Transmutation capability of the High Flux Reactor BR2 for Minor Actinides and Long Lived Fission Products

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ABSTRACT

The high flux reactors are presently the only existing facilities with the highest potential for transmutation of minor actinides and long lived fission products. The material testing reactor BR2 at MOL, BELGIUM with, in routine operation, an effective thermal neutron flux ranging, in the core, from 2 to 4 10^{14} n/cm².s; in a flux trap, up to 8 10^{14} n/cm².s; and in a reflector position, up to 4 10^{14} n/cm².s, and a large epithermal contribution (about 0.4 10^{14} n/cm².s per unit lethargy at 5 eV in a core position).

In the sixties 'and early seventies, important amounts of Actinides have been irradiated for isotopic heat source production and fabrication of higher actinides (Am, Cm, Bk...). The expertise and hardware for capsule irradiation is available at the C.E.N./S.C.K. laboratories as a result of the Pu fuel pin irradiations in extreme conditions.

The preferred form of minor actinides irradiation is the oxide form homogeneously mixed with MgO or $A1_2O_3$. In case of 1-129 specific target compositions have to be selected in order to avoid target-capsule wall interactions and the Tc-99 source form depends on the accompanying nuclides.

The quantity of nuclides which can potentially be irradiated in the BR2 at the highest flux positions (inside fuel elements) are illustrated for the most important nuclides : Np, Am (Cm), 1-129 and Tc-99 and the spent target compositions are derived for a few representative cases. The neutron shadowing and local flux depressions are taken into account in order to obtain realistic data which can be extrapolated to taller capsules and larger irradiation facilities.

The interaction between the cladding material or the capsule wall is of primary importance for the selection of long term, high fluence irradiations.
On the basis of these data, a chemical-metallurgical recycle scheme is designed in order to process the spent targets and to reirradiate the residual nuclide inventory.
A global mass balance is given for the minor actinides in the target capsules and in the driver fuel in order to derive the overall radiotoxicity reduction obtainable in a P & T scheme based on the use of high flux thermal neutron reactors.

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TRANSMUTATION CAPABILITY OF THE HIGH FLUX REACTOR BR2

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1. INTRODUCTION

In the framework of a comprehensive Partitioning and Transmutation (P&T) option, the part taken by Transmutation is crucial to achieve a reduced long term radiotoxic potential.

While Geological Disposal is an option which aims at protecting the environment from unwanted contamination by actinides and Long-Lived Fission Products (LLFP) during a period of up to 10000 years and in the best case up to 100000 years, it cannot guarantee eternal confinement. The actinides and the LLFP with half lives equal to or surpassing the mentioned periods and which are critical for the validity of the disposal option are given in Table I (taken from [1]).

The conventional backend of the fuel cycle with reprocessing, storage of HLLW and vitrification constitutes the ultimate steps during which man can influence the source term. In this paper, uranium and plutonium are supposed to be recycled for 99 % into thermal or fast fuel. The residual plutonium and the so-called minor actinides (M.A.) are according to the present practice trapped into a borosilicate matrix which is suitable for storage of fission products and actinides during thousands of years but not for 10000 years or more.

Iodine-129, with a half-life of 16.7 M years, is presently discharged into the ocean as a fraction of the Medium Level Waste effluents. Its presence in the marine biosphere will steadily increase the global collective dose. Even if iodine were trapped and stored, there is no geologic confinement capable of withholding this nuclide from worldwide circulation.

TABLE I

Actinides and Long Lived Radionuclides composition of PWR - UO_2 spent fuel. and its HLW equivalent at 33.000 MWd/T after 5 years cooling.

ACTINIDES					FISSION PRODUCTS				
NUCLIDE	t1/2(y)	C i /THM	g/THM	HLW FRACTION	NUCLIDE	t1/2(y)	Ci/THM	g/THM	HLW FRACTION
U 232	70	$3.27\ 10^{-2}$			Sr 90	28.5	$6.63\ 10^{4}$	485	
U 234	$2.44\ 10^{5}$	0.94	150		Y 90	equil	$6.63\ 10^{4}$	0.126	
U 235	$7.\ 10^{8}$	$1.79\ 10^{-2}$	8250	1% to	Zr 93	$1.5\ 10^{6}$	1.79	736	
U 236	$2.34\ 10^{7}$	0.262	4050	HLW	Tc 99	$2.1\ 10^{5}$	13.2	841	100 % to
U 238	$4.47\ 10^{9}$	0.317	943000		Pd 107	6.5 10	0.102	231	HLW
Np 237	$2.14\ 10^{6}$	0.308	437	100% to HLW	Sn 121	50	0.146	4.5	
Pu 238	87.7	$2.39\ 10^{3}$	140		Sn 126	10^{5}	0.542	19.2	
Pu 239	24110	340	5470		I 129	$1.57\ 10^{7}$	$3.12\ 10^{-2}$	229	0.1% to HLW
Pu 240	6550	509	2230	1% to	Cs 135	$2\ 10^{6}$	0.352	324	
Pu 241	14.4	$9.85\ 10^{4}$	956	HLW	Cs 137	30.1	$9.5\ 10^{4}$	1100	100 % to
Pu 242	$3.7\ 10^{5}$	1.86	486		Ba 137	equil.	$9.4\ 10^{4}$	178	HLW
Am 241	432.6	1002	296		sm 151	93	324	44.2	
Am 242	142	3.8	0.39						
Am 243	7370	16.7	83.8	100% to HLW	LIGHT ELEMENTS				
Cm 243	28.5	14.8							
Cm 245	8500	0.138	0.804		c 14	5730	0.143	0.0315	0.1 % to HLW
Cm 246	4730	0.225	0.731		Ni 59	$7.5\ 10^{4}$	17.5	216.6	100% to
					Ni 63	100	24.10^{3}	24.28	HLW

Technetium-99 , with a half-life of 2.1 E+5 years is present as a metal or oxide in the insoluble residues and as soluble TcO_4^- in HLLW. According to present practice, both fractions are combined and vitrified. Due to its long half-life and variable valency states, this nuclide will undoubtedly migrate through a geological barrier which is not sufficiently reducing. It will have a radiological impact on the groundwater in the vicinity of a geological repository.

Taking into account these well known arguments in favour of partial P&T for very long-lived radionuclides, we have examined the potential inventory reduction of these nuclides by transmutation in a high neutron flux facility. Several so-called "Materials Testing Reactors" (MTRs) are available throughout the world which provide neutron fluxes of E+14 to E+15 n/cm^2s. In Europe a number of MTRs are still operating : OSIRIS in France, HFR in the Netherlands and BR2 in Belgium.

2. THE HIGH FLUX MATERIALS TESTING REACTOR BR2 [2to 12]

BR2 is a heterogeneous thermal high flux MTR designed in 1957 for S.C.K./C.E.N. by the Nuclear Development Corporation of America (NDA) . It has been built on the site of S.C.K./C.E.N. at Mel, Belgium. Routine operation of the reactor started in 1963 and, to this very day, continues to contribute to the development of many large nuclear projects within the European Community and for nuclear partners throughout the world.

The reactor is cooled and moderated by pressurized light water (12 bar) in a compact core of highly enriched uranium positioned in and reflected by a beryllium matrix. The ultimate cooling capacity, initially foreseen for 50 MW, has been increased in 1971 to 125 MW.

The beryllium" matrix has 79 cylindrical holes in a hexagonal lattice of 96.44 mm pitch at the reactor midplane (Fig. 1); all channels have individual access through holes in the top cover of the reactor vessel. Individual guide tubes join the holes in the top cover to the holes in the matrix and thus create 79 individual reactor channels. There are 64 standard channels (84 mm diam.), 10 small peripheral channels (50 mm diam.) and 5 large channels (200 mm diam.). All the channels

can receive fuel elements, control rods, beryllium plugs or experiments , which allows a great flexibility for a variety of core arrangements. The five 200 mm channels and thirteen of the 84 mm standard channels have additional apertures through the bottom cover of the reactor vessel, which allows through-loop experiments.

