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Burning transuranium isotopes in thermal and fast reactors

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Abstract

Energy production in nuclear power plants on the basis of fission processes lead inevitably to fission products and to the generation of new actinide isotopes. Most of these fission products are rather shortlived and decay within less than about 500 years to stable nuclides. However, a few of them, e.g. ^{99}Tc and ^{129}I, are longlived and may contribute to the radiotoxicity and hazard associated with an envisaged repository for their long-term disposal in a stable geologic formation, e.g. a salt dome. The majority of the generated actinide isotopes are fairly longlived, e.g. ^{239}Pu with a halflife of more than 20 000 years. Therefore, their direct storage poses a heavy burden on the capacity and the possible environment impact of a repository. Furthermore, the energy content of these actinides could be deployed for producing additional nuclear fission energy after recovering them from unloaded irradiated fuel by suitable reprocessing techniques. Various possibilities exist for burning these actinides in different types of reactors, e.g. in light water reactors (LWRs), or LMFRs, adhering to available technology, or in actinide burners particularly designed for the purpose of their efficient incineration. The different options will be discussed in the paper. Transmutation of the manmade actinides and longlived fission products will require advanced technologies e.g. regarding reprocessing losses, remote fabrication techniques, and most probably, isotope separation processes. However, the almost complete elimination of these nuclides resulting from fission energy production in a continued recycling process may be the only feasible way to limit the effects of nuclear power generation to a tolerable and fair level for generations to come.

1. Introduction

After Chadwick, in the early 1930s, had demonstrated the existence of the neutron, which Rutherford had predicted approximately one decade earlier, it took less than another decade for Hahn and Straßmann to discover nuclear fission shortly before the beginning of the Second World War. Proof of nuclear fission of the ^{235}U isotope was the identification of the fission products generated in the process, most of them radioactive, changing into stable atoms by emitting radiation or particles, mostly within a relatively short period of time.

In 1940, Seaborg (1951 Nobel Prize winner) and coworkers discovered the first artificial element, plutonium, which later was found in very low concentrations also in natural ores.

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Plutonium[1], practically not occurring in nature, constituted the first transuranium element of which larger quantities (several tens of kilograms) were produced in so-called atomic piles within a few years. The first operation of such a pile, now called nuclear fission reactor, i.e. the management of the nuclear fission chain reaction on a technical scale, was achieved by Wigner in Chicago approximately at the same time. The transuranium element plutonium acquired negative fame after the atom bomb containing it was dropped on Nagasaki 3 days after the first atom bomb had been dropped on Hiroshima on August 6, 1945, which had contained the ^{235}U isotope as a fissionable material.

Producing plutonium on a kilogram scale at that time took immense efforts of all scientists and engineers involved, not to mention the huge costs of this military project. The ^{239}Pu isotope was the key product because its properties made it particularly suitable for use as a fissile material in a bomb. The approach chosen to produce ^{239}Pu can be recognized comparatively easily from the section of the Karlsruhe Chart of the Nuclides in Fig. 1. Neutron capture in ^{238}U initially generates ^{239}U, which relatively quickly (23.5 min halflife) decays into ^{239}Np, subsequently (2.355 days halflife) going on to decay into ^{239}Pu. ^{239}Pu is quite longlived, with a halflife of roughly 24 000 years. This is why plutonium is the main constituent of the transactinides generated in reactor operation (Table 1).

Table 1
Annual arisings of transuranium elements worldwide

Element	Production rate (kg per year)
Np	3400
Pu	68 000
Am	2740
Cm	335

[1] Readers interested in the properties of plutonium, the associated risk, and in the precise meaning of specific terms used in nuclear engineering can seek more detailed information in e.g. GSF (1989), Koelzer (1989, 1997), Volf (1989), Deutsches Atomforum (1996), Spektrum der Wissenschaft (1997).

Producing larger quantities of ^{239}Pu also required the corresponding number of neutrons. Special nuclear reactors were built and operated to produce them. The fuel used was either natural uranium with approximately 0.7% of ^{235}U, or slightly enriched uranium with a content of approximately 3% of ^{235}U. When a neutron is absorbed in a ^{235}U nucleus, this mostly results in nuclear fission, and the fission neutrons released may trigger a chain reaction. Less frequently, neutron capture leads to ^{236}U. As the probability of neutron absorption processes in ^{235}U becomes significantly larger as the neutron energy decreases, so-called thermal neutrons are used in these plants. These 'slow' neutrons are obtained by the moderation (which means slowing down) of 'fast' neutrons produced in nuclear fission with an energy in excess of 1 MeV. In this thermalization process, the neutron energy is reduced by more than a factor of 10^7. Well thermalized neutrons have a mean energy of approximately 0.025 eV. They are then in a state called thermal equilibrium with the ambient media at room temperature and operating temperature, respectively. Yet, these thermal neutrons are not really 'slow' in the conventional sense, their mean velocity still being around 2200 m s^{-1}. Nuclear reactors in which most neutron reactions, primarily fission and capture, are caused by thermal or low-energy neutrons, are usually referred to as 'thermal reactors.' These include light water reactors (LWR), of which two variants, namely pressurized water reactors (PWR) and boiling water reactors (BWR), are operated for electricity generation at Philippsburg not far from the Karlsruhe Research Center. In 'fast reactors' with so-called 'hard' neutron spectra, most of the nuclear reactions induced by neutrons occur at considerably higher energies (and correspondingly higher neutron velocities), namely in the range of several 100 keV. Reactors of this type have been developed for a long time within the Fast Breeder Project at the (then) Karlsruhe Nuclear Research Center. As a prototype of this development line in the Federal Republic of Germany, the SNR-300 at Kalkar was to furnish experience in the operation of these novel plants. In France, the large Superphénix (SPX) facility has completed approximately

