STATUS OF TRANSMUTATION STUDIES IN A FAST REACTOR AT JNC

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Abstract

This paper presents the status of studies on transmutation of minor actinides (MA) and long-lived fission products (LLFP) in a fast reactor at JNC, which includes four areas of work: (a) design studies, (b) nuclear data measurement and evaluation, (c) fuel fabrication and irradiation test, (d) strategy studies. It was found that the mixed hybrid MA-loading method, where Np nuclide is dispersed uniformly in the core and target subassemblies containing Am, Cm and rare earth nuclides are loaded into the radial blanket region, has a great potential to achieve effective transmutation of MA without serious drawbacks in terms of core performance. Fission cross section ratios of minor actinide nuclides (^{237}Np, ^{241}Am, ^{243}Am, ^{242m}Am, etc.) in the fast and epithermal neutron energy region have been measured to evaluate the accuracy of MA nuclear data. As for fabrication and investigation of irradiation behaviour of MOX containing MA, the systematic program has been planned. The irradiation test of Np- and Am-contained MOX fuel is planned in JOYO. In step with the JOYO MK-III schedule, the irradiation test will be initiated from around 2003.

Introduction

Japan Nuclear Cycle Development Institute (JNC) was organised to establish the nuclear fuel cycle in Japan in 1st October 1998. JNC is now conducting research and development on FBRs and nuclear fuel reprocessing technology to achieve this objective.

One of the distinctive features of a fast reactor is its good neutron economy. Utilising the excess of neutrons enables us to construct flexible cores such that they incinerate minor actinides(MAs) and long lived fission products (LLFP) to reduce radiotoxicity and breed or burn Plutonium in consideration of Plutonium balance.

Some of the MA nuclides (Np, Am, Cm) contained in residual waste from reprocessing have extremely long-lived radiotoxicity [1]. Means of reducing the radiotoxicity of the MA nuclides are presently under investigation. The MA nuclides could produce useful energy if converted into short-lived fission products by neutron bombardment. From this standpoint, a nuclear reactor provides the obvious means for transmutation of MA nuclides. Among the various nuclear reactors, a fast reactor is considered to have the greatest potential to transmute MA effectively, because of its hard neutron spectrum [2-6].

Transmutation of ^{99}Tc and ^{129}I by neutron capture as a result of irradiation in nuclear reactors will yield the stable isotopes ^{100}Ru and ^{130}Xe, respectively. However, due to the small neutron cross sections, the transmutation efficiency in LWRs is low. Moderated subassemblies in fast reactors are more appropriate devices for the transmutation of the fission products [6-8].

This paper presents the status of studies on transmutation of minor actinides (MA) and long-lived fission products (LLFP) in a fast reactor at JNC , which includes four areas of work : (a) design studies, (b) nuclear data measurement and evaluation, (c) fuel fabrication and irradiation test, (d) strategy studies.

Feasibility studies on MA transmutation

Feasibility studies have been performed to investigate the basic characteristics (transmutation rate, burn-up reactivity, Doppler coefficient, sodium void reactivity, maximum linear heat rate, etc.) of a fast reactor core with MA transmutation, the following items were considered:

