.3	105.8	190.3
Total		8498.2		728.6

TABLE 3.
The energy required to accelerate the elemental species of LLFPs which will be disposed uniformly in outer solar system and in an Earth orbiting plane. The corresponding accelerator power used for ejecting the LLFPs generated by a 1GWe light water reactor.

The above values were calculated for LLFPs which were separated out isotopically from the other elements. When the isotopes are not separated, the total product has to be ejected into outer solar space; the acceleration energy needed is almost same as the isotopic requirement without taking into account the small mass difference of isotope, Then the power for accelerating all the LLFPs elements produced by one 1 GWe LWR to dispose uniformly in outer space and in the earth's orbit plane, are respectively, 8.5 KW and 0.73 KW, as shown in third and fifth columns in Table 3.

These powers are far smaller than the energy required to transmute the LLFPs by spallation neutrons where the neutrons are multiplied by the subcritical assembly, which is in the order of a few 10s MW.

To prevent these charged ions being trapped by the magnetic field in outer space, the ions should be neutralized in the same way as the neutral beam injection in the magnetic fusion reactor[9].

To reduce the high current, we might to accelerate the singly charged clustered LLFPs, composed of 100-1000 ions.

5. CONCLUSION

Accelerators can provide great flexibility for various nuclear options, as well as markedly improving reactor safety. The Thorium-^{233}U fuel cycle which has high conversion factor for thermal neutrons, can be started by using military Pu or Pu accumulated from operating LWRs, together with the Pu fuel cycle which has superior neutron economy. To reduce the costs and energy needed for transmuting LLFPs, they can be deposited into outer solar space with high-current, low-energy electro-static or RFQ accelerator.

6. ACKNOWLEDGEMENT

The authors would like to express their thanks to Drs.H.Rief, G.Maise, H.Takashita, H.Harada, and Prof. S.Mikami for valuable discussions, and for providing the information for writing this report, and Dr. Woodhead for editorial work. This work was performed under the auspices of the U.S. Department of Energy under Contract No. DE-AC02-76CH00016, and under The Power Reactor and Nuclear Fuel Development Corporation, Japan Atomic Energy Research Institute.

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Nuclear Data for the OMEGA Project

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In order to provide a reliable database for the OMEGA project, two types of the evaluated data files will be compiled at the JAERI Nuclear Data Center in cooperation with the Japanese Nuclear Data Committee. One is the JENDL Actinoid File which contains neutron induced-reaction data for about 90 actinoid nuclides, and the other is the JENDL High Energy File for neutron and protons up to a few GeV.

JENDL Actinoid File

The JENDL Actinoid File will be compiled to meet the data requirements for study of actinide burner reactors. This file will contain the data for about 90 nuclides from ^{208}Tl to ^{255}Fm, in the neutron energy region from 10^{-5} eV to 20 MeV. Table 1 shows the nuclides to be stored in JENDL Actinoid File. Among them, data for about 60 nuclides are already existing in JENDL-3.2. However, reevaluation is needed for data of important minor actinides. The new evaluation will be done for about 30 nuclides. In 1993, a new evaluation was made for ^{244}Pu and ^{237}Pu.[1)]

The neutron cross sections for the fission, (n,2n), (n,3n) reactions have been calculated[2)] consistently for 71 isotopes of $^{227-234}$Th, $^{229-233}$Pa, $^{230-240}$U, $^{235-239}$Np, $^{236-247}$Pu, $^{239-245}$Am, $^{238-251}$Cm, $^{245-249}$Bk and $^{249-252}$Cf using the Hauser-Feshbach code STAPRE[3)] and GNASH[4)]. Shell, superfluid and collective effects in nuclear level density have been taken into account. Neutron transmission coefficients were calculated using the coupled channel code ECIS[5)]. All experimental data available for the above isotopes have been used for model testing. A consistency between theoretical and experimental data was achieved, and the theoretical prediction of neutron cross sections has been made.

JENDL High Energy Files

JENDL High Energy Files will contain the data for neutrons and protons up to 50 MeV and those up to a few GeV. The former files are required for design of a material irradiation test facility for fusion neutronics, and the latter ones for an accelerator-driven radioactive waste transmutation system. The data up to 50 MeV are evaluated, mainly, by means of SINCROS-II system[6] based on GNASH[4], ELIESE-3[7], DWUCK-4[8] and auxiliary programs. For very light mass nuclei, another code will be used. High priorities are put on H, Li, C, N, O, Al, Si, Cr, Fe, Ni, Cu for the file of neutron-induced reactions, and Li, C, Al, Cr, Fe, Ni, Cu, Mo, W and Ta for proton-induced reactions. The evaluation will be completed at the end of 1995.