One of the most characteristic features of BR2 is its geometry : the hexagonal Be-matrix prisms form a hyperboloidal bundle with channels which are close to each other at the midplane but more apart at the lower and upper ends, i.e. at the penetrations through the covers of the reactor pressure vessel (Fig. 2). In this array, a very high fuel density is achieved in the middle part of the vessel (reactor core) while leaving enough space at the covers for easy access to the channel openings.

The neutron chain reaction is controlled by a number of Cd(active part)-Be(rod follower) shim safety rods and by a regulating rod, which occupy matrix lattice positions.

The standard fuel elements loaded in the 84 mm diameter channels (Fig. 3) 'are assemblies of concentric plates or **tubes of about 84 mm outer diameter, each having a** 762 mm **fuel length. The water gap between the plates is 3 mm wide, the plate thickness is 1.27 mm. A fuel plate consists of a cermet mixture of fully enriched (93 %) U AI_x and burnable poisons** (B_4C **and** Sm_2O_3), aluminium **clad. A standard fuel element of the VIG type consists of six concentric plates and allows for an experimental space having a diameter of 25.4 mm while a fuel element of the VG type consists of five concentric plates of the same composition and allows for an experimental space of 34 mm.**

The presently used VIG fuel elements (with so-called cermet G fuel) contain, when fresh, 400 g U-235 in the form of UAl_x (1.3 gU/cm^3) + 3.8 g boron (B_4C) + 1.4 g samarium (Sm_2O_3).

The reactor core contains 10 to 13 kg U-235 (30 to 40 fuel elements, not all fresh). The concentration at discharge of the fuel elements is about 50 % of the initial fissile content value.

A typical BR2 reactor cycle consists of 7 days shut-down (for reloading), followed by 21 days operation. The shut-down

periods can be prolonged up to about 40 days for inspection, maintenance, etc.

The present nominal heat flux at the hot spot is 470 W/cm², the maximum value allowed for nominal cooling conditions (probable onset of nucleate boiling) is 600 W/cm². The nominal full power depends on the core configuration used; at present, it ranges from 60 to 100 MW.

Typical neutron fluxes are (in the reactor hot spot plane) :

- thermal conventional neutron flux : $v_0 \int_0^{0.5\ eV} n(E)\,dE$:

  2 to 4E+14 n/cm2s in the reactor core
  2 to 9E+14 n/cm2s in the reflector and core flux trap (Hi)

- fast flux : $\int_{0.1\ MeV}^{\infty} \phi(E)\,dE$:

  4 to 7E+14 n/cm²s in the reactor core.

Typical BR2 neutron 'flux specra are shown on Fig. 4.

3. TARGET PREPARATION

3.1. QUANTITIES TO BE IRRADIATED

In order to assess the magnitude of the M.A. conditioning operations including chemical purification from rare earths, conversion to oxide, encapsulation and/or incorporation in fuel elements, it is necessary to quantify the M.A. inventory produced by reprocessing operations from spent LWR (33000 MWd/T after 5 years cooling). Table II shows the M.A. data per THM discharged per GWe/year and per large reprocessing unit.

The total amount of Np + Am discharged per year from all reprocessing plants amounts at present to 3100 kg.

The quantities of long-lived fission products Tc-99 and 1-129 are shown in the lower part of Table II for the same type of fuel and according to the same subdivision.

The other important LLFP , Cs-135 and Zr-93 are not discussed in this paper since they would require a preliminary isotopic separation from the other isotopes of Cs and Zr to be transmuted efficiently. Other methods than neutron induced transmutation ought to be developed for that purpose.

TABLE II :

OCCURRENCEOF M.A. INSPENTFUEL PER THM AND PER GWe CAPACITY
OVERALL PRODUCTION CAPACITY OF THE MAJOR REPROCESSING PLANTS

Nuclide	Specific yields		M.A. Annual production in kg/year		
	g/THM	kg/GWe	La Hague UP2 + UP3	Sellafield THORP	Rokkasho (+ Tokai)
Np-237	437	11.8	700	524	349
Am-(241 + 243)	380	10.26	608	456	304
Cm-(242-247)	1.5	0.045	2.4	1.8	1.2
Pu(0.7 %)	" 65	1.75	104	78	52
Tc-99	841	22.7	1345	1009	841
1-129	229	6.18	366	274	229

3.2. METALLURGICAL ASPECTS

The target preparation has to be carried out in order to meet the requests which are imposed by safety reasons and feasibility considerations. The main parameters to be specified are :

- physical and chemical forms;
- compatibility and geometrical stability;
- thermal conductivity or diffusivity;
- axial nuclide density.

To avoid parasitic neutron absorption, zircaloy has been chosen as cladding material and aluminium alloys as irradiation device material.

3.2.1. Neptunium and Americium Targets

For fabrication reasons, it is suggested to prepare the M.A. targets by a sphere-pack oxide method resulting from the sol-gel route. In this case, NpO_2 and/or Am_2O_3 are mixed with Al_2O_3 or, preferably, MgO powder and vibrocompacted to obtain a bulk density of 75 % of the theoretical density.

No change in the metallurgical or geometrical stability is expected during the first step of transmutation if the centerline temperature does not reach the melding point of the eutectic compound. The fissile nuclide will behave roughly as UO_2 and the geometrical stability of the target can be assured as long as the temperature does not reach the melting point ($T < 2000°K$).

The compatibility between the target matrix and the cladding material does not give rise to any problem. The fission products' mixture is not different from that resulting from $U0_2$ but a slight change in their distribution could induce an oxidizing reaction between alkaline metal type fission products and zircaloy. Some metallurgical in-pile tests would have to be carried 'out to investigate this phenomenon and to verify any absence of chemical stress-corrosion cracking by fission products, in particular by iodine.

The thermal conductivity or diffusivity would have to be measured by in-pile measurements if the target is to be irradiated at a centerline temperature close to the melting point.

3.2.2. Iodine targets

Due to its high volatility even at low temperature, its geometrical instability (which depends on the melting point of the compound used), and its high reactivity with the zircaloy cladding, iodine must be loaded as iodide compound. Oxidized species have to be avoided in order to limit oxygen liberation after transmutation to Xe-130. Na, Ca and Ba iodides have been considered and CaI_2 has been chosen as the reference compound because of its higher volume concentration in iodine and its higher melting point (750°C). The target is to be fabricated by loading sphere-pack CaI_2 into the zircaloy capsule and vibrocompacting it to 70 % T.D.

The compatibility between the capsule material and the CaI_2–Ca mixture is assured as long as oxygen or oxidized compounds are absent during transmutation. The starting procedure has to be properly qualified because absorbed oxygen, zircaloy and metallic calcium react to form calcium-zirconate in equilibrium with metallic Zr and Ca. This reaction comes to a halt when all free oxygen has been consumed and no other parasitic reactions are to be expected. The reaction rate can be slowed down by keeping the cladding surface temperature low enough. In-pile metallurgical tests are needed to verify the compatibility between the cladding and the CaI_2 loading to investigate the metallurgical stability and the potential axial iodine migration.

The Xe-130 formation will require a vented target with an inner pressure close to the BR2 water pressure in order to avoid any stress between the outside and the inside of the capsule as a very long irradiation time is required to achieve depletion of the target. For safety reasons, it will be necessary to load the capsule in a closed loop device which will limit external contamination in case of accidental cladding failure.