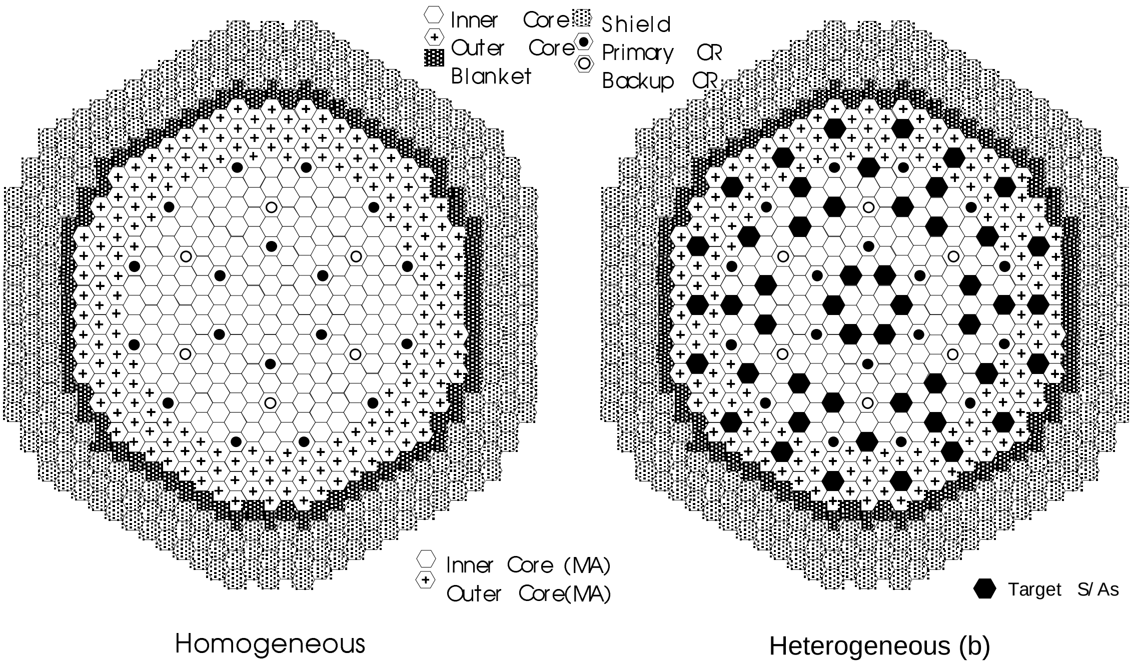
1. Study on loading method of MA in the core (homogeneous, heterogeneous, hybrid, blanket, etc.).
2. Selection of fuel material for MA transmutation (oxide, inert matrices such as Al_2O_3, CeO_2, etc.)
3. Study on the maximum tolerable amount of rare earth (RE) nuclides.
4. Effect of MA recycling on core characteristics and fuel cycle system.
5. Influence of uncertainties of MA nuclear data.
6. Influence of MA containing fuel on reactor plant and fuel cycle.

The main results of the studies are summarised as follows.

Study on MA loading method

The MA transmutation in a fast reactor core has no serious drawbacks in terms of core performance, provided that the homogeneous loading methods shown in Figure 1 can be employed with a small ratio of MA to fuel (~5% wt). Since a 1 000 MWe-class LWR produces about 26 kg of MA per year, a fast reactor with 5%wt MA loading can transmute the MA mass from six LWRs.

Figure 1. **MA loading method**



The heterogeneous MA loading method as shown in Figure 1 can be made feasible by optimising the fuel design, loading pattern and the coolant flow of the MA-loaded fuel subassemblies. The reduction of the fuel pin diameter and the Pu enrichment is essential to reduce the power of MA-loaded fuel in the heterogeneous MA loading method.

The hybrid MA loading method, where Np nuclide is dispersed uniformly in the core and target subassemblies containing Am, Cm and rare earth nuclides are loaded into the radial blanket region, can transmute a large amount of MA without serious drawbacks in terms of core performance. The transmuted mass of MA is about 530 kg/cycle as shown in Table 1, which is almost 16 times the mass produced by an LWR of the same power output.

The MA loading in the blanket region causes no problems from the viewpoint of core performance. Minor actinides are transmuted at a rate of 6% per cycle in the axial and radial blanket regions.

It was found that the hybrid MA loading method has the potential to achieve the maximum transmutation of MA with no special design considerations.

