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COMMUNICATIONS

TRANSMUTATION OF SPENT FUEL IN REACTOR VVER–440

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This study deals with a specific way of transmutation process, decreasing the amount of long-lived and high-active radionuclides. The goal of the work is to analyse the change of the critical radionuclides concentration (critical from the point of view of radioactive waste management) during burn-up process in LWR.

K e y w o r d s: transmutation, spent nuclear fuel, radionuclides, combined fuel assembly

1 INTRODUCTION

The nuclear establishment, aware of its social and long-term responsibility, has always carefully considered the future management of irradiated fuel discharged annually from some 400 commercial nuclear power plants now in operation.

The management of irradiated fuel should ensure that the biosphere is protected under economically acceptable conditions without entailing unfavorable short-term consequences and the public must be convinced of the effectiveness of the methods. Since the spent fuel contains very long-lived radionuclides, some protection is required for at least 100 000 years. Two ways are possible:

- we can wait for the natural decay of the radioactive elements by isolating them physically from the biosphere by installing successive barriers at a suitable depth in the ground. This strategy leads to deep geological disposal;
- we can make use of nuclear reactions that will transmute the very long-lived wastes into less radioactive or shorter-lived products.

Whatever the solution chosen for highly radioactive wastes, deep geological repository disposal will always be necessary. There exists a volume of longlived and intermediate level waste arising from irradiated structural materials and losses during the various operations of the nuclear fuel cycle that is 20 times greater than that of the high-level waste. Tests are in progress to try to reduce the volume of these wastes, but there remains a lower threshold below which we cannot reasonably go. For humankind, the risks of a waste storage site depend on its radiotoxicity and the possibility of transfer to the biosphere. This transfer can occur after failure of the barriers and subsequent migration of the elements into the surrounding geosphere. Uncertainties regarding the transfer mechanisms, however, as well as the fact that we cannot totally dismiss the possibility of the waste coming into contact with the biosphere following a geological upheaval or accidental intrusion foster the distrust of the public. These uncertainties can only be approached by a statistical analysis of the possible threats. Different irradiated fuel management approaches can be envisaged:

- *Deep geological disposal of irradiated fuel without reprocessing*. This solution may be the least expensive and requires the least handling. On the other hand, it implies some waste of energy, the formation of which are in fact uranium and plutonium mines.
- *Alternative strategy of reprocessing of the spent fuel followed by deep geological disposal of wastes* has been chosen by France, UK, Japan and other countries. Uranium and plutonium are quantitatively separated from the other nuclides with yields ranging from 99.7 to 99.9 % [1]. The recovered uranium is reenriched and recycled in light water reactors (LWRs) thus reducing the need for fresh uranium ore. The plutonium can be used in LWRs (or even better in fast reactors) as mixed oxide fuel (MOX). Production of MOX fuel implies that the reprocessing has taken place shortly prior to fuel production in order to keep the radiation levels in the production plant as low as possible. By storing separated plutonium, ^{241}Am will grow-in from decaying ^{241}Pu. According to this approach the considerable energy content of the spent fuel is put to work in LWRs or fast reactors. The minor actinides and highly radioactive fission products are embedded in glass and are to be placed at the proper time into deep geologically sealed repositories. Their radiotoxicity decreases with a factor of 10 to 100 in 10 000 years. The recycling of plutonium in LWRs decreases the growth rate of plutonium stocks.
- *Advanced reprocessing* involves the separation, not only of uranium and plutonium, but also that of the so-called "Minor Actinides" (neptunium, americium, curium) and some long-lived fission products into single element or

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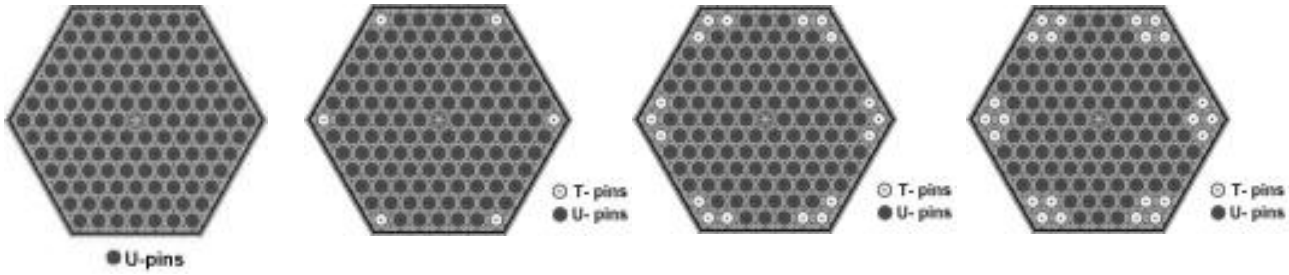