3.2.3. Technetium Target

If Tc has been separated from other platinum metals, it can be used in its metal or lower oxide form (TcO_2) to be irradiated in a zircaloy capsule. No particular problems have to be expected during transmutation as it transforms into stable and metallurgically similar ruthenium. No second barrier is required nor any special safety device.

4. IRRADIATION DEVICE

In order to permit large scale transmutation of M.A. and LLFP in a MTR very stringent safety requirements have to be fulfilled. The BR2 reactor has a longstanding experience in that type of irradiations, particularly with the production of transuranic elements in the sixties and with a large scale Ra-226 irradiation programme in the early seventies. Quantities of the order of 600 g of Ra-226 have been irradiated to produce gramme quantities of Ac-227 and Th-228.

To avoid general contamination of the reactor in case of accidental failure of the capsule, special irradiation devices have to be designed :

- to maintain the cladding temperature as low as possible, to isolate the primary coolant circuit of BR2 from the highly active targets,
- to permit an adequate siting of the capsules within a VIG or a VG fuel element,
- to become easily deactivated after prolonged residence inside the reactor.

Based on the experience of previously existing experimental irradiation devices (hydraulic rabbit, PRF) a closed pressurized water circuit was chosen as basic design. This loop is cooled by the primary cooling water of the BR2 reactor and the water temperature would be roughly 10”C above the primary cooling circuit temperature. The internal pressure of the loop equals that of the reactor cooling circuit in order to allow the use of light alloys and small wall thicknesses. The device has to be protected against local overheating in case of accidental ‘blockage of the cooling duct. Therefore, a bypass-cooling with a heat dissipation capacity exceeding the critical heat flux has to be foreseen. Different experimental arrangements are shown on Fig. 5, which displays four different target-fuel assembly configurations within a VIG and a VG fuel element.

To perform scouting neutronic computations it was decided to consider two target diameters for Am, Np and I : 0.836 and 1.696 cm depending on the loading within a VIG or a VG fuel element. For Tc, the target diameters considered were 1.170 and 2.030 cm.

The bottom parts of the Tables IV and V show the results of the thermal computations of the linear fission power and the centre-line temperature of targets loaded with Am and Np. The Am targets with a large diameter must be limited in Am content in order to avoid target centre melting.

Furthermore, it can be stated that below 400 W/cm, no change in geometrical stability is expected for Am and Np targets while below 150 W/cm no accidental events can occur.

For CaI_2 targets, the main point is to avoid any volatilization of iodine or iodine compound. Therefore the centre-line temperature has to be kept below 700°C and consequently the maximum linear power must be lower than 40 W/cm. Tests on the geometrical stability of $Ca1_2$ are of utmost importance before starting an isotopic transmutation programme.

5. IRRADIATION CONDITIONS AND CALCULATED TRANSMUTATION YIELDS

Transmutation reactions of Np-237 and Am-241 by thermal neutron absorption yield during extended irradiation programmes of several years essentially Pu-238 with a half-life of 88 years and fission products. Am-243 is slowly transformed into Cm-244 with a half-life of 18 years.
Neutron capture of Tc-99 yields inactive Ru-1OO while 1-129 transforms into gaseous Xe-130. (See further, Table VIII).

The neutron calculations were carried out, in one-dimensional cylindrical geometry, with following codes and neutron libraries :

- for the Am and Np targets :
  with the ANISN neutron transport code [13] (part of the MARS-AMPEX code system [14]), with a coupled fast-thermal 27 group library obtained by group-collapsing (from 218 to 27 groups) from [15].
  In the 218 and 27 group libraries, self-shielding corrections in the resonance region are allowed for (Nordheim formalism, with the NITAWL code) for the nuclides Am-241 and Np-237 (not for Am-243).

- **for the I and Tc targets :**
  with a S.C.K./C.E.N. version of the DTF-IV neutron transport code [16] (part of the MULCOS code system [17]), with the coupled fast-thermal 40 group library [18]. The 40 group cross-section sets (without resonance self-shielding correction) for 1-129 and Tc-99 were calculated at S.C.K./C.E.N. starting from the resonance parameters given in [19].

The composition of the reference targets is shown in Table 111.

TABLE 111: PROPOSED TARGET COMPOSITIONS"

IRRADIATED NUCLIDE	BULK DENSITY	TARGET COMPOSITION
Am-241 Am-243	75 % T.D.	$Am_2O_3 + Al_2O_3$ weight ratio Am-241/Am-243 = 3.53 vol. ratio $Am_2O_3/(Am_2O_3 + Al_2O_3)$ considered : 10; 50; 90 %
Np-237	75 % T. I).	$NpO_2 + Al_2O_3$ vol. ratio $NpO_2/(NpO_2 + Al_2O_3)$ considered : 10; 50; 90 %
1-129	70 % T.D.	CaI_2
Tc-99	`'75 % T.D.	Metallic technetium

The target length adopted in the present study is 60 cm. This length has been chosen for the following reasons :

- 60 cm corresponds to the fuel length of many MTRs;
- BR2 has a fuel length of 76.2 cm, leading to an extrapolated core height of about 90 cm. As a result, the ratio (axially averaged value/maximum) for a cosine-shaped axial flux profile amounts to 0.827 for a 60 cm target, which remains sufficiently near to unity to be acceptable.

In addition, the 60 cm target could possibly be split into two 30 cm capsules, which could regularly be inverted, yielding a practically uniform transmutation over the whole irradiated target. length.

The target diameters considered are, as indicated above :

- for Am, Np, I (double barrier irradiation) :
  0.836 cm in a VIG element
  1.696 cm in a VG element
- for Tc (single barrier irradiation) :
  1.170 cm in a VIG element
  2.030 cm in a VG element.

As already explained, the cladding material adopted for all targets is zircaloy-4.

The irradiation conditions adopted for the neutron irradiation in BR2 are as follows :

- the clad target and the surrounding aluminium tubes are introduced into a VIG or VG type fuel element with a burn-up of 40 %;
- the surroundings of the channel containing this VIG or VG fuel element are arranged in a hexagonal configuration ("crown") of channels of which 4 contain fuel elements of the VIG type with 20 % burn-up and 2 contain a beryllium reflector plug;
- the power of this hexagonal "crown" amounts to 8 MW;
- the irradiation duration is assumed to be 200 EFPD (= 1 calendar year).

This corresponds practically to an irradiation in a BR2 channel such as. C-41, C-101, B-240, C-259, B-300, C-319 etc. on Fig. 1.

The target volumes, the quantities of actinides or fission products contained in one target before irradiation, the percentage transmutation obtained after 200 EFPD and the corresponding quantities of actinides or fission products transmuted per target are indicated in Tables IV, V and VI for americium, neptunium and fission product targets, respectively.

The percentages and quantities of Am-241, Am-243 and Np-237 transmuted after 200 EFPD are indicated' in Fig. 6, as a function of the vol.% Am_2O_3 (or NpO_2) in the $Am_2O_3 + Al_2O_3$ (or $NpO_2 + Al_2O_3$) matrix.

TABLE IV : CALCULATED TRANSMUTATIONS IN THE AMERICIUM TARGETS; CALCULATED FISSION POWER VALUES AND DEDUCED TEMPERATURES

TARGET COMPOSITION	Am_2O_3 - Al_2O_3					
	10-90	50-50	90-10	10-90"	50-50	90-10"
target diameter (cm)	0.836			1.696		
target length (cm)	60					
target diameter (cm^3)	32.935			135.548		
g in target						
Am-24 1	20.45	102.3	184.1	84.17	420.9	757.6
Am-243	5.790	28.95	52.11	23.83	119.2	214.5
% transmuted after 1 year (200 EFPD) irradiation						
Am-241	86.3	513	36.6	70.5	30.6	20.2
Am-243	493	22.6	15.6	36.2	13.4	8.90
g transmuted after 1 year (200 EFPD) irradiation						
Am-241	17.7	52.5	67.3	593	129	153
Am-243	2.86	6.55	8.13	8.63	15.9	19.1
absorption rate depression *						
Am-241	1.42	3.40	5.56	2.02	7.17	10.9
Am-243	1.41	3.27	4.18	2.04	5.16	6.53
linear fission power (W/cm) (AM-241 + AM-243)	38.8	128	204	122	421	700
fission rate depression * (Am-241 + Am-243)	1.21	1.42	1.41	136	1.49	1.43
target surface temperature (K)	368	470	559	401	595	774
target axis temperature (K)	457	800"	1158	704	2090"	above melting point

* depression : value at target surface (outer radius) /value in target axis.