Table 1. **Comparison of core performance for various MA loading methods**

Item	Reference (No MA)	Homogeneous loading	Heterogeneous loading	Homogeneous loading	Hybrid loading
MA and RE loaded in the core region	–	Np,Am,Cm: 5% RE: 0%	Np,Am,Cm: 49% RE: 0% (Number of target S/As:39)	Np,Am,Cm: 5% RE: 10%	Np: 9.8% RE: 0%
Matrix of target	–	–	UO_2	–	Al_2O_3
MA and RE loaded in the blanket region	–	–	–	–	Am,Cm: 46% RE: 46% (Number of target S/As: 72)
Core height (cm)	100	100	100	100	100
Cycle length (days)	456	456	456	456	456
Number of batches	3	3	3	3	3
Pu enrichment (inner core/ outer core) (wt%)	15.4/18.6	16.6/20.1	15.4/18.6	20.0/24.2	19.0/23.4
B.U. reactivity loss (%δk/kk')	3.31	2.12	1.83	3.71	0.90
Max. linear heat rate (driver/target) (W/cm)	420	407	439/309	413	406/174
Void reactivity	1.0	1.3(1)	1.3(1)	1.4(1)	1.5 (1)
Doppler coef.	1.0	0.6(1)	0.7(1)	0.5(1)	0.45 (1)
MA transmutation Amount (kg/cycle) Rate(%/cycle)	– –	172 10.9	186 11.3	164 10.3	529

(1) Relative values.

Selection of fuel material for MA transmutation

Different types of inert matrices, instead of UO_2, for the heterogeneous MA-loading method have been investigated, they avoid the buildup of higher actinides via ^{238}U and achieve a high MA transmutation rate. Inert matrices of Al_2O_3 and CeO_2 were examined in this study. The MA transmutation rate of the target subassembly using inert matrices is larger than that of the target subassembly using UO_2 The use of inert matrices in the target subassembly effectively increases the MA transmutation rate.

Study on the permissible re level in homogeneously loaded MA

Systematic parameter survey calculations were performed to investigate the basic characteristics of a fast reactor core loaded homogeneously with MA which contains RE, and also to establish a MA and RE loading method which has no serious influence on the core design. The homogeneous loading of MA and RE has no serious effects on the reactor core performance, provided that the amounts of MA and RE in the fuel are less than 5 and 10wt% respectively. In the case of adding Am, Cm and RE in the radial blanket region, it is possible, from the viewpoint of core performance, to insert ~50wt% of Am and Cm, and ~50wt% of RE in the target assemblies.

Effect of MA recycling on core characteristics and fuel cycle system

The effects of MA recycling on the core characteristics and the fuel cycle system in the homogeneous loading method were evaluated. The recycling of MA in a fast reactor is feasible from neutronic and thermal-hydraulic points of view. However, during multiple recycling the Np fraction is significantly reduced compared to the unirradiated feed, and the fraction of Cm is greatly increased because of neutron capture in Am. The accumulation of Cm as a result of the MA recycling will bring about some problems concerning fuel handling and reprocessing, because of an increase in both the decay heat and the neutron emission rate from ^{244}Cm.

Influence of uncertainties of MA nuclear data

The influences of the uncertainties upon nuclear characteristics were evaluated for a large LMFBR core loaded with MA of 5%. Sensitivity analysis on cross sections was carried out and uncertainties of nuclear characteristics were roughly evaluated. Uncertainties of nuclear characteristics are rather large compared with those of conventional cores. Some cross sections of minor actinides (^{237}Np, ^{241}Am, ^{238}Pu, ^{243}Am and ^{244}Cm) need to be improved.