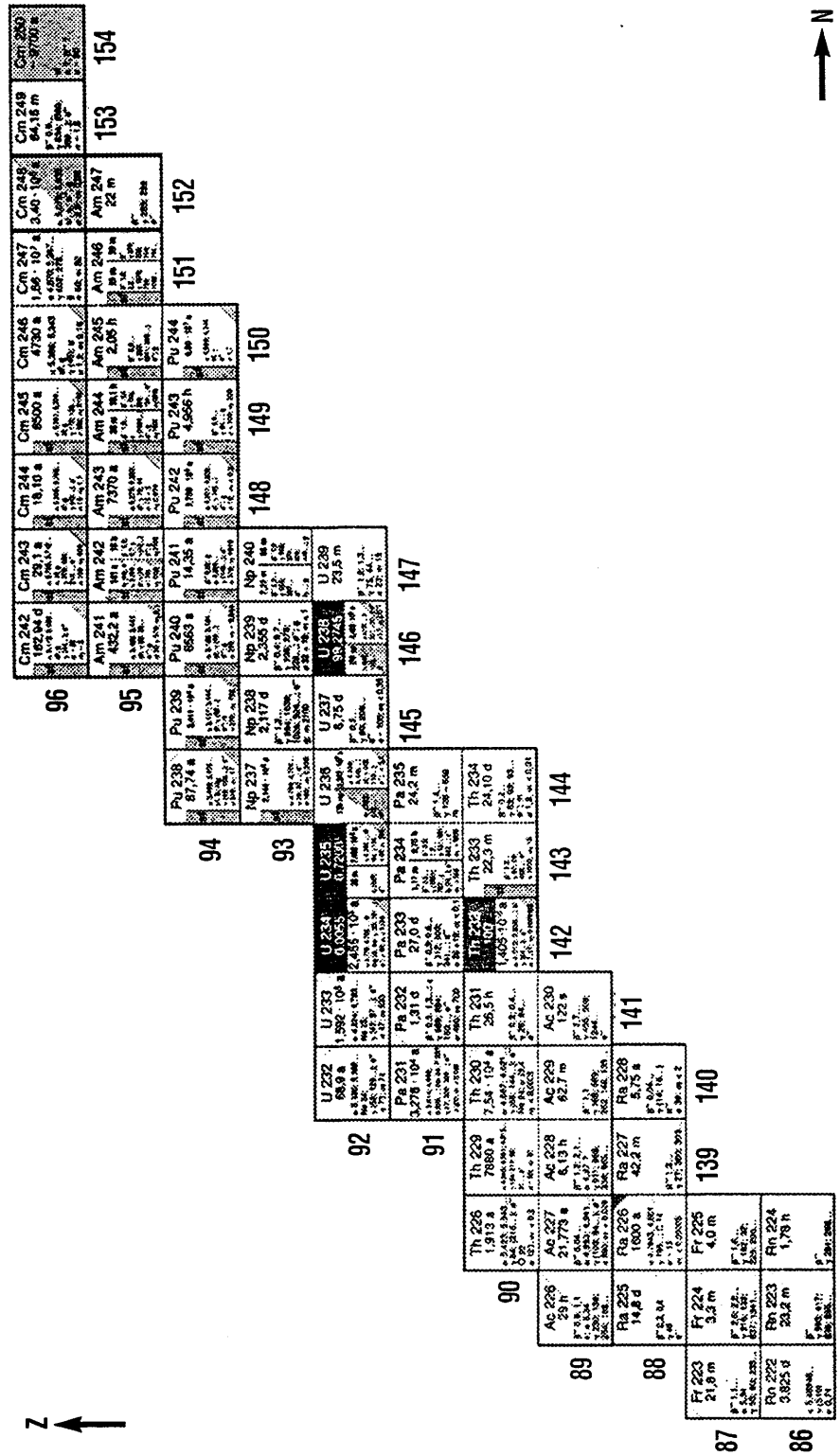

Fig. 1. Extract from the Karlsruhe Chart of the nuclides, sixth edition, 1995, W. Seelmann-Eggebert et al.

10 years of power operation, not all of them troublefree.

Fig. 1 shows the elements in the Periodic Table arranged by proton numbers in the atomic nucleus and of electrons in the atomic shell, respectively. Isotopes with the same numbers of neutrons in their atomic nuclei are positioned vertically below one another. It appears from these diagrams that neptunium, with the proton number of 93, is the first transuranium element. By analogy, thorium, with the proton number of 90, is considered the first transactinium element, or transactinide for short. The diagram also explains the way in which ^{239}Pu is produced on the basis of ^{238}U, as discussed above, and shows the formation paths of the other important isotopes of the transuranium elements, americium (Am) and curium (Cm), as well as the transitions resulting from radioactive decay. Thus, three consecutive α-decays, $^{242}\mathrm{Cm} \rightarrow {}^{238}\mathrm{Pu} \rightarrow {}^{234}\mathrm{U} \rightarrow {}^{230}\mathrm{Th}$, give rise to the thorium isotope ^{230}Th from ^{242}Cm. The colored chart of the nuclides shows these processes and the different types of nuclear decays much more clearly. It also indicates that ^{230}Th, as a result of two other α-decays, $^{230}\mathrm{Th} \rightarrow {}^{226}\mathrm{Ra} \rightarrow {}^{222}\mathrm{Rn}$, finally produces ^{222}Rn, and thus one of the radioactive isotopes of the noble gas radon, from the immediate precursor isotope of radium, ^{226}Ra. The decay of ^{222}Rn, which also occurs in ore mines, mainly determines radiotoxicity[2] as a result of inhalation in the environment of dumps of overburden or inside buildings made of granite rock. Inhaling high concentrations of ^{222}Rn implies an increased radiation hazard because of the shortlived decay products generated. It should also be pointed out that the ^{234}U isotope mentioned above mainly occurs as an intermediate product in the very slow decay of ^{238}U (Koelzer, 1997).

2. The purpose of burning transuranium isotopes

While plutonium was only available in kilogram amounts (almost exclusively for military purposes) worldwide half a century ago and, accordingly, was very expensive, a considerable quantity of this technically produced transuranium element had accumulated in the meantime. The worldwide arisings of transuranium elements are shown in Table 1 (taken from Beauvy, 1995). The arisings of 68 t per year of Pu are in good agreement with the value of 75 t per year Pu contained in spent fuel for an installed nuclear capacity of 350 GW(e) total electricity output in 1997 reported in an IAEA bulletin (Oi, 2000). As mentioned above, plutonium, which is originally produced by neutron capture in ^{238}U, makes up the lion's share. Other neutron capture reactions, e.g. in ^{235}U and in ^{236}U, as well as radioactive decay reactions, e.g. $^{241}\mathrm{Pu} \rightarrow {}^{241}\mathrm{Am}$, give rise to considerable amounts of other transuranium elements, e.g. approximately 3 t each of neptunium and americium. In Germany, some 5 t of plutonium are annually produced (see G. Heusener's introductory contribution in this issue).

In addition to the plutonium produced in the ongoing operation of nuclear power plants generating electricity, there is also the military sector in the nuclear weapon states with an estimated stockpile in excess of 100 t, part of which is being released in the disarmament efforts by nuclear weapon states. It is not very encouraging to observe only little progress since 1993 when at a session of the ANS Summer Meeting devoted to nuclear weapons material utilization the following conclusion was drawn (Steyn, 1993). "The disposition of the high-grade plutonium that will become available as nuclear weapons are dismantled is fast becoming a major international nuclear nonproliferation issue. While neither the United States nor Russia seems to have developed a clear and acceptable policy in this regard, beyond storage in the interim, both must do so soon." Although apparently urgent, no clear-cut decision has been taken nor is an agreed strategy or a unique procedure or a consistent policy identifiable. As a consequence of the stringent criteria ensuring the safety and security especially of nuclear weapons grade plutonium, the formerly expensive material meanwhile has come to be regarded frequently as an unwelcome follow-on product of reactor operation either to be stored in

[2] For an explanation of radiotoxicity, see, e.g. (Koelzer, 1997), p. 145.