Fig. 1. Models of profile assemblies a) VVER-440 assembly, and assemblies with b) 6 T-pins, c) 18 T-pins d) 24 T-pins.

element-group packages with similar nuclear and/or chemical properties. In this way suitable solutions can be designed to improve conditioning or to set up transmutation scenarios. Transmutation of Pu and minor actinides will reduce the radiotoxic potential of high-level waste but has little effect on the release rate of radioactivity to the environment since the very low solubility of the actinides is the controlling transfer factor to the biosphere. Further R&D is required to investigate all the aspects of this way of waste management in order to be able to truly assess its benefits or consequences for the fuel cycle. Among the problems to be solved are the partitioning of hazardous materials with a high efficiency and their subsequent transmutation.

2 SIMULATION OF TRANSMUTATION PROCESS IN REACTOR VVER—440

The considered transmutation requires sufficient neutron flux that is initiator of the process. There are various sources of neutrons depending on transmutation technology. Various methods have physical and economic advantages and disadvantages. The considered way seems to be a less effective but cheaper one.

A simulation of the process was made by spectral code HELIOS [2]. The assembly VVER-440 was used at User's input in the burn-up process, see Fig. 1a, [3]. Burnt-up 50 000 MWd/tU was reached with parameters as follows [4]:

- enrichment: 3.6 % ^{235}U
- temperature of coolant: 553 K
- concentration of boron: 0.5245 g/cm^3

After burn-up the assembly was cooled in the spent fuel pool during 5 years, and its material composition was changed.

The first step of designed reprocessing is total removal of uranium. The rest of spent fuel is concentrated into the original volume and density of oxide fuel at the next step. New fuel pins, the so-called T-pins are constructed, as plutonium is the major fission material. Fuel assemblies containing T-pins have only low reactivity. For this reason, the next step is a formation of special uranium fuel assembly profiled by T-pins (6, 18 or 24 pins). Uranium pins (120, 108 or 102 U-pins) are identical with those in original fresh VVER-440 fuel; see Fig1. b – d. The geometry of profiled assembly is the same as that of standard assembly VVER-440. The enrichment of all U-pins in profiled assembly is the same.

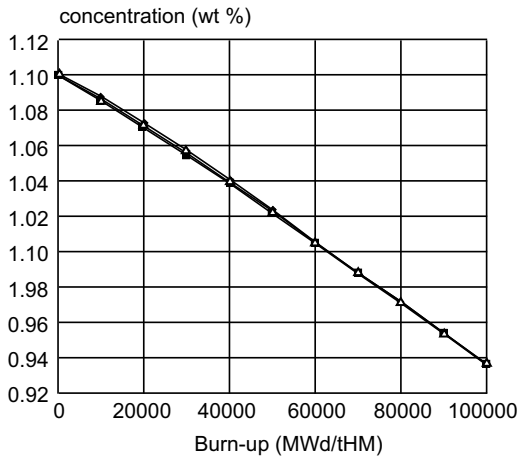

Fig. 2. The concentration characteristics of Sr-90 during burn-up. Legend: ♦ 6 T–pins, △ 18 T–pins, ■ 24 T–pins

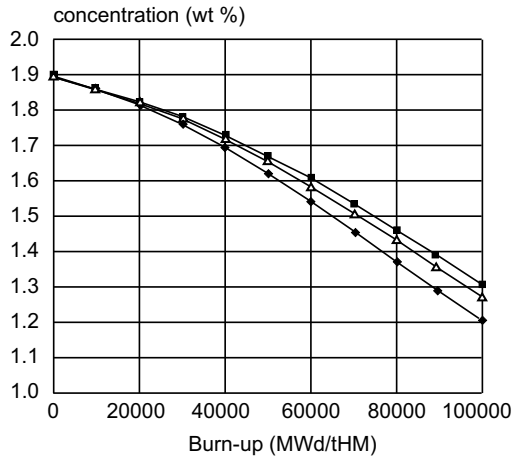

Fig. 3. The concentration characteristics of Tc-99 during burn-up. Legend: ♦ 6 T–pins, △ 18 T–pins, ■ 24 T–pins