TABLE V : CALCULATED TRANSMUTATIONS IN THE NEPTUNIUM TARGETS; CALCULATED FISSION POWER VALUES AND DEDUCED TEMPERATURES

TARGET COMPOSITION"	NpO_2 - Al_2O_3					
	10-90	50-50	90-10	10-90"	50-50"	90-10"
target diameter (cm)	0.836			1.696		
target length (cm)	60					
target volume (cm^3)	32.935			135.548		
g in target Np-237	24.18	120.9	217.6	99.s1	497.5	895.6
% transmuted after 1 year (200 EFPD) irradiation Np-237	57,3	36.4	27.4	48.1	23.8	16.6
g transmuted after 1 year (200 EFPD) irradiation" Np-237	13.9	44.1	59.7	47.8	118	149
absorption rate depression * Np-237	1.09	1.s3	1.98	1.22	2.22	3.13
linear fission power (W/cm) Np-237	16.1	78.1	140	61.4	296	535
fission rate depression * Np-237	0.999	0.998	0.995	1.001	0.996	0.988
target surface temperature (K)	341	413	484	362	514	668
target axis temperature (K)	378	611	880	510	1495	above melting point

*** depression : value at target surface (outer radius)/value in target axis**

TABLE VI : CALCULATED TRANSMUTATIONS IN THE FISSION PRODUCT TARGETS; CALCULATE I) TEMPERATURES

TARGET COMPOSITION	CaI_2		Tc	
target diameter (cm)	0.836	1.696	1.170	2.030
target length (cm)	60		60	
target volume (cm^3)	32.935	135.548	64.509	194.193
g in target 1-129 Tc-99	78.94	324.9	522.5	1573
% transmuted after 1 year (200 EFPD) irradiation 1-129 Tc-99	8.6- 9.5**	6.8 -7.6	3.4 -6.1	2.2 -4.1
g transmuted after 1 year (200 EFPD) irradiation. 1-129 Tc-99	6.8 -7.5	22-25	18 - 32	34-64
absorption rate depression * I-129 Tc-99	1.06-1.05	1.13-1.12	133-1.98	1.70- 2.80
target axis temperature (K)	< 1000	< 1000		

* depression : value at target surface (outer radius)/value in target axis

** 1st value : thermal neutron contribution

2nd value : thermal neutron contribution + epithermal neutron contribution, without resonance self-shielding correction.

6. BR2 IRRADIATION CAPABILITY

Am and Np may be burnt in a target having diameter 1.696 cm or 0.836 cm. The above calculations have given the burn-up of the targets after 200 EFPD. If the thermal neutron flux is ϕ = 2.OE+14 n/cm²s in the fuel elements at the mid-plane, we obtain $\sigma_f.\phi.t$ = 1.963. This gives a non-negligible anti-reactivity in a core loading containing 10 kg U-235. Taking into account the external leakage of neutrons (β_{eff} = O.0072 and κ_∞ = 1.30) and weighting the axial thermal neutron flux and the importance function on a fresh fuel element and on the target length, we obtain in a mean channel (Wi = 1.0) :

	diam. :1.696 cm			diam. :0.836 cm		
	Am-241	Am-243	Np-237	Am-241	Am-243	Np-237
mass loaded(g)	420.9	119.2	497.5	184.1	52.11	217.6
vol. actinide in target	50		50	90		90
% transmuted after 200 EFPD						
mean	30.6	13.4	23.8	36.6	15.6	27.4
max.	35.7	16.0	28.0	42.4	18.5	32.1
anti-reactivity ($)	0.519 +	0.065	0.490	0.270 +	0.033	0.246

An experimental target with diameter 1.696 may be loaded in a BR2 fuel element of type VG (5 plates) positioned in a channel C (Wi = 0.9).

For a massive transmutation of Am and Np in standard fuel elements of type VIG (6 plates) it should be noted that :

1) the **permission has to be obtained to cool the targets directly by the primary circuit of the reactor BR2; this** permission may be obtained after a successful experimental programme.

2) the number of targets is routinely limited to 6-8 $ of antireactivity depending on the burn-up at elimination of the fuel elements (50-40%) [20].

This allows the irradiation of 12 targets of large diameter 1.696 cm in the channels C (Wi = 0.9) and D (Wi= 0.7), (whilst the channels A and B in the central ring have a statistical weight of about Wi = 1.5) or of 24 targets of small diameter 0.836 cm in the channels A, B, C, D. In both cases the consumption rate of Am and Np will be equivalent.

Advanced driver fuel elements of the type VIH containing 520 g U-235 instead of 400 g U-235 could be fabricated in the future to load 24 targets of large diameter with 50 vol.% actinide content in the channels A, B, C, D. In that case 3.5 kg Am or 2.8 kg Np could be burnt yearly in BR2.

7. DISCUSSION

The thermal data of tables IV and V indicate that the highest acceptable concentration of Am resp. Np in a 1.696 cm diameter capsule is a 50/50 ratio of actinide to aluminium. In small diameter targets (O. 836 cm) no critical temperature level is attained but the quantities which can be irradiated per target are 4 times smaller.

As was to be expected, the percentage of actinide transmuted per target in 200 EFPD decreases as actinide/aluminium ratio and target size increase. The percentages of Am-241 and of Np-237 transmuted per 200 EFPD are roughly the same and range from 27 to 86 % in the small targets and from 24 to 70 % in the large targets. The Np-237 and Am-241 content of a target can be depleted in a few years of irradiation.
The Am-243 depletion is smaller because of the lower neutron absorption cross-sections.

The transmutation yields of 1-129 and Tc-99 are shown in Table VI and appear to be very small due to the very low neutron capture cross-sections.

The use of a high flux reactor for target transmutation studies has to be viewed from the overall mass balance in the reactor fuel. Table VII [21] shows the quantities of fissile material depleted and actinides + LLFP formed per reactor fuel element and in the whole reactor core operating during 200 EFPD.

By comparing these data with the projected depletion in the target capsules it appears that more Tc-99 and 1-129 are formed in the U-235 fuel than can be transmuted in the target.
However, the reflector of the BR2 reactor provides the opportunity to irradiate in 20 channels (with 84 mm diameter) where the unperturbed thermal neutron flux amounts to about 3E+14 n/cm2s.
These positions can be used for irradiation of targets with low absorption cross-sections such as iodine and technetium.
No calculation has been made for these cases but one can assume that about 400 g of iodine can be transmuted per year.

TABLE VII : URANIUM BURNUP AND NUCLIDE YIELDS IN BR2 FUEL ELEMENTS (grammes) [21]

Nuclide	Scenario : 50% burn-up at discharge (5 cycles) 3.5 years cooling time	
	per VIG fuel element	in 60...75 VIG fuel elements per year
U-235	200	12000...15000
Np-237	4.235E-1	25..32
AM-241	2.696E-3	0.16...0.20
Am-243	4.071E-5	0.002...0.003
Tc-99	4.127E0	248..310
1-129	8.903E-1	53...67
U-235 disappeared	200	12000...15000

On the other hand the Np-237 and Am-241/243 formation in the reactor fuel is very small compared to the potential transmutation yield in the actinide target capsules.