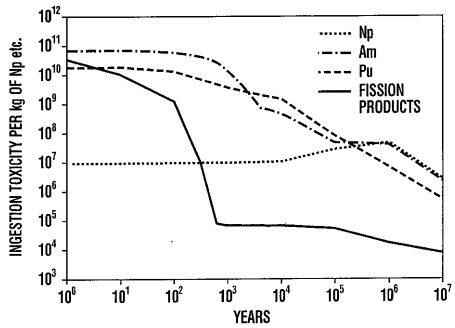

Fig. 2. Time dependence of the radiotoxicity of fission products and transuranium elements, including their decay products. Complete transmutation of the transuranium elements into fission products can reduce radiotoxicity roughly 1000 times after a storage time of approximately 500 years.

safe underground repositories or to be disposed of by transmutation. On the other hand, it should be borne in mind that the energy content of plutonium is roughly comparable to the one of the ^{235}U isotope. Obviously, there exist two almost contrary opinions or goals regarding the value of plutonium, (1) considering plutonium as a potential resource material that can be deployed for energy production; and (2) considering plutonium as an unavoidable by-product of nuclear energy production that could be immobilized safely together with surplus weapons grade plutonium in a suitable repository for permanent disposal. The idea of plutonium actually being a valuable material is emphasized especially by the Russian side in discussions about a potential direct disposal of weapons grade plutonium. A joint US/Russian study (Moses et al., 1998) came to the conclusion, 'the BN-600 reactor is capable of converting up to 1.3 metric tons of surplus weapons-derived plutonium to spent fuel each year.' Various options for plutonium in fast and light water reactors under consideration in Russia have also been described by Chebeskov et al. (1996). In the United States, this kind of direct disposal of weapons grade plutonium is under serious consideration, of course, with the appropriate safety precautions. At about the same time, 1998, in a US study with contributions of several national laboratories (Strachan et al., 1998) intentions for direct disposal can be deduced from the finding: 'two candidate immobilization forms for the disposal of surplus weapons-usable Pu, glass and ceramic were tested and characterized to provide sufficient data for an informed selection of one form for further R&D'.

Some of the transuranium isotopes, other than plutonium, arising in nuclear power production are extremely longlived (Fig. 1 for the halflives) and their radiotoxicity must not be neglected when assessing the long-term hazard potential of the transuranium elements generated in nuclear reactors. Because of their comparatively low concentrations, these transuranium elements (mainly neptunium, americium, and curium with ^{237}Np, ^{241}Am, ^{243}Am, and ^{242}Cm, ^{244}Cm as the main isotopes), are frequently referred to as 'minor actinides' (MAs). These MAs can be considered as fertile isotopes too, similar to ^{238}U, although their energy potential may be less than that of e.g. plutonium.

Numerous studies performed by various research teams have shown that the disposal only of plutonium will not greatly reduce the long-term hazard potential associated with transuranium elements. This objective can be attained only by eliminating most of the MAs at the same time. Transuranium elements can only be destroyed by ultimately being converted into fission products, i.e. they must either be fissioned directly (and 'burned' in this way) or transferred into easily fissionable nuclei by preceding neutron capture processes. Although the energy potential of MAs might be considered as to be only modest, efforts for using their energy content are under discussion, as is obvious e.g. from the Japanese options making extra gains from actinides and fission products (OMEGA) project, mentioned e.g. in Salvatores et al. (1998). One decisive aspect allowing the long-term hazard potential to be diminished is the halflife of the radioactive fission products, i.e. of the fragments generated in nuclear fission. With very few exceptions, the radiotoxicity of fission products will have decreased to almost negligible levels after roughly 500 years (Fig. 2). Consequently, there has been serious thinking about not necessarily keeping fission products in an underground geologically safe repository, but instead storing them in protected

containers in monitored stores so as to be retrievable, e.g. as vitrified material. This type of controlled annealing of the 'ash' produced in nuclear fission can be ensured over several centuries, in the opinion of the proponents of this final storage option, and the safety of such stores can be maintained even over long periods of time such as these. The fission products with long halflives referred to above are produced in nuclear reactors in much smaller quantities than are the transuranium elements. Arisings are roughly comparable to those of minor actinides (Table 2, taken from reference 1 in Broeders and Broeders, 2000). It is doubtful whether the curium isotopes ^{243}Cm and ^{244}Cm, listed in Table 2, with halflives of approximately 30 and 20 years, respectively, can really be considered longlived, as they decay into ^{239}Pu and ^{240}Pu, respectively, relatively quickly.

It should also be pointed out that some studies were conducted about the disposal of longlived dangerous fission products (LLDFPs) by transmutation into stable nuclides. Nuclear fission reactors can also be used for this purpose, but they must have a particularly favorable neutron balance because of the surplus neutrons required. This is where reactors or accelerator-driven subcritical assemblies with mainly 'fast' neutrons offer a basic advantage, as the ratio of fission reactions to capture reactions is much higher for high-energy neutrons on the average, and for that reason the neutron surplus is higher than for low-energy, so-called thermal or epithermal, neutrons.

Table 2
Contents of transuranium isotopes and longlived fission product isotopes in spent LWR fuel elements (for a PWR of 3 GW thermal power from which an annual 33 t of fuel with a burnup of 33 GWd per t is unloaded; assumed decay time, 10 years)

Nuclide	(kg per year)
^{238}Pu	4.52
^{239}Pu	166.0
^{240}Pu	76.7
^{241}Pu	25.4
^{242}Pu	15.5
^{237}Np	14.5
^{241}Am	16.6
^{243}Am	2.99
^{243}Cm	0.01
^{244}Cm	0.58
^{79}Se	0.17
^{85}Kr	0.40
^{90}Sr	13.5
^{93}Zr	23.2
^{99}Tc	24.7
^{107}Pd	7.3
^{126}Sn	0.96
^{129}I	5.8
^{135}Cs	9.4
^{137}Cs	31.8

The LLDFPs greatly contributing to the hazard potential, ^{99}Tc and ^{129}I, can be transformed into stable nuclides relatively easily. For other LLDFPs, such as cesium (Cs) or palladium (Pd), this is much more difficult and expensive to achieve, probably only by means of modern technology. In addition to the radioactive isotopes mentioned above, there are other stable or far less hazardous isotopes of the same elements, which occur as fission products. They should be separated from the LLDFPs, if possible, as they would otherwise cause considerably higher neutron consumption when irradiated in reactors for transmutation purposes together with the LLDFPs of the same element (e.g. ^{135}Cs together with the much shorter-lived ^{137}Cs). On the other hand, isotope separation by advanced laser technology is not really a utopian proposition. However, it means venturing into new technical fields for the quantities of material arising and, accordingly, would require considerable expense in research and development if transmutation also of these LLDFPs were considered an important objective.