The evaluation of nuclear data up to a few GeV is based on the theoretical calculation, systematics of cross sections, and a fitting to available experimental data. The evaluation for H, ^{28,29,30}Si, ^{52}Cr, ^{58,60}Ni, ^{63,65}Cu, ^{208}Pb and ^{209}Bi has been completed already in the energy range up to 1 GeV. ALICE-F[9] was used for their theoretical calculation except for H. Figure 1 shows the ^{209}Bi(p,x)^{206}Po cross section.

In order to investigate an evaluation method for fissile nuclei, the intranuclear cascade-exciton model taking into account the fission process[10] was used for neutron and proton cross-section calculations in the energy range from 50 MeV to 1 GeV. The calculations showed that the experimental data available for ^{232}Th, ^{235}U, ^{238}U, ^{237}Np and ^{239}Pu for neutrons and protons can be reproduced rather accurately (within 5 to 10 %) by the adopted model which takes account of the fission barrier height values by Sierk[11], level density systematics by Iljinov et al.[12] with ground state shell and pairing corrections by Truran et al.[13], and with taking into account $B_f(E^*)$ and $B_f(L)$ dependencies. Therefore the intranuclear cascade-exciton model was used for neutron and proton cross-section calculations for ^{232}Th, ^{232}Pa, ^{235}U, ^{238}U, ^{239}Pu, ^{237}Np, ^{238}Np, $^{241-243}$Am and $^{242-248}$Cm in the energy range from 100 MeV to 1 GeV. In the energy range from 20 to 100 MeV the GNASH code[4] which takes account of the decay of 9 subsequent compound nuclei was used for calculation of neutron fission and (n,xn) cross sections for ^{232}Th, ^{235}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, $^{241-243}$Am and $^{242-247}$Cm. The fission and (n,xn) reaction cross sections of ^{239}Pu are given in Fig. 3.

Application of the quantum molecular dynamics (QMD) theory is also being investigated, and remarkable progress has been achieved.[14] A new code based on the QMD theory is under development in JAERI, and hopefully it may be a useful tool for high energy nuclear data evaluation. The double-differential cross section of ^{56}Fe(p,xn) at the proton energy of 1.5 GeV is compared with experimental data in Fig. 3. The solid curve shows the result of the QMD+statistic decay model, and the dashed one denotes a result of cascade+evaporation model code NUCLEUS[15]. It was found that QMD gives almost identical results on the double-differential (p,xn) reaction cross sections with the conventionally used cascade model. The QMD, however, gives a better prediction of various isotope production cross sections.

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Table 1 Nuclides in JENDL Actinoid File

^{208}Tl	×	^{210}Pb	×	^{210}Bi	×
^{210}Po	×	^{222}Rn	×	^{223}Ra	J3
^{224}Ra	J3	^{225}Ra	J3	^{226}Ra	J3
^{228}Ra	×	^{225}Ac	J3	^{226}Ac	J3
^{227}Ac	J3	^{227}Th	J3	^{228}Th	J3
^{229}Th	J3	^{230}Th	J3	^{231}Th	×
^{232}Th	J3,A	^{233}Th	J3	^{234}Th	J3
^{229}Pa	×	^{230}Pa	×	^{231}Pa	J3
^{232}Pa	J3	^{233}Pa	J3	^{230}U	×
^{231}U	×	^{232}U	J3	^{233}U	J3,A
^{234}U	J3	^{235}U	J3,A	^{236}U	J3
^{237}U	J3	^{238}U	J3,A	^{234}Np	×
^{235}Np	×	^{236}Np	J3	^{237}Np	J3,B
^{238}Np	J3,B	^{239}Np	J3	^{236}Pu	J3
^{237}Pu		^{238}Pu	J3,B	^{239}Pu	J3,A
^{240}Pu	J3,A	^{241}Pu	J3,A	^{242}Pu	J3,B
^{244}Pu		^{246}Pu	×	^{247}Pu	×
^{241}Am	J3,B	^{242}Am	J3,B	^{242m}Am	J3,B
^{243}Am	J3,B	^{244}Am	J3	^{244m}Am	J3
^{240}Cm	×	^{241}Cm	J3	^{242}Cm	J3,B
^{243}Cm	J3,B	^{244}Cm	J3,B	^{245}Cm	J3,B
^{246}Cm	J3,B	^{247}Cm	J3	^{248}Cm	J3
^{249}Cm	J3	^{250}Cm	J3	^{245}Bk	×
^{246}Bk	×	^{247}Bk	×	^{248}Bk	×
^{249}Bk	J3	^{250}Bk	J3	^{246}Cf	×
^{248}Cf	×	^{249}Cf	J3	^{250}Cf	J3
^{251}Cf	J3	^{252}Cf	J3	^{253}Cf	J3
^{254}Cf	J3	^{251}Es	×	^{252}Es	×
^{253}Es		^{254}Es	J3	^{254m}Es	×
^{255}Es	J3	^{255}Fm	J3		