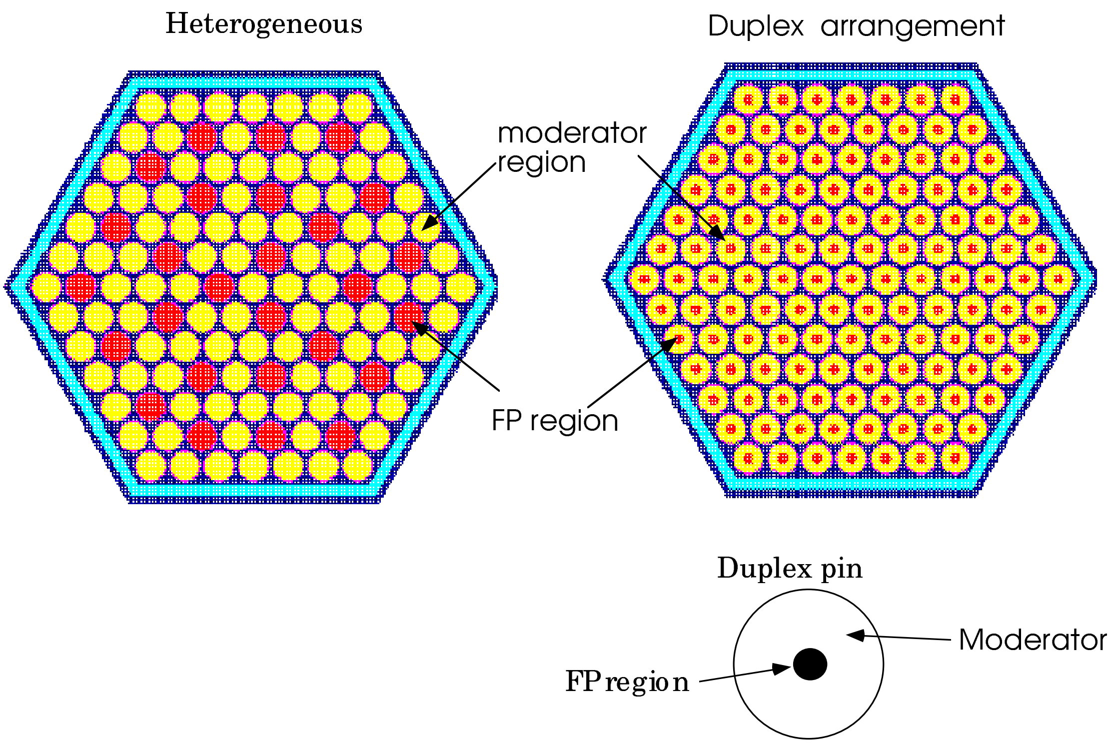
Influence of MA containing fuel on reactor plant and fuel cycle

Both the decay heat and neutron emission rate of the MA (Np, Am and Cm)-loaded fuel are very large in comparison with MOX fuel without MA. The dominant element of these fuel properties is 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.

Feasibility studies on FP transmutation

A moderated target subassembly was used for FP transmutation. The subassembly consists of moderator pins and FP target pins distributed between the moderator pins. The moderated target subassemblies were loaded in the radial shield region of the fast reactor. A new concept of duplex pellets was also examined: a moderator annulus surrounding a ^{99}Tc core, as shown in Figure 2, adopted to get a better ^{99}Tc transmutation performance.

Figure 2. **Configuration of moderated target sub-assemblies**



Systematic parameter survey calculations were performed to investigate the basic characteristics of FP transmutation in the blanket region of a fast reactor. The arrangement of the moderator and the target pins in the subassembly, the moderator material and the volume ratio of target to moderator were selected as parameters. The results of the calculations are shown in Table 2. The transmutation rate of ^{99}Tc in the new target subassembly is higher than that in the subassembly consisting of separate $ZrH_{1.7}$ moderator pins and ^{99}Tc target pins, as shown in Figure 2. A maximum ^{99}Tc transmutation rate of about 10%/year was obtained by using the new target subassembly loaded in the blanket region of the fast reactor. The new target subassembly can achieve an optimum transmutation performance by adjusting the volume ratio of $ZrH_{1.7}$ to ^{99}Tc in the duplex pellet.