One general difficulty associated with burning transuranium elements and in the transmutation of LLDFPs is the fact that normally irradiation in reactors is not sufficient to render these materials harmless almost completely by a once-through treatment, given the present state of the art, e.g. with respect to the maximum radiation dose tolerated by steel used for fuel pins, fuel subassemblies and structural core components. Consequently, multiple recycling of the residues of incomplete burning is necessary. This is bound to result in the requirement that reprocessing of these residual materials must allow only a very small fraction to go into the waste because, otherwise, the desired

objective of reducing the long-term hazard potential of repositories cannot be achieved. For the most important transuranium element, plutonium, the technical-scale separation techniques, especially by the PUREX process (Plutonium–Uranium Recovery by Extraction), are being improved continuously so as to optimize the separation factors, i.e. minimize the amounts of plutonium ending up in the waste. Separation techniques proven either on a laboratory scale or in pilot plants exist also for important MAs. Some preliminary tests were conducted in CYRANO, the French CEA plant in Fontenay-aux-Roses, which is operated by the DIAMEX process using dimethyldibutyltetradecylmalonamide for the co-extraction of trivalent actinides (Am, Cm) and lanthanides. They constituted the basis on which the ATALANTE laboratory of the CEA was set up in Marcoule. Finally, separation of the lanthanides from Am and Cm must be achieved in a selective extraction step. The CEA is currently testing an electrochemical process called SESAME (Séparation par Extraction Sélective de l'Am par Méthodes Electrochimiques) (Bouquiun, 1996) which stands a fair chance of achieving the desired goal. These processes are expected to allow the almost complete extraction of MAs in reprocessing. As an alternative to traditional wet chemical reprocessing, different process techniques could be used when required by which the volumes of transuranium elements and LLDFPs in the waste could be limited to tolerable levels. Such processes have already been used in pilot plants or are being tested on a laboratory scale.

3. Removal from the biosphere of transuranium elements and longlived fission products

Because of their very long life, some radioactive daughter products arising in energy production by nuclear fission represent a long-term hazard potential to the environment. Consequently, these substances must be prevented from entering the biosphere (as far as it is important to human existence) in the long run.

Under these conditions, there are only two feasible ways of removing these hazardous materials from the biosphere, (1) direct disposal in geologically stable underground formations; (2) disposal by transmutation into stable or short-lived nuclides. Since, it will hardly be possible to transmute all of the dangerous long-lived fission products and reprocessing wastes, these two ways should be considered and followed as complementary strategies. In particular, even very efficient transmutation capabilities most probably will not eliminate the need for disposal of a presumable significantly reduced amount of high level radioactive waste, i.e. transmutation cannot completely replace geologic repositories.

Opinions differ about the long-term stability of geologic formations (salt domes, hard rock, clay deposits) and the requirements they must meet. In addition, different rules and regulations as well as conditions apply nationally and locally. The reliability of risk forecasts can be supported only by conclusions about existent or non-existent past changes within geologic periods of time, and by accelerated testing experiments under simulated realistic conditions. In view of the prevailing public skepticism it must be pointed out, however, that the prehistoric natural nuclear reactor in Oklo, Gabon, Africa (of course, without any special protective measures) restricted the spread of the daughter products of nuclear fission generated at that time within a range of 10–20 m around their original source (Métivier et al., 1995). Discussions, sometimes biased and very emotional, about the risk associated with direct disposal, the tolerability by the present generation and future generations, the reliability of long-term observance of conventions on behavior and protective norms after millennia, will not be continued in this article[3].

[3] It may be of historical interest to note that, according to the memoirs of Weinberg (1994)), Enrico Fermi must have recognized the acceptance problem early on and expressed his concerns accordingly: ''It is not clear that the public will accept a source of energy that produces so much activity or that might be subject to diversion of bomb material by terrorists.''

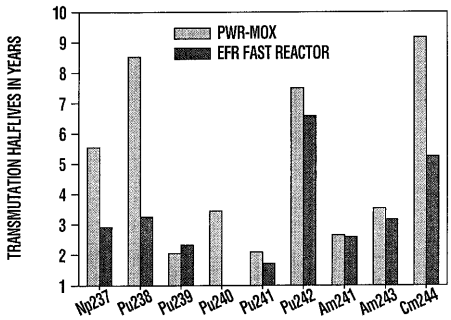

Fig. 3. Typical transmutation halflives for transuranium nuclides in a thermal reactor (PWR MOX) and in a reactor of European Fast Reactor (EFR) type both operated on plutonium-bearing mixed oxide fuel. Short transmutation halflives have a positive impact on transmutation.

Let us instead explain the other possibility of disposing of manmade radioactive hazardous material, namely, conversion into stable or shortlived nuclides. Of course, only nuclear physics processes can achieve this kind of conversion. At the current state of the art, neutrons are the particles best suited to 'destroying' all radwaste. This destruction can be achieved in two ways, (1) by fissioning the transactinide elements; and (2) by transmutating longlived hazardous fission products into shorter-lived or stable nuclides by neutron capture (i.e. incorporation of a neutron or, perhaps, several neutrons may generate either a stable nuclide or a nuclide decaying in a much shorter period of time than the source nuclide and, in this way, changing into stable nuclide).

Burning is possible only by nuclear transmutation, i.e. by nuclear processes. The cross-sections, as a measure of reaction probabilities, indicate that primarily neutrons can be used as reaction partners in these nuclear reactions. The reaction probabilities for other particles of low mass, e.g. protons (charged hydrogen atoms) or α-particles (charged helium atoms), are normally considerably lower than those for neutrons. In order to ensure that the incineration process caused by neutrons proceeds as effectively as possible, a facility with high neutron density is required. As the reaction rate is proportional to the product of neutron density and neutron velocity, these neutron physics studies normally employ the so-called neutron flux density (frequently referred to only as neutron flux) as the most useful quantity, measured in neutrons per cm^2 and second, which characterizes the neutron distribution as a function of space and time. Another factor influencing the reaction rate is the so-called cross-section, a measure of the probability of a neutron reacting with a specific nuclide. The transmutation halflives shown in Fig. 3 are a useful tool in assessing the suitability of various types of reactors for actinide transmutation. It will be seen below that the transmutation halflife must not be used as the sole criterion in assessing suitability. The extra consumption of neutrons required for transmutation (Salvatores et al., 1994) is only mentioned as one example, as no further reference will be made to this aspect below.

Until very recently, primarily critical nuclear reactors were considered the most suitable plants for annihilation. Over the past few years, sub-critical accelerator-driven facilities of the fast-reactor-type originally discussed for breeding fissile material have been included in transmutation considerations. In the paper by Broeders and Broeders (2000) in this publication, special mention will be made of this type of plant. This article will, therefore, be restricted largely to conventional types of reactors in which the burning of transactinide elements and LLDFPs represents an aspect additional to the usual generation of energy from nuclear fission. Only two reactor lines will be discussed:

1. LWRs or, even more specifically, PWRs, with the other variant, BWRs, in principle being eligible as well;
2. fast reactors whose neutron physics behavior and the computer codes required for analysis make them rather similar to the fast breeder reactors studied in detail at the FZK some time ago.