Per irradiation position a maximum of 540 g Am and 500 g Np can be loaded into one target capsule. The depletion per 200 EFPD is 30 % for Am-241 and 24 % for Np-237, i.e. 129 g resp. 118 g are transmuted per year in one target.

The number of target capsules which can be loaded into the reactor is limited by the antireactivity of the targets and the number of available positions (24). In the case of BR2, a total antireactivity of 6 to 8 $ can be accepted in the reactor at the present time, while each capsule produces an antireactivity of about 0.5 $.

Under these circumstances, a maximum of about 12 targets can be loaded with a total mass of about 6 kg Am or Np or a mixture of both. The transmutation capacity of the reactor if it were dedicated to such R&D programmed is about 1.5 kg Am or Np per year.

As indicated in Table VIII, Am-241 is transmuted into Am-242 (about 10 % into the 141 y isomer, about 90 % into the 16 h isomer); hence about 90 % is finally transmuted into Cm-242, which, with a half-life of 163 days, decays into Pu-238.
Am-243 is transmuted into Am-244 which decays rapidly into Cm-244 (18 y).
Np-237 is transmuted into Np-238 which decays (with a half-life of 2.1 days) into Pu-238.
Hence the bulk of the two actinide target types considered in this paper is ultimately transmuted into Pu-238.

The 2200 m/s cross-sections (σ) and the resonance integrals (RI) of the main nuclides involved in these transmutations are indicated in Table IX [19].

The "half-life" of both Am-241 and Np-237 equals 2 years or even less, depending on the type of target, when irradiated in the BR2 reactor.

TABLE VIII : MAIN TRANSMUTATION REACTIONS (simplified) OCCURRING IN THE TARGETS STUDIED IN THIS PAPER

Am-241	$(n_f y)$	Am-242 m	(n, γ)	Am-243	
	≈ 10%				
		↓ 141 y			
			16h	163d	
	(n, y)	**Am-242 g**	→	**Cm-242** →	Pu-238
	≈ 90%				
	(n, f) FP				
			26m...10h		
Am-243	(n, y)	Am-244	→	Cm-244	
	(n, f) FP				
			2.1d		
Np-237	$(n_r y)$	Np-238	→	Pu-238	
	(n, f) FP				
			12h		
1-129	**(n, y)**	**1-130**	→	Xe-130	
			16s		
Tc-99	$(n_r y)$	Tc-100	→	Ru-100	

TABLE IX :2200 m/s CROSS-SECTIONS AND RESONANCE INTEGRALS OF THE MAIN NUCLIDES INVOLVED IN THE TRANSMUTATION REACTIONS STUDIED IN THIS PAPER [19]

NUCLIDE	t½	REACTION TYPE	σ (barns)	RI (barns)
Am-241	433 y	n,γ tot	587	1425
		n,γ -152 y Am-242m	54	195
		n,γ → 16.1 h Am-242g	533	1230
		n,f	3.2	14.4
Am-243	7370 y	n,γ tot	75.1	1820
		n,γ .26 m Am-244m		
		n,γ -10.1 h Am-244g	3.8	94
		n,f	0.1983	9
Cm-242	163 d	n,γ	16	110
	. .	n,f	< 5	
Cm-244	18.1 y	n,γ	15.2	650
		n,f	1.04	12.5
Np-237	$2.14\ 10^6$y	n,γ	175.9	640
		n,f	21.5	6.9
Pu-238	87.7 y	n,γ	540	33
		n,f	17.9	162
1-129	$1.6\ 10^7$y	n,γ tot		36
		n,γ -9.2 m I-130m	18	
		n,γ - 1236 h I-130g	9	
Xe-130	St	n,γ tot	< 26	
		n,γ -11.8 d Xe-131m	0.45	16
		n,γ - st Ce-131g		
Tc-99	$2.14\ 10^5$y	n,γ	20	340
Ru-100	St	n,γ	5.0	11.2

8. CONCLUSIONS

The high flux reactor BR2 with a routine thermal output of 60 MWth has a limited irradiation potential for 12 targets of 500 g (Np + Am) each and a transmutation throughput of about 1.5 to 1.7 kg (Np + Am) per year. This transmutation capability can be used for investigating at the technological scale the formation of transmutation products (Pu-238, Pu-239, FP.. .) in a thermal neutron spectrum with a high contribution of epithermal and fast neutrons as well as the metallurgical behaviour of the targets. With upgraded fuel the irradiation potential could be increased up to, maximum, 24 targets.

The quantities which are potentially produced by the reprocessing plants (3100 kg (Np + Am) per year) would require extensive irradiation facilities amounting to about 110 GWth high flux reactor capacity and consume 27E+3 kg U-235 per year (200 EFPD).

The "half-life" of Np and Am in the BR2 reactor is about 2 years in routine operation, which means that a target has to be irradiated 10 years to yield a 97 % depletion. This period can be reduced by incorporating a target reprocessing step after e.g. 2 or 3 years irradiation.

The irradiation of 1-129 as CaI_2 and Tc as metal does not provide a rapid transmutation (5.8 % per year) but due to the occurrence of a high thermal flux in the reflector, the targets may stay in the peripheral positions of the reactor for a very long time till depletion.
The 1-129 targets must be equipped with a Xe venting system which has to be carefully protected against any temperature excursion to avoid iodine contamination and corrosion of the venting circuits.

The Tc-99 targets do not require any special monitoring since the end-product Ru-100 has nearly the same physico-chemical properties as the target nuclide.