Table 2. **Results of ^{99}Tc transmutation performance parameter survey**

Loading method of FP pins	Number of pins in sub-assembly	Number of FP pins	Radius of FP pin	Transmuted amount (kg/y)	Transmutation ratio (%)
Heterogeneous	127	37	0.5	41.1	1.8
Heterogeneous	127	22	0.5	27.2	2.5
Duplex	127	127	0.2	38.1	3.5
Duplex	127	127	0.063	10.8	9.8
Duplex	217	217	0.2	46.7	2.5
Duplex	217	217	0.063	17.1	9.1

The effects on main core characteristics of loading target subassemblies were also analysed. It was found that the power density of the core fuel adjacent to the target is rather high and is about the same as the maximum in the core. However, the power spike is much mitigated compared to the case of loading target subassemblies in the core region.

Several calculations were performed to determine the ^{129}I transmutation performance. ^{129}I was loaded as NaI. The isotopic concentration of ^{129}I was 76.5% and the remainder ^{127}I. The transmutation rate of ^{129}I was 5.2% and the transmuted amount was 18 kg in a year. The amount of ^{129}I produced by a 1 000 MWe class PWR is about 5 kg, so the transmuted amount of ^{129}I was equal to the output from 3 PWRs.

Measurements of nuclear data for MA and FP transmutation

In MA burner core analyses, nuclear data for MA nuclides and fission products are of primary importance. However, nuclear data for many MA nuclides are still not known to the desired accuracy. Accurate experimental data of neutron cross section for MA are indispensable to establish MA transmutation technology by FBRs.

Fission cross section of MA nuclides

Fission cross section ratios of minor actinide nuclides (^{237}Np, ^{241}Am and ^{243}Am) relative to ^{235}U in the fast neutron energy region have been measured to evaluate the accuracy of MA nuclear data, using a back-to-back (BTB) fission chamber at YAYOI fast neutron source reactor. The experimental results were compared with the fission cross sections in the JENDL-3.2, ENDF/B-VI and JEF-2.2 libraries. It was found that calculated values for ^{241}Am using the JENDL-3.2, ENDF/B-VI and JEF-2.2 data are higher by 19%, 21% and 18%, respectively, than the measured value in the centre of the core.

Making use of BTB fission chambers and a lead slowing-down spectrometer coupled to a 46MeV electron linear accelerator at Kyoto university, the fission cross sections of ^{237}Np, ^{241}Am, ^{242m}Am, ^{243}Am have been measured relative to that for ^{235}U (n,f) reaction in the energy range from 0.1eV to 10 keV. Each of the fission cross sections in the JENDL-3.2 and ENDF/B-VI libraries was compared with the measurement.

Capture cross section of MA nuclides

The keV-neutron capture cross sections of ^{237}Np were measured to evaluate the accuracy of the nuclear data libraries using the 3-MV Pelletron accelerator of the Research Laboratory for Nuclear Reactors at the Tokyo Institute of Technology (TIT). The measurement was relative to the standard capture cross sections of ^{197}Au. A neutron time of flight method was adopted with ns-pulsed neutron source the accelerator and a large anti-Compton NaI(Tl) gamma-ray detector.

As a results, the capture cross sections of ^{237}Np were obtained with the error of about 4% in an incident neutron energy region of 10 to 500 keV. The present data are compared with other experimental data and the evaluated values of JENDL-3.2, and it was found that JENDL-3.2 provided good evaluations for ^{237}Np.

Decay heat of MA nuclides

The decay heat power of nuclear reactors is an important quantity for design, safety and costs of operation of a nuclear reactor. Extensive works have been done for decay heat measurements of major nuclides such as ^{235}U, ^{238}U, ^{239}Pu and ^{241}Pu and the accuracy has been established. No experimental decay heat data exist up to now for minor actinides such as ^{237}Np, ^{241}Am and ^{243}Am that are important for MA burner design.

The objective of this study is to measure the decay heat of minor actinide nuclides irradiated by the fast neutron energy spectra accurately in order to verify the decay heat calculation and thus establish confidence in the calculational method.