4. Transmutation in existing light water reactors

Reusing or recycling the plutonium bred in LWRs in plants of the same type represents the state of the art in several countries, especially in Germany and France (Schlosser et al., 1999). Up to 50% of the standard fuel elements (UOX fuel

Table 3
Rate of transuranium element production in LWRs; the numbers refer to the quantity (kg t^{-1}) of original heavy metal loading, existing at the given burnup for a mean rating of 164 W cm^{-1}. When comparing the data associated with different burnups (33 and 50 GWd per tHM, respectively), it should be borne in mind that relative to the same energy generation, the values associated with 33 GWd per tHM must be multiplied by a factor of 1.5

Material	Burnup							
	33 GWd per tHM				50 GWd per tHM			
	^{235}U enrichment				^{235}U enrichment			
	3.2%	3.5%	4.0%	4.5%	3.2%	3.5%	4.0%	4.5%
Pu	9.592	9.581	9.561	9.537	11.733	11.777	11.855	11.932
^{237}Np	0.4254	0.4306	0.4368	0.4408	0.6453	0.6650	0.6916	0.7119
^{239}Np	0.0883	0.0846	0.0791	0.0745	0.1002	0.0965	0.0907	0.0854
^{241}Am	0.0370	0.0373	0.0374	0.0371	0.0540	0.0565	0.0604	0.0638
^{243}Am	0.1002	0.0869	0.0694	0.0561	0.3220	0.2914	0.2467	0.2095
Transuranium elements	10.243	10.220	10.184	10.146	12.855	12.886	12.944	13.003

with enriched uranium) may be replaced by specially designed mixed oxide fuel elements (MOX fuel containing Pu instead of ^{235}U as the main fissile material). This fraction was chosen so that the reactivity coefficient of coolant density, which is important for safe operation of the reactor, remains within the permissible range. Table 3 lists the production of plutonium and MA in conventional UOX LWRs as a function of the uranium enrichment level and the burnup of fuel unloaded from the reactor. 33 GWd per ton HM is a typical value for reactor operation in recent years, while 50 GWd per tHM is an optimistic level envisaged by reactor operators for the near future. The energy produced can be converted into fissile material consumption, and used as an approximation to the energy content of a fissile material, by the rule of thumb according to which fission of 1 g of heavy metal produces approximately 1 MWd or 24 000 kWh. Consequently, a burnup of 50 GWd per tHM means about 50 kg of each ton of heavy metal loaded were fissioned. It is surprising, first of all, that this level can be achieved with a ^{235}U enrichment of less than 5%. This is explained by the conversion of the non-fissionable uranium isotope ^{238}U into fissile ^{239}Pu, which occurs continuously in the fuel rods of a reactor in operation.

After reprocessing, the first-generation plutonium extracted from the unloaded fuel elements can be processed into MOX fuel and reloaded in the reactor. As reactor operation is continued with multiple recycling, the quality of the plutonium tends to deteriorate, i.e. its content of the fissile isotopes ^{239}Pu and ^{241}Pu, decreases, as is seen in Fig. 4. This deterioration may develop to a stage at which any further use of this higher-generation plutonium would require an excessively large plutonium fraction in the MOX fuel elements that could impair the safe operation of a reactor loaded with that fuel (for a more detailed discussion of the influence of the isotopic composition of plutonium upon reactor physics

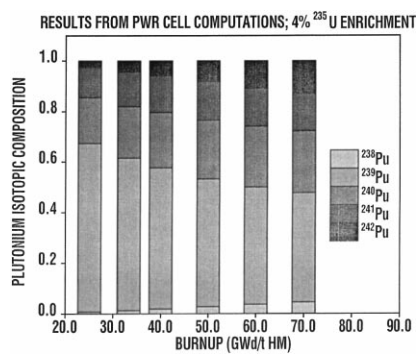

Fig. 4. Changes in plutonium isotopic composition as a function higher discharge burnups in an LWR.

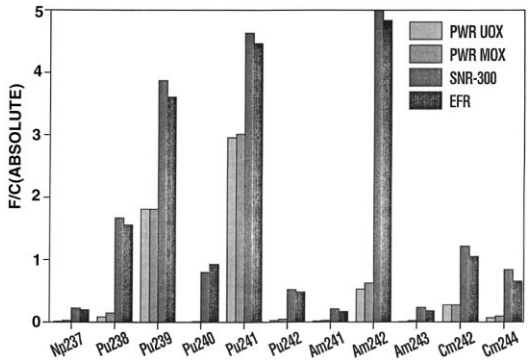

Fig. 5. Ratio of fission to capture reaction rates, F/C, in thermal and fast reactors. High F/C ratios favor burning longlived nuclides.

parameoters important for behavior during normal operation and under accident conditions, see Broeders, 1996 and the references quoted there). One possible way out of this dilemma is to fabricate MOX with enriched uranium. Such MOX fuel can be considered in the light of two different aspects, (a) the enriched uranium is added to plutonium; energy generation from the plutonium decreases the amount of natural uranium to be provided annually for reactor operation. As a result of the reduced U_{nat} requirement, reserves will extend over a longer period of time. (b) The addition of ^{235}U to the so-called dirty plutonium allows this radiotoxic element to be burned further in a proven type of reactor, thus avoiding or reducing its accumulation in interim stores or repositories.

Detailed studies as described, e.g. in Broeders (1996), and similar studies from other research laboratories have shown that the quantity of plutonium can be kept at a constant level after a certain startup period, and the operation of about 10 UOX reactors would necessitate the simultaneous operation of six MOX reactors of the same size. The necessary degree of ^{235}U enrichment depends on the desired burnup, varying between rather low levels at 33 GWd per tHM and approximately 4% enrichment at a discharge burnup of 50 GWd per tHM. It must be pointed out, however, that this procedure would cause a sizeable part of the plutonium to be converted into MAs, i.e. the quantities of neptunium and americium accumulating would increase considerably in the course of this continuous power production (Wiese, 1999). An explanation will be given below of the existing possibilities to burn these MAs and thus reduce or limit the arisings of MAs.

In conclusion of this paragraph, it should be pointed out (Broeders, 1996) that the computer codes and the associated data library employed in these studies, especially the nuclear data, were thoroughly verified and backed by numerous experimental results.

5. Transmutation in fast burner reactors

MOX LWRs allow plutonium to be transmuted and the whole plutonium inventory to be stabilized. However, this implies a more pronounced growth of americium arisings. In a nutshell, some 30% of the spent plutonium are converted into MAs. Moreover, it must be remembered that also neptunium is produced in UOX and MOX LWRs. Initially, the quantity of neptunium does not seem to be too remarkable — approximately 15 kg per year in a modern large LWR, i.e. approximately 10 kg per GW_e year. However, if one takes into account the number of reactors, e.g. in Europe, and their envisaged service life of roughly 40 years, an inventory of several tons will be seen to accumulate over the years.