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CONFIGURATE 12H

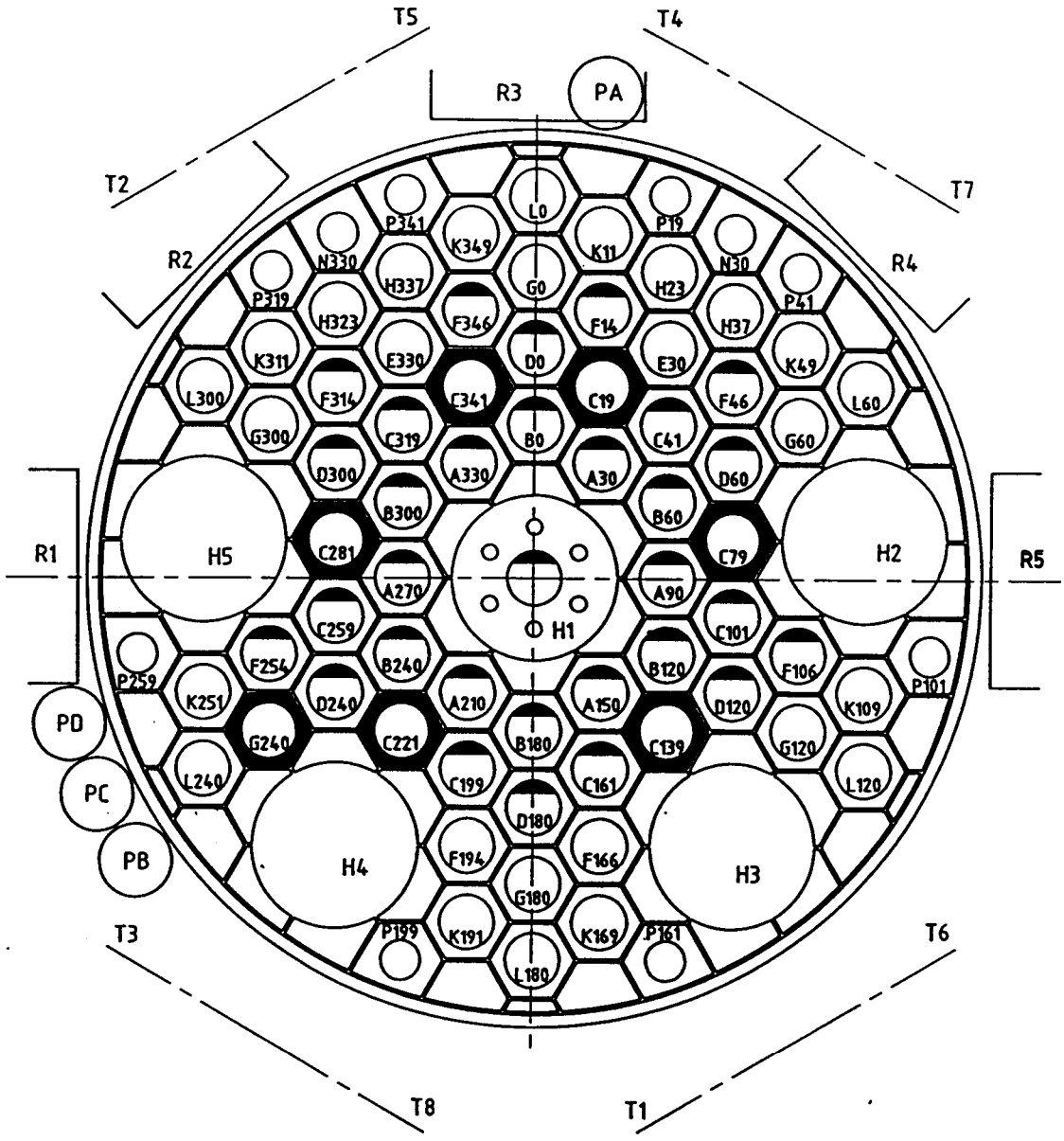

FIG. 1: BR2 core : Cross-section at reactor mid-plane with a typical loading

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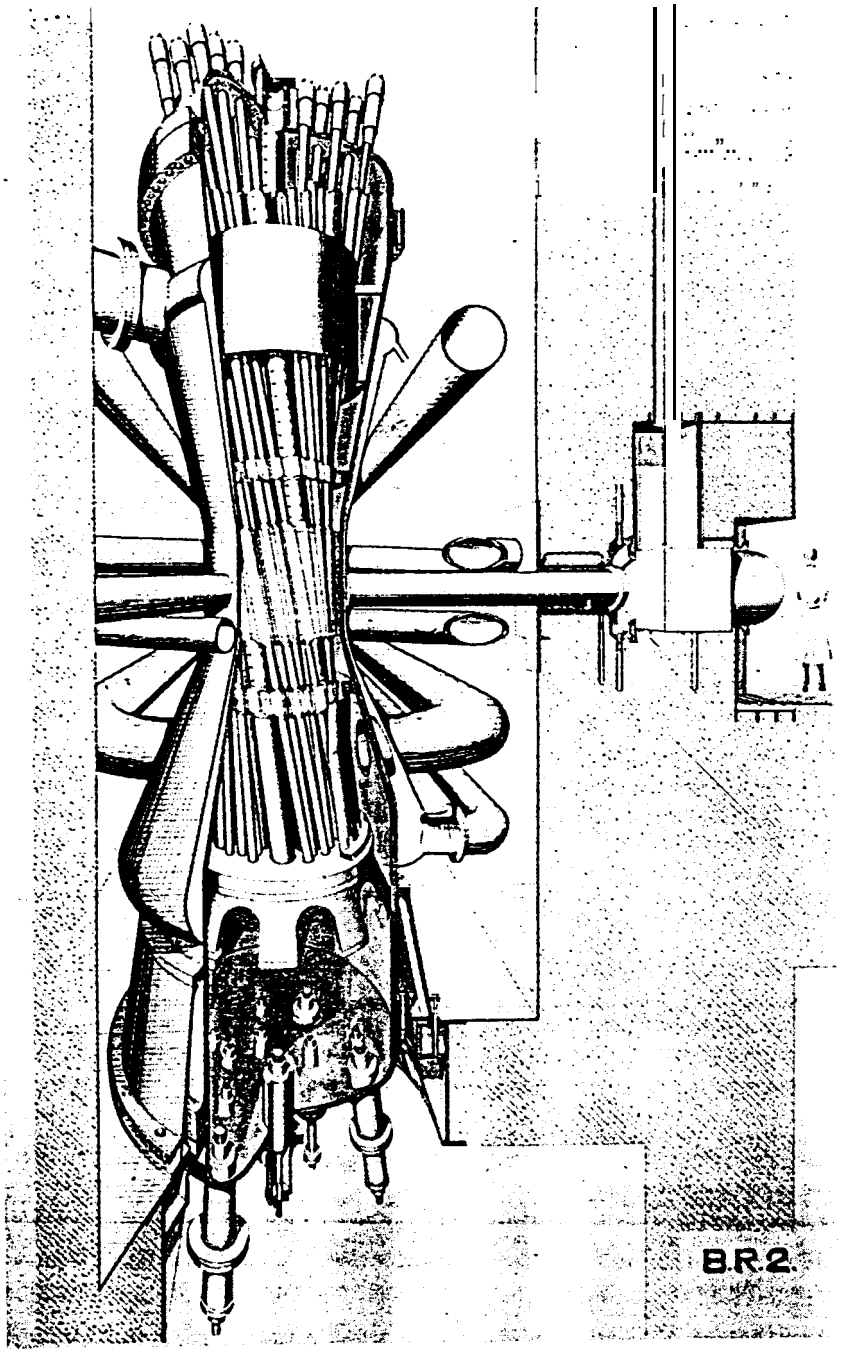