A: major actinide, B: important for the actinide burner reactor, J3: already in JENDL-3, ×: no evaluated data

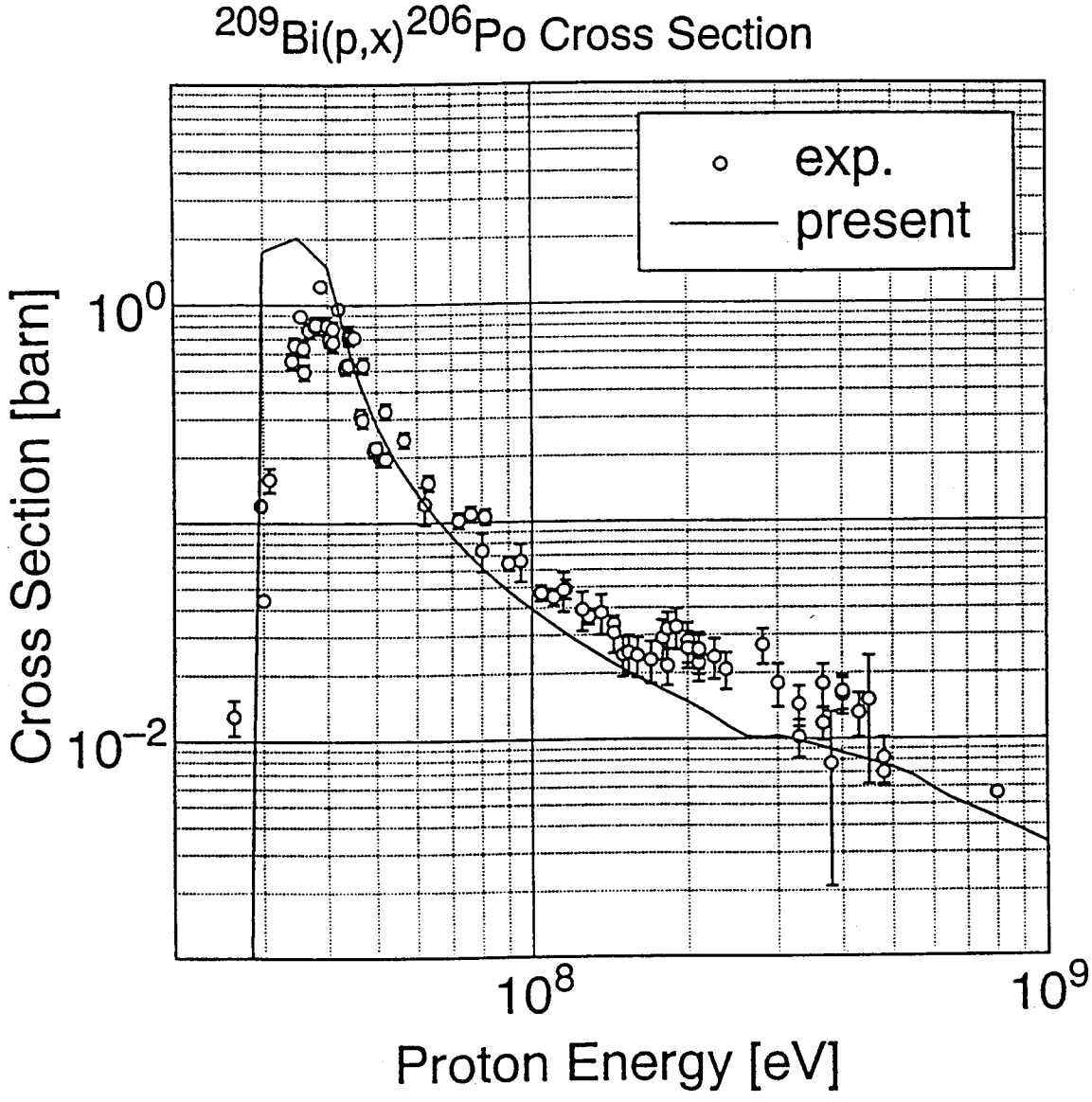