The sample irradiations of ^{235}U (reference) and ^{237}Np were performed in the fast neutron source reactor YAYOI of Tokyo University. Gamma and beta decay heat released from fission products of the samples were measured using the radiation spectrometry method. A NaI(Tl) scintillation detector was used for measurement of gamma ray. A plastic scintillation detector and a proportional detector were used for beta ray measurement.

Capture cross section of RE nuclides

While it is essential to consider all actinide containing wastes in a total actinide recycling scheme, the high-level waste (HLW) certainly presents the most difficult partitioning problem. Especially, separation of the trivalent minor actinides (MA) (Am, Cm, and higher products) from rare earth (RE) nuclides is notoriously difficult. Several methods have been successfully applied to isolating an actinides/RE fraction from the HLW. The impact of RE nuclides in MA containing fuels on the core performance of FBRs was investigated. As a result, it was found that the effect of RE nuclides on core characteristics is large. Accurate neutron cross section data of RE nuclides become necessary for designing the MA burner core. The data, however, are quite inadequate both in quality and in quantity.

Measurements of keV-neutron capture cross sections of RE nuclides (^{147}Sm, ^{148}Sm, ^{150}Sm, ^{140}Ce, ^{141}Pr, ^{153}Eu, ^{143}Nd, ^{145}Nd) have been performed to evaluate the accuracy of the nuclear data libraries using the 3-MV Pelletron accelerator of TIT. The capture cross sections of RE nuclides were obtained with the error less than 5% in an incident neutron energy of 10 to 500 keV.

A comparison was made between the present experimental data and the evaluated values of JENDL-3.2. The comparison shows that the JENDL-3.2 overestimates the keV-neutron capture cross sections of ^{147}Sm and ^{150}Sm by 5 to 15% and 5 to 20%, respectively, underestimates those of ^{140}Ce, ^{141}Pr and ^{153}Eu by about 30%, 5 to 20% and about 10%, respectively, and estimates those of ^{148}Sm very well.

Capture cross section of FP

The thermal neutron capture cross section and the resonance integral of radioactive fission products have been measured. For the measurement of thermal neutron cross section, new method named isotope ratio method has been developed. The thermal neutron capture cross section and resonance integral has been measured for ^{99}Tc, ^{129}I and ^{135}Cs [9-11].

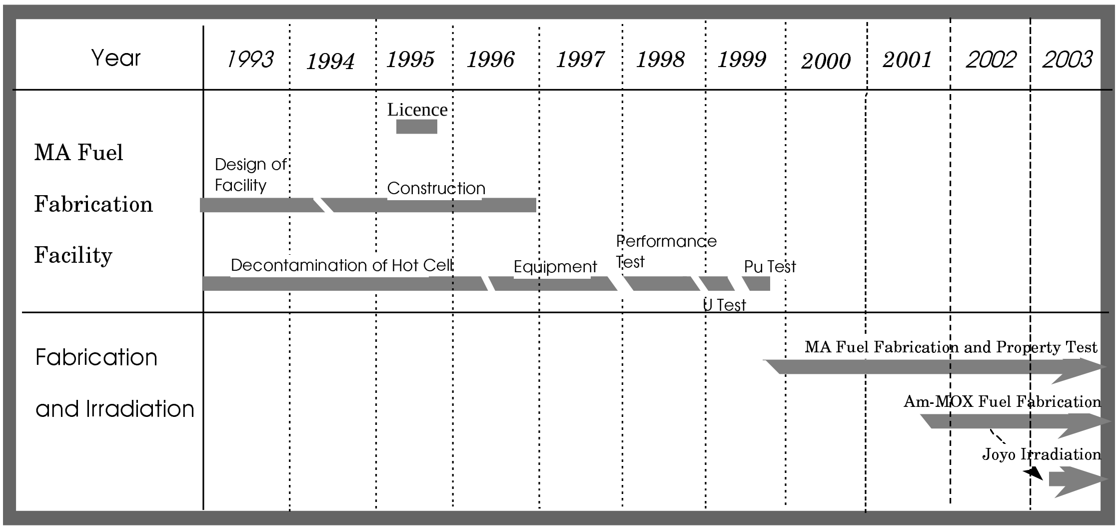
Transmutation rate

As a part of MA nuclear data evaluation, the analysis of irradiated ^{237}Np sample in Joyo has been performed. Additional irradiation test of Np, Am and Cm samples in JOYO was started in August, 1994.