As mentioned above, reactors with hard neutron spectra, in which most nuclear reactions are initiated by high-energy 'fast' neutrons, lend themselves particularly well to burning MAs, such as neptunium and americium. As evident from Fig. 5, this applies in particular to the nuclides which are practically non-fissionable in thermal reactors, such as ^{237}Np, ^{238}Pu, ^{240}Pu. For ^{239}Pu and ^{241}Pu, likewise, fast reactors are to be preferred over thermal reactors because they have a higher ratio of fission to capture reactions for neutrons. Moreover, they can be designed so as to achieve a higher rate of plutonium burning per unit of energy generated than, e.g. MOX LWRs. For this reason, a suitable fast burner reactor for the transmutation of plutonium and MAs was designed at the French Research Center of Cadarache, which closely cooperates with FZK in numerous fields. FZK made important contribu-

tions to a number of extensive detailed investigations into this newly developed concept briefly referred to as CAPRA[4]. These studies are to demonstrate that this type of reactor, which has roughly the same total power as the SUPERPHÉNIX 1 (SPX1) reactor operated in France for the past 10 years with German participation, can achieve a plutonium burning rate of approximately 600 kg of Pu per GW_e year. At the same time, it is to be demonstrated that CAPRA has safety features comparable to those of the SPX1.

In view of the variety of reactor concepts proposed in the past, CAPRA represents only one special variant of the many fast reactor designs. It is for this reason that the existing data and computation methods were sufficient to allow the potential of this concept to be estimated. However, the design was characterized by a number of special features, which had to be studied in detail. To burn as much plutonium as possible, the generation of fresh plutonium from uranium must be reduced. In CAPRA, this is achieved by decreasing the mean uranium concentration in the core. The absence of this material, which acts as a neutron absorber, would unduly raise the neutron multiplication factor of the reactor. In order to restore the equilibrium between neutron production and neutron losses necessary for operation of the reactor, CAPRA must reestablish the neutron balance by increased neutron leakage losses from the reactor core and, perhaps, by additional absorbers. For this purpose, void volume is incorporated in the reactor and material which is relatively transparent to neutrons (a so-called diluent) is used so that the neutrons can escape from the reactor core quite easily.

This dilution is achieved by three measures, (M1) fuel pellets are manufactured with a bigger center hole, so-called hollow pellets; (M2) the fuel elements not only contain the 336 fuel rods but an additional 133 rods containing no fuel, which can either be empty or filled with steel or a suitable moderator material; and (M3) finally, the reactor core with a total of 451 elements is not filled with fuel elements and control and shutdown elements only, but also contains some 50 diluent elements. The material they contain can be adapted to the desired purpose, i.e. increase neutron leakage losses or neutron losses due to absorption. In addition, also proper positioning of these special elements can influence the power distribution.

By definition, CAPRA has been designed to burn dirty plutonium generated, e.g. by recycling UOX LWR plutonium in MOX LWRs. Moreover, also MAs are to be transmuted in CAPRA. Dirty plutonium and MA-bearing fuel deteriorate important safety characteristics of the reactor. The change in multiplication properties (a) as a result of a change in coolant density or of a complete loss of the sodium coolant (sodium void effect), or (b) as a result of a change in fuel temperature (Doppler effect). As was shown in detailed studies by the partners in the CAPRA project, safety characteristics comparable to those of conventional fast reactors can be restored by using an effective moderator as the filler material. The most promising candidate at the present time is $^{11}B_4C$. To avoid unnecessary confusion, it should be pointed out expressly that it is not the highly neutron absorbing boron isotope ^{10}B, but only the very weakly neutron absorbing ^{11}B which is to be used in this boron carbide.

The envisaged objectives of a plutonium incineration rate of approximately 600 kg per GW_e year, and transmutation of a sufficient quantity of neptunium can be met in the CAPRA design to a high degree. The transmutation of americium, which is to be irradiated in special elements near the outside of the reactor core, does not yet quite come up to expectations. This aspect will be mentioned again below. The transmutation caused by long-term neutron exposure of americium, and the corresponding changes in nuclide masses, are shown in Fig. 6. Fig. 7 indicates that, as a result of radioactive decays, considerable changes in mass will occur over prolonged periods of time even after the end of irradiation.

Finally, attention will be drawn to a complication almost unavoidable in the transmutation of MAs. The spent fuel elements contain a fraction, in some cases considerable, of the ^{238}Pu isotope. Small quantities of this isotope are already being

[4] CAPRA stands for Consommation Accrue de Plutonium dans les Rapides meaning increased consumption of plutonium in fast reactors.

used in isotope batteries, e.g. in space technology. However, the amounts arising in MA transmutation clearly exceed the present demand for isotope batteries. The relatively high concentrations of ^{238}Pu in the spent fuel elements dedicated for MA transmutation could make reprocessing of these fuel elements difficult. Certainly, it will also be indispensable for these reasons to build remotely controlled fuel fabrication facilities. Highly automated handling in fuel reprocessing and refabrication already constitutes part of the present customary procedures. In order to reduce the exposure of the operating personnel, and also for cost reasons, remote-handling technologies will have to be advanced even further in the future. It is to be expected, therefore, that the higher radiation levels caused by the transmutation of MAs will slightly aggravate fuel processing. However, if the necessary provisions are made in plant design, MA transmutation is not likely to require new process steps to be developed from scratch. As mentioned above, the additional separation steps for recovery of the MAs tested on a laboratory scale and in pilot plants, respectively, still need to be scaled up to industrial-size reprocessing plants. For the sake of completeness, it should also be added that in addition to the usual PUREX reprocessing technique, which has been proven worldwide, other processes may be applied in recycling spent fuel, too. A considerable background of experience exists for molten salt processes and pyrochemical as well as pyrometallurgical processes. More recent studies deal with the development potential of these reprocessing techniques which, compared with the PUREX process, have not yet reached maturity.

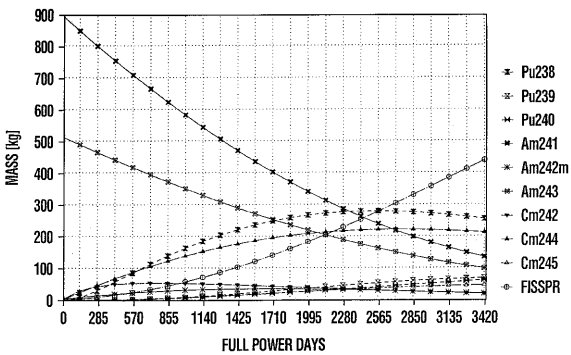

Fig. 6. Changes in nuclide masses during long-term irradiation of americium in the first reflector row of the CAPRA design. Over the residence time of 3420 full-power days, the steel of the fuel rods and the fuel elements suffers pronounced radiation damage of approximately 200 dpa (displacements per atom).