Fig. 1 $^{209}Bi(p,x)^{206}Po$ Cross Section

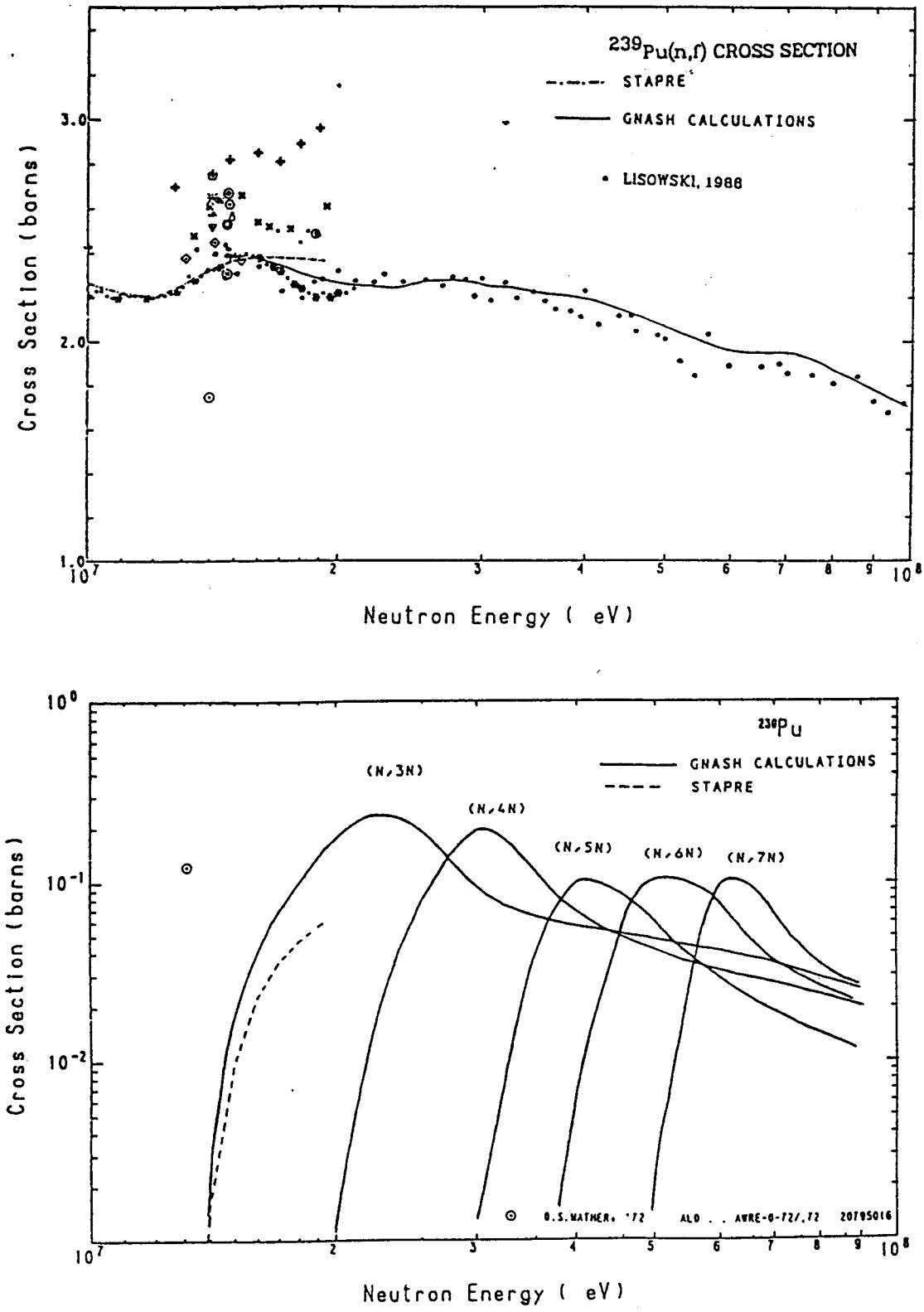

Fig. 2 ^{239}Pu fission and (n,xn) reaction cross sections.