Fabrication of MOX fuel containing MA

As for fabrication and investigation of irradiation behaviour of MOX containing MA, the systematic program has been planned in JNC. Two fabrication methods, pellet and vibro-packing have been studied for Neptunium-based fuel pins. The pellet type Np-based fuel will be fabricated at Tokai Works of JNC, and the fabrication of Np-based fuel by vibro-packing method will be performed at PSI in collaboration with JNC. For Am-based fuels, the Alpha-Gamma Facility (AGF) at Oarai Engineering Centre of JNC has already been reformed to fabricate MOX fuel pins containing Am at first as shown in Figure 3 and then containing Am and Np. Remote assembling will be conducted in the Fuel Monitoring Facility (FMC). Both facilities will provide test beds for the post irradiation examination. The irradiation test of Np- and Am-contained MOX fuel is planned in JOYO. In step with the JOYO MK-III schedule, the irradiation test will be initiated from around 2003.

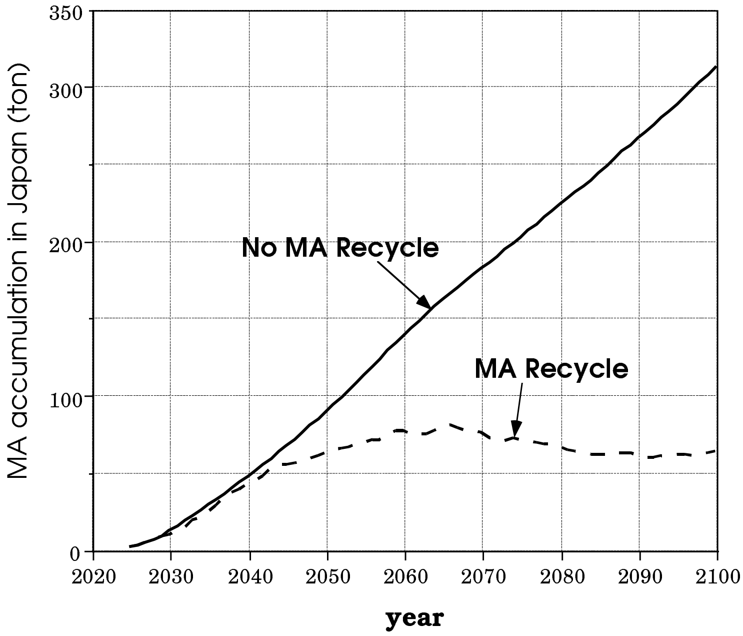
Figure 3. **Fabrication of MOX fuel containing Am**



Mass balance of MA in Japan

The MA mass balance was analysed according to the predicted nuclear energy production in Japan. Plutonium and MAs are recovered from the LWR and Pu-thermal reactors, recovered Pu and MAs are multiply recycled in fast reactors. Nuclear power generation is assumed to increase to 1 000 MWe/y, with the introduction of commercial fast reactors starting in the year 2030. New reactors are assumed to be totally FBR, and all spent fuel discharged from LWR and Pu-thermal reactors is assumed to be reprocessed. The total MAs transferred into the high level waste are calculated to be 310 tons from LWR, Pu-thermal LWR and FBR without recycling. In the case of recycling MAs into LMFRs after the year 2030, the MAs remaining in the fuel cycle in the year 2100 is reduced to about 60 tons, 80% less than without recycling , as shown in Figure 4 below.

Figure 4. **Effect of transmutation in reducing accumulation MA**



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