FIG. 2: General view of the BR2 reactor

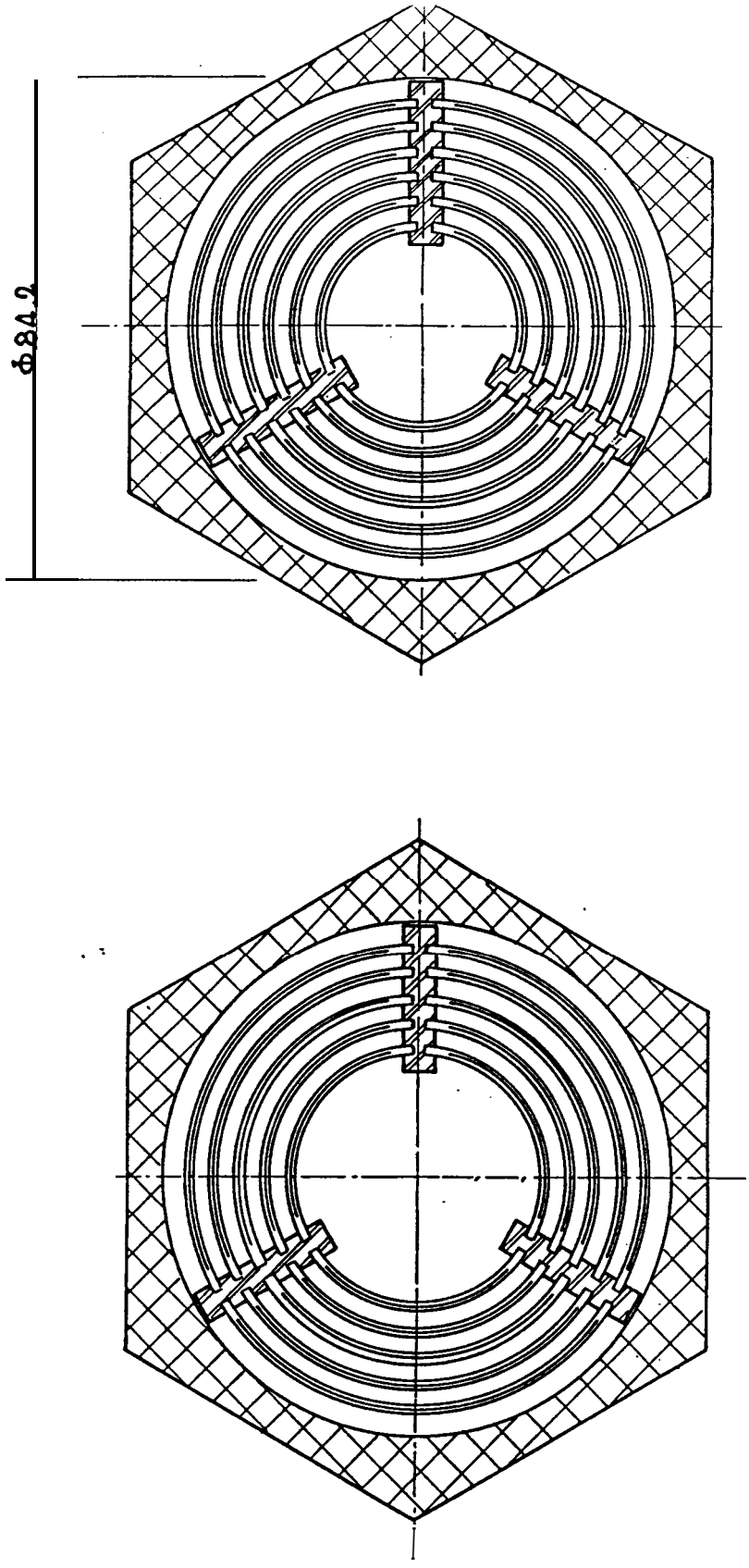


FIG. 3: BR2 six-plate (VIG) and five-plate (VG) fuel elements in standard channels

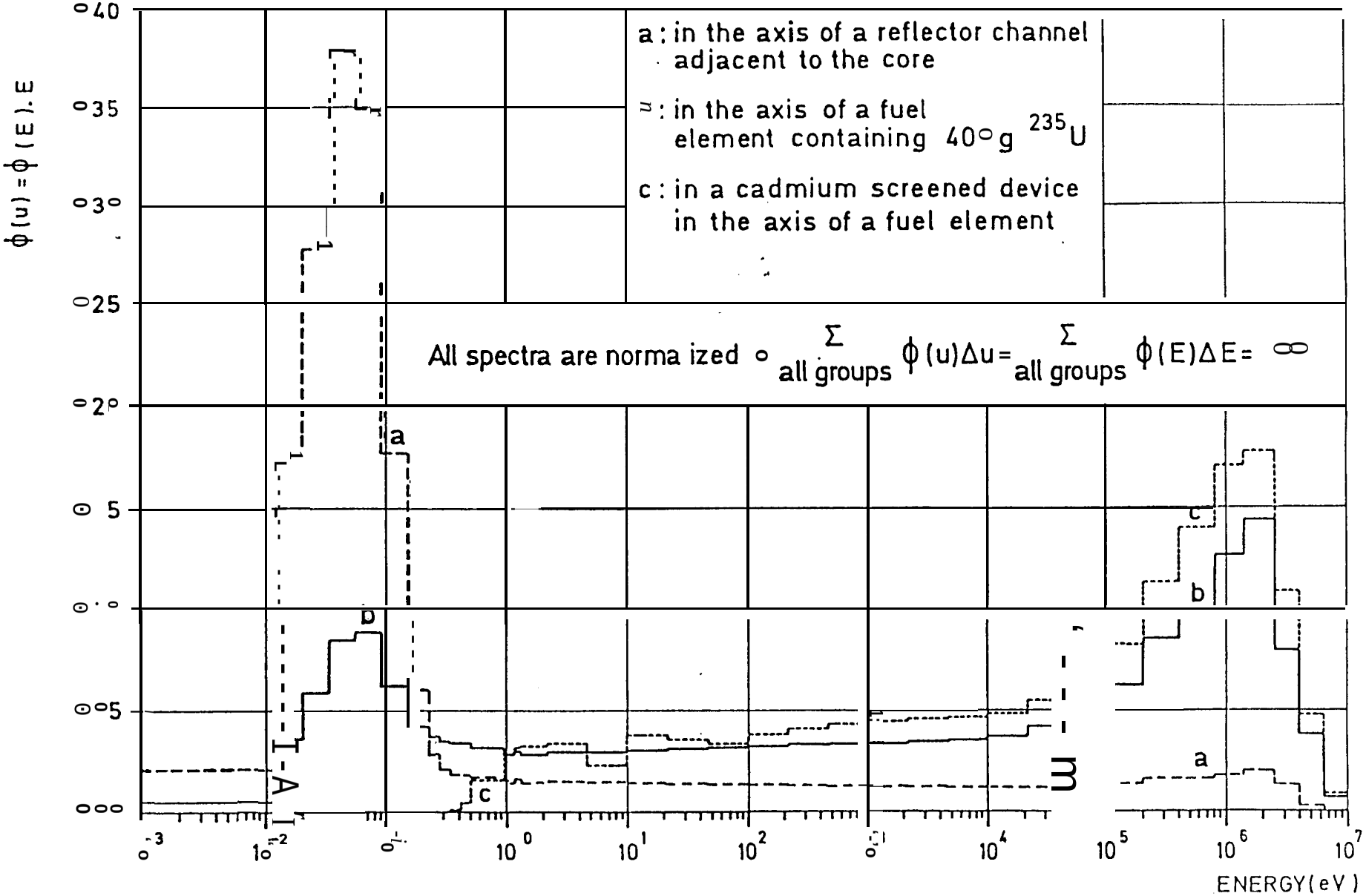
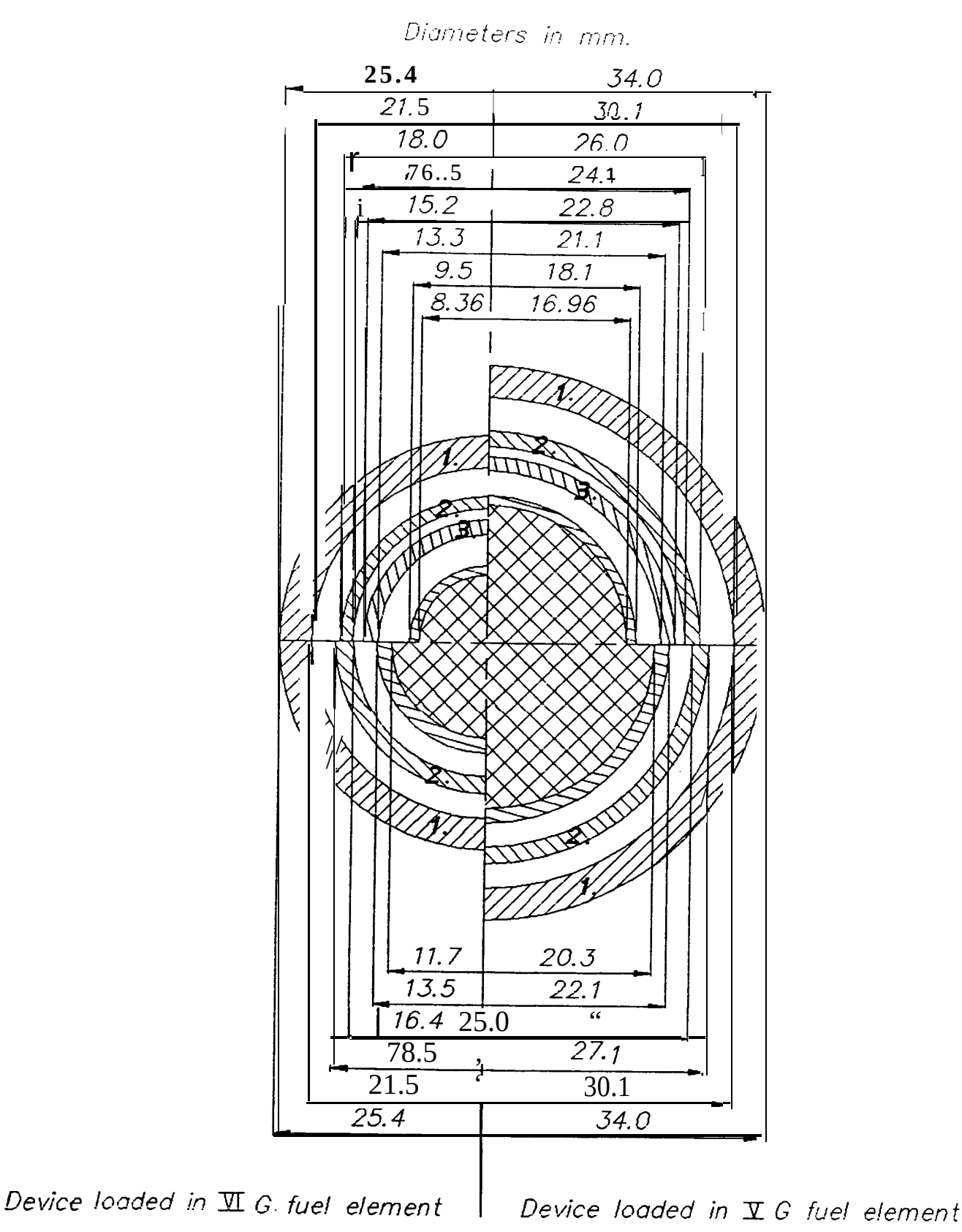
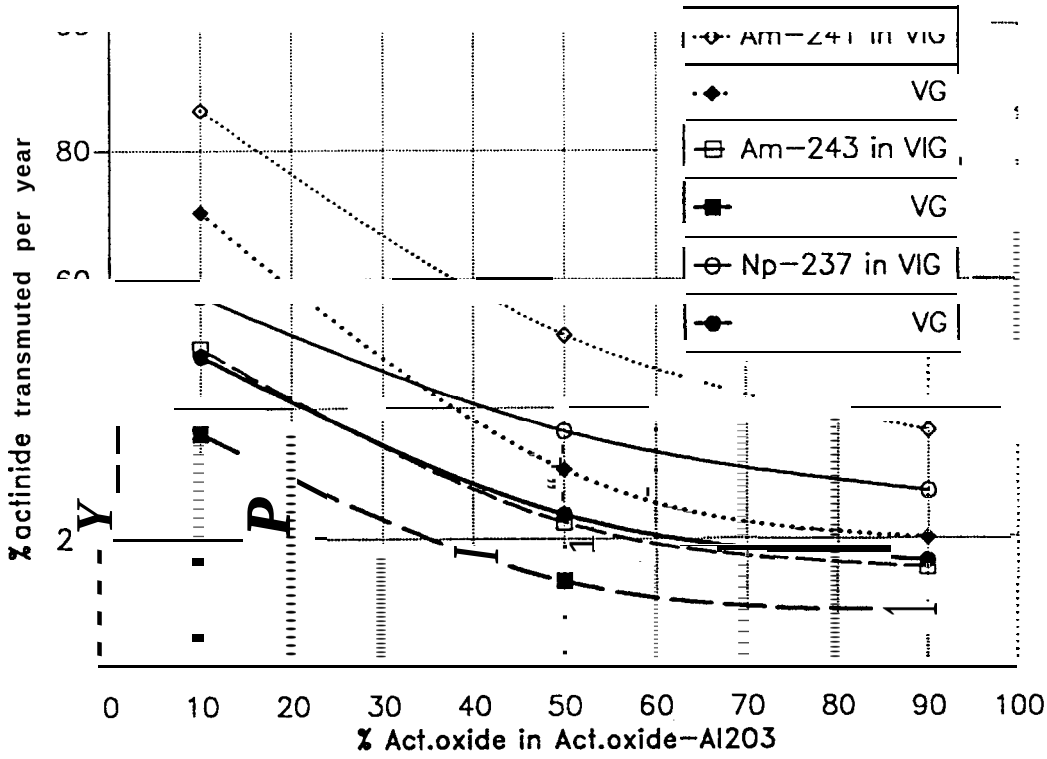