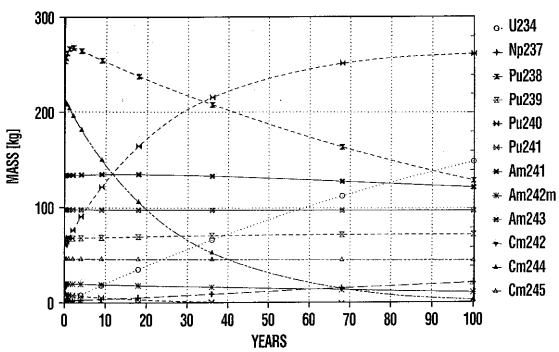

Fig. 7. Long-term interim storage of material unloaded as in Fig. 6 drastically reduces the masses of ^{242}Cm and ^{244}Cm. The initial rise in ^{238}Pu mass results from the rapid ^{242}Cm decay, which is difficult to recognize. Moreover, the increase in ^{240}Pu due to ^{244}Cm decay, and that of ^{234}U by ^{238}Pu decay, are clearly evident.

6. Mixed transmutation strategies for a nuclear reactor park

Worldwide, LWRs produce the largest share of energy generated from nuclear fission. It is not unreasonable, therefore, to exploit the potential of these running plants for transmutation. At present, most of them are fueled with enriched uranium. If a sufficient number of plants equipped with this type of reactor were modified for operation with plutonium-bearing fuel (either MOX fuel consisting of PuO_2 and UO_2, where depleted uranium could be used, or MOX fuel with relatively dirty plutonium plus uranium enriched to the appropriate level), plutonium arisings could be kept roughly at a constant level. However, a number of objections can be raised against this concept, (a) natural uranium reserves are not unlimited, and so a long-term concept must be prepared which does not necessarily have to use enriched uranium and which, moreover, reduces the consumption of natural uranium (pro-

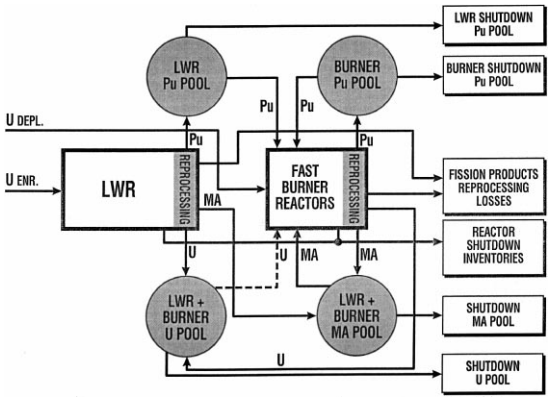

Fig. 8. Mass flows in a mixed nuclear power park of LWRs and fast burner reactors. Under the assumption of the operation of this park terminating at a specific date, also the amounts of material existing in the reactors and in the shutdown pools must be taken into account when considering the radiotoxicity potential.

tection of resources); (b) the necessary ratio of MOX- to UOX-reactors is quite high, approximately 3:5, i.e. the net consumption of plutonium by three MOX reactors roughly compensates the plutonium production by five UOX reactors; a smaller percentage of MOX reactors therefore would be desirable in the interest of economic viability; and (c) the concept described above, which is restricted to the use of LWRs, is unable to cope with the continuously rising volume of MAs.

For all these reasons, mixed strategies exploiting the benefits of different reactor types must be found. Only one example out of the large variety of possible combination strategies will be explained here, namely a European nuclear park with an aggregate power of 120 GWe, corresponding to 80 LWRs of 1500 MWe each, for instance, 84 reactors of the Philippsburg-2 type or 82 reactors of the French N4 type. Each N4 reactor annually produces approximately 250 kg of plutonium and some 15 kg each of neptunium and americium. The schematic diagram in Fig. 8 shows the mass flows assumed for the scenario under study, based on a common transmutation strategy of LWRs and fast burner reactors of the CAPRA type. Simplifying assumptions were made for the model calculations, which simulated real approaches only in an approximate way, but did not completely correspond to the details of actual operating practice. Economical considerations based on present or expected interest rates and past depreciations, respectively, surely will lead to deviations in specific cases from the simple computational model chosen. The idealized approach selected here is meant to show primarily the existing possibilities and the resultant consequences so that the necessary preconditions as well as any bottlenecks or vulnerabilities can be identified.

The following assumptions are essential to the scenario under study (further details are described, e.g. in Wiese, 1997), (1) power production is kept at a constant level; depending on the availability of Pu, the N4 PWRs will be replaced successively by CAPRAs; (2) all the plutonium from N4 PWRs and CAPRAs will be made available to operating and new CAPRAs, respectively; (3) all of the neptunium will be added homogeneously to the CAPRA fuel; and (4) all of the americium produced will be irradiated in CAPRA reflector positions for transmutation.

Special reference must be made to some constraining factors, (C1) the plutonium content in the CAPRA fuel should not exceed 45% because, otherwise, there may be considerable solubility problems in reprocessing the spent fuel; (C2) the fuel loaded (containing so-called dirty plutonium and neptunium) must not significantly impair the safety characteristics of the CAPRA reactors, i.e. the key reactivity coefficients must remain within acceptable margins (these coefficients mainly characterize the changes in the multiplication factor of the reactor due to changes in the fuel temperature and coolant density, respectively).

The scenario described in Fig. 8 furnishes some important findings. Given an LWR:CAPRA ratio of approximately 89:11% for the aggregate electricity generation of the nuclear park, the LWR plutonium generated can be consumed in the CAPRA reactors. If the CAPRA fuel is regularly added some 5% of neptunium, the neptunium produced in LWRs in the above split of power generation can be destroyed in the CAPRAs. However, some restrictions should be mentioned.

(R1) The irradiation of americium in the first reflector row of the CAPRAs assumed here is not

sufficient to prevent americium arisings from increasing. This is due mainly to the rather large quantities of ^{241}Am produced by the decay of ^{241}Pu. (R2) The quantity of plutonium recovered from the CAPRAs will increase with time, and the quality of this plutonium irradiated in CAPRAs will deteriorate.

The first restriction can be mitigated or removed by several measures, (M1) also the second reflector row in the CAPRAs is used for Am irradiation. Because of the lower neutron density, this means longer in-pile times of the respective fuel elements. (M2) The addition of moderating materials can reduce the mean neutron energy; as the reaction probability increases with decreasing neutron energy, this allows higher conversion or fission rates, respectively, to be achieved.