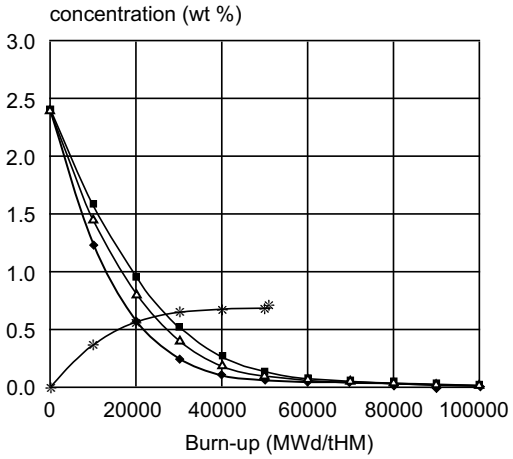

Fig. 4. The concentration characteristics of Pu-239 during burn-up. Legend: ♦ 6 T–pins, △ 18 T–pins, ■ 24 T–pins, ∗ U–pin

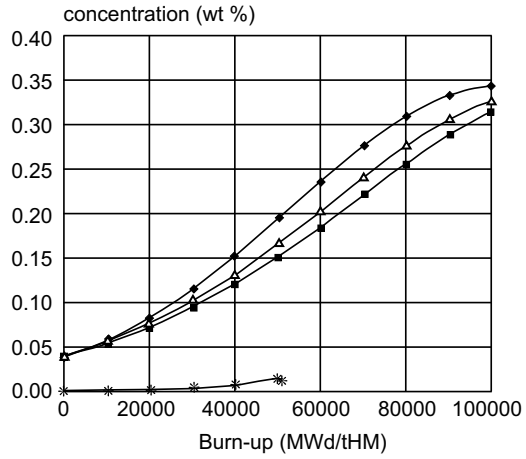

Fig. 5. Concentration characteristics of Cm-244 during burn-up. Legend: ♦ 6 T–pins, △ 18 T–pins, ■ 24 T–pins, ∗ U–pin

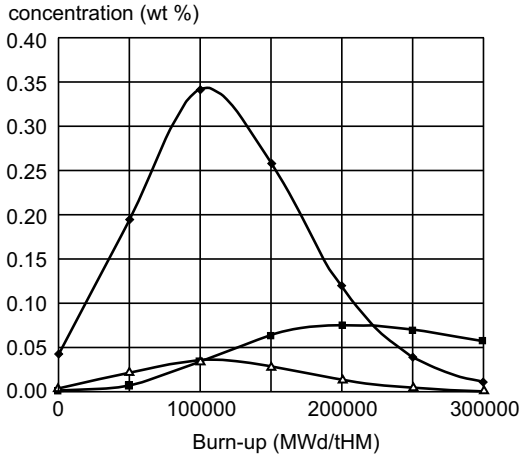

Fig. 6. Concentration characteristics of: ♦ Cm-244, △ Cm-245, ■ Cm-246 during burn-up.

Three enrichments of U-pins were taken into account: 3.6, 4.0 or 4.4 % ^{235}U. The resulting number of combinations (with T-pin numbers) was nine. All 9 types of the so-called combined assemblies (CA) were burnt-up to 100 000 MWd/tHM.

3 RADIONUCLIDE CONCENTRATIONS DURING BURN–UP

Changes of selected radionuclide concentrations were investigated only. Fission products ^{90}Sr, ^{93}Zr, ^{99}Tc, ^{129}I, ^{137}Cs, and actinides ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{244}Cm were taken into account.

Burn-up calculation results of combined assemblies with 6, 18 and 24 T-pins and enrichment of U-pins 3.6 % ^{235}U are presented below. The other combinations have similar characteristics during burn-up. The differences are induced by influences of T-pin number and enrichment value of U-pins.

The concentrations of fission products as ^{90}Sr, ^{99}Tc during burn-up up to 100 000 MWd/tHM are shown in Figs. 2 and 3.

Minimal influence of T-pin number can be seen. Concentrations of actinides as ^{239}Pu, ^{244}Cm during burn-up up to 100 000 MWd/tHM and breeding of these actinides during burn-up (up to 50 000 MWd/tHM) in reactor VVER-440 plus their stay in spent fuel pool for 5 years are shown in Figs. 3 and 4. Concentration of ^{244}Cm is growing only during the burn-up process. It can be seen in Fig. 5. However, when the assembly burn-up was simulated up to 300 000 MWd/tHM ^{244}Cm concentration started to decrease at burn-up above 100 000 MWd/tHM. ^{244}Cm is transmuted into the ^{245}Cm and ^{246}Cm (Fig. 6).