FIG. 4 : Typical neutron flux spectra in BR2



1. Pressure tube.
2. Tube separating up and down flow.
3. Tube separating by-pass and up flow.

FIG. 5 : **Cross-section of the experimental devices loaded in BR2 fuel elements VIG (with six plates) and VG (with five plates)**



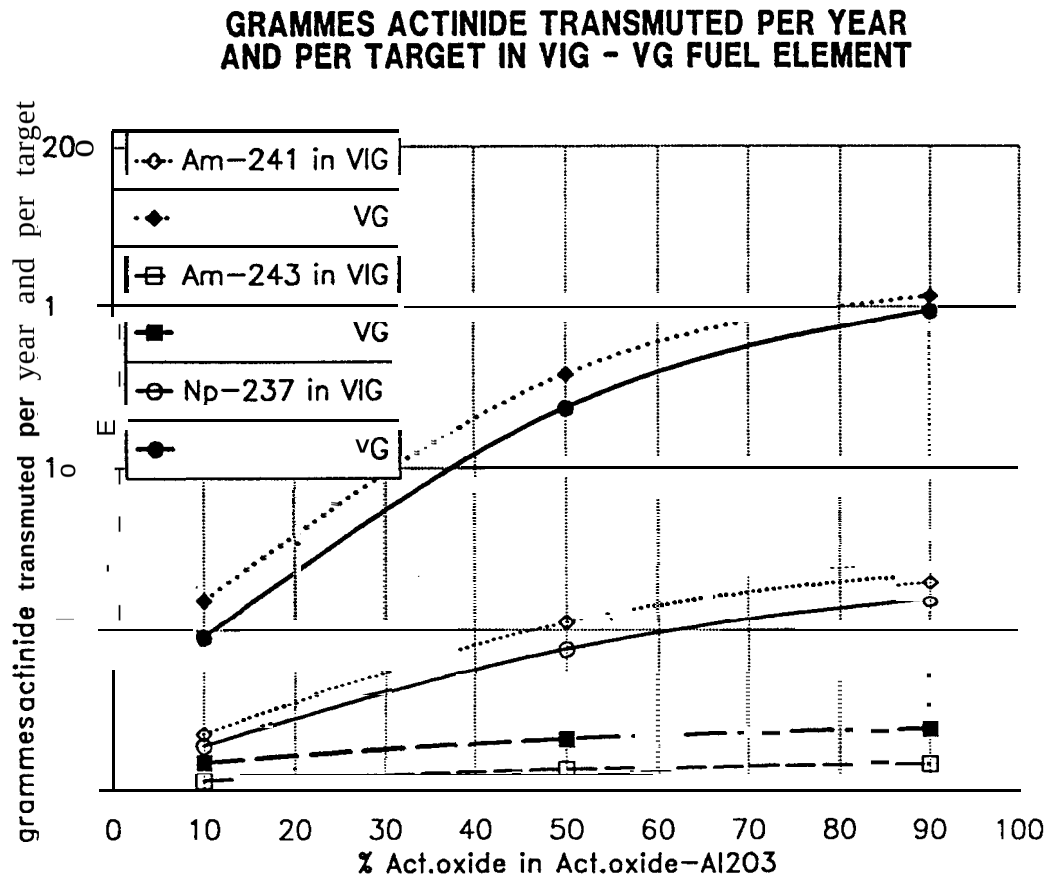


FIG. 6 : Percentages and quantities of Am-241, Am-243 and Np-237 transmuted after 200 EFPD irradiation in BR2