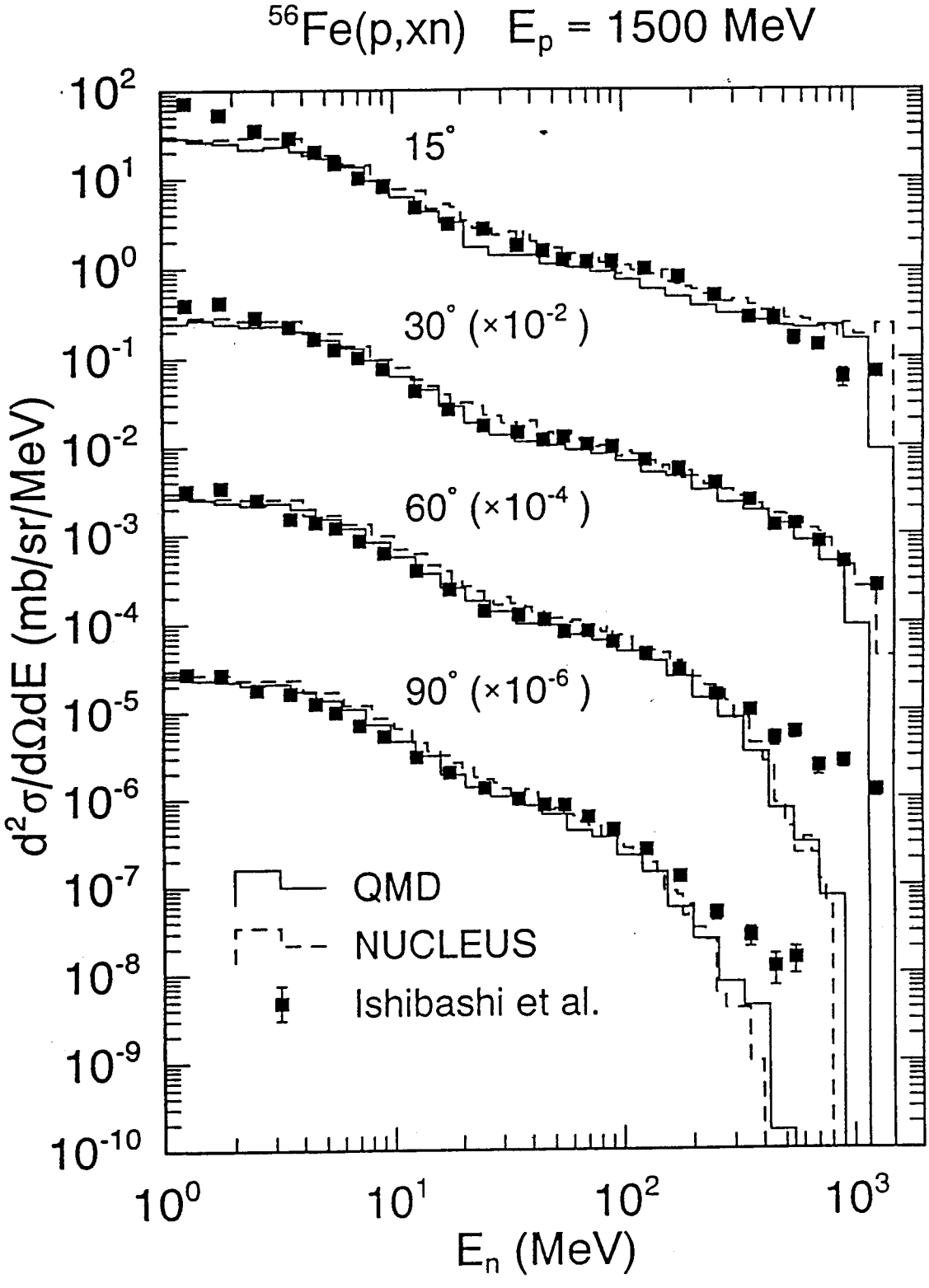

Fig. 3 Double-differential ^{56}Fe(p,xn) reaction cross section at 1.5 GeV