Positive effect can be seen on transmuted material masses. The mass of fresh fuel pellets (UO_2) in assembly VVER-440 is 136.36 kg. After fuel burn-up up to 50 000 MWd/tU and all spare uranium oxides removal from spent fuel, the nuclide mass of the rest, containing the transuranium elements, fission and decay products, is about of 7.2 kg. Masses of transuranium elements are displayed in Table 1 and in Fig. 7.

Table 1. The mass comparison of transuranium elements.

		U	Np	Pu	Am	Cm
mass (g/pin)	spent U-pin from VVER-440 assembly	904.50	0.77	13.26	0.78	0.12
	"fresh" T-pin from combined assembly	0	2.89	49.10	2.92	0.47
	spent T-pin from combined assembly	0	0.13	3.78	1.66	3.82

In the case that the T-pin has the same geometry and fuel density as the original pin of fresh VVER-440 assembly, 7 T-pins can be created as a result of one assembly reprocessing.

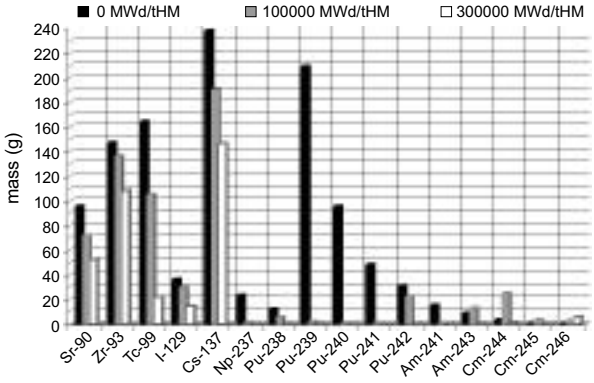

Fig. 7. The mass of transmuted products from spent fuel VVER-440 in T-pins

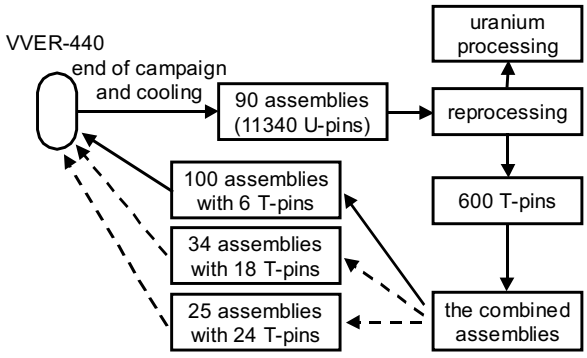

Fig. 8. Considered scheme for possible partitioning and transmutation of spent fuel

Taking into consideration that generally 90 spent fuel assemblies are removed from VVER-440 reactor during refuelling, the total mass of spent fuel without uranium oxides (spent fuel rest) is more than 650 kg. It follows 600 T-pins can be created from the spent fuel rest. These T-pins can profile 100, 34 or 25 combined assemblies with 6, 18 or 24 T-pins, respectively. The considered model for possible application of the partitioning and transmutation technology is shown in Fig. 8.

4 CONCLUSION

Partitioning and transmutation techniques offer the possibility of removing by-products that contribute to the long-term risks of storage from spent nuclear fuel and transmuting them in transmutation facilities to less harmful products. The by-products as follows were taken into account: fission products ^{90}Sr, ^{93}Zr, ^{99}Tc, ^{129}I, ^{137}Cs, actinides ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, ^{244}Cm.

The calculation possibilities offered by HELIOS, were used to model and to calculate the burn-up of standard.

The fuel assembly for the reactor VVER-440 resulting in nuclide composition was used as a starting point for partitioning-transmutation. A combined assembly was profiled radially by non-uranium pins (T-pins) with a content of plutonium, higher actinides and fission products. The transmutation process in these pins was observed during burn-up.

Time-dependent concentrations of the mentioned by-products at combined assemblies were analysed. A decrease of all concentrations was demonstrated but usually at very high burn-up levels in comparison with recent burn-ups. It was shown that transmutation process is theoretically possible also in reactor VVER-440. It is necessary to analyse a more realistic transmutation fuel cycle and to estimate efficiency and applicability of transmutation process in the reactors VVER-440.

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