With respect to the second restriction, it should be pointed out that a specific plutonium isotopic composition was assumed as a design basis for the CAPRA reactors. If fuel with much dirtier Pu were to be loaded, this design would have to be modified. CAPRA reactors are very flexible designwise, and there is ample room for design modifications. If the plutonium unloaded from the CAPRAs is not to be accumulated, various options may be followed, (O1) the dirty plutonium recovered is recycled in CAPRAs specially modified for the purpose; of course, this increases the CAPRA share in electricity generation. (O2) In addition, MOX LWRs are used to burn plutonium, which may be loaded, for instance, with relatively clean plutonium from UOX LWRs.

Another option should be mentioned briefly, (O3) the use of uranium-free fuel can prevent fresh plutonium from being generated during reactor operation. Consequently, the rate of plutonium burning can be increased to the theoretical maximum of approximately 1000 kg per GW_e year. As oxide fuel cannot be used in this application because of the solubility problems referred to above, the potential and the properties of nitride fuel, specifically of PuN, are being studied again with great intensity in various countries. This concept, which appears to be promising at first sight, still requires extensive research and development efforts. In addition to time-consuming in-pile and reprocessing experiments, also the safety characteristics of uranium-free reactors must be analyzed, for it must be ensured that the Doppler effect in conventional reactors caused primarily by ^{238}U can be replaced to a sufficient extent by the ^{240}Pu fraction of dirty plutonium.

Other options are the accelerator-driven subcritical facilities (Broeders and Broeders, 2000), whose transmutation potential is evaluated in (Salvatores et al., 1997). In the same reference it was shown that a change from the uranium to the thorium fuel cycle could greatly reduce the production of plutonium and of other transuranium elements. Comparisons of the long-term radiotoxicity of the waste arising in both fuel cycles show no clearcut advantage of the thorium cycle (Salvatores et al., 1995, 1997), as the long halflives of ^{233}U, ^{234}U, and ^{231}Pa, as well as their high radiotoxicity, have a negative impact on the thorium cycle.

Following a Japanese proposal (Mukayama, 1997), the so-called double strata concept will be included in the systems studies within the French program of radioactive waste treatment (Salvatores et al., 1996). In order to minimize the overall cost, a two-tier approach is to be taken to burn large quantities of plutonium and neptunium mainly in conventional thermal or fast reactors, and to burn the smaller arisings of the americium and curium MAs, in a few specially designed accelerator-driven systems (ADS).

7. Summary and outlook

Generating nuclear power by fissioning heavy nuclei gives rise to radioactive fission products. With a few exceptions, these are relatively short-lived. Consequently, the radiotoxicity of fission products has dropped drastically after a period of approximately 500 years. In many instances, this level is considered practically safe, and normal protective measures could be sufficient which are comparable to those applying to hazardous waste.

Nuclear power generation also gives rise to artificial heavy isotopes. In the LWR line now mostly used, these are primarily plutonium, neptunium, americium and curium. The decision whether the spent fuel elements from these reac-

tors are to be disposed of directly or reprocessed may differ depending on the priorities attached to specific evaluation criteria. In Germany, for instance, the amended version of the Atomic Energy Act not too long ago fundamentally changed the legal situation of direct disposal. Right now, direct disposal is regarded by many electricity utilities as the economically most viable solution. In that case, an enormous radiotoxicity potential would be accumulated in repositories, which no licensed locations have as yet been identified for. Should nuclear power generation be considered a transitory episode in the history of mankind, this could be the appropriate option.

Under ethical aspects and, particularly, in view of the long-term use of nuclear fission reactors, strong doubts and concerns are expressed about the justification and viability of this solution. If one accepts the need to recycle artificially produced nuclides, primarily plutonium and the so-called minor actinides, i.e. neptunium, americium, curium (also for reasons of resource utilization and reduction of the long-term radiotoxicity of the arisings of transuranium nuclides), burning these nuclides in conventional LWRs and in specially designed fast burner reactors represents a technology available for use right now by which the desired goal can be achieved in the long term. Strategic studies have shown that this can be put into effect on a medium term only if reactors burning plutonium on a major scale of the MOX LWR and fast burner types, such as CAPRA, are commissioned in the near future.

The reactor physics design of the plants suitable for this purpose can be performed on a reliable basis.

Optimization and safety studies of new concepts, however, still require a certain amount of research and development work. A considerable development effort must be mounted also for the most complete possible recycling, with minimum waste arisings especially of the minor actinides. This also applies to the remote handling techniques and to any isotope separation techniques, which may have to be made available.

If nuclear fission reactors are considered the best choice to the supply energy to future generations in the industrialized and developing countries, or if they are accepted as the lesser evil, then, from the present point of view, the transmutation of transuranium nuclides and longlived radiotoxic fission products is the only feasible way to limit the effects of nuclear power production to a tolerable and fair level for generations to come.

References

Beauvy, M., 1995–1996. Les composés d'actinides à l'état solide. CLEFS CEA No. 31, Hiver, pp. 19 f.

Bouquiun, B., (Ed.), 1996. Le futur du Retraitement: Une Synergie de Procédés Améliorés et d'Approches Nouvelles, CLEFS CEA No. 33, Automne, p. 38 f.

Broeders, C.H.M., December 1996. Investigations Related to the Buildup of Transurania in Pressurized Water Reactors, FZKA 5784.

Broeders, C.H.M., Broeders, I., 2000. Neutron physics analyses of accelerator-driven subcritical assemblies. Nucl. Eng. Des. 202 (2–3), 209–218.

Chebeskov, A.N., Kagramanyan, V.S., Malenkov, A.V., 1996. Problems of excess plutonium utilization in nuclear reactors, NATO Advanced Research Workshop on Advanced Nuclear Systems Consuming of Excess Plutonium, October 13–16, Moscow, Russia.

Deutsches Atomforum/Arbeitskreis Technik, 1996. PLUTONIUM-die Fakten. INFORUM Verlags- und Verwaltungsgesellschaft, Bonn.

GSF (Ed.), 1989. Plutonium; mensch und umwelt, Ein Magazin der Gesellschaft für Strahlen- und Umweltforschung, 6. Ausgabe, München.

Koelzer, W. (Ed.), 1989. Plutonium, KfK 4516.

Koelzer, W., 1997. Lexikon zur Kernenergie. FZK, Stabsabteilung, Öffentlichkeitsarbeit, pp. 208–209.

Métivier, H., Colle, C., Germain, P., 1995–1996. La géochimie des actinides, CLEFS CEA, No. 31, Hiver, p. 59.

Moses, D.L., Chebeskov, A.N., Matveev, V.I., Maltysev, V.V., Vasiliev, B.A., 1998. Plutonium disposition in the BN-600 fast-neutron reactor at Beloyarsk. Proceedings of the Third Topical Meeting on DOE Spent Fuel and Fissile Materials Management. Charleston, SC, USA, November 8–11, p. 597.

Mukayama, T., 1997. Importance of the double strata fuel cycle for minor actinide transmutation. Proceedings of the third International Information Exchange Meeting on Partitioning and Transmutation. OECD-NEA/PT Report No. 13, Cadarache, December 1997.

Oi, N., 2000. Plutonium challenges, changing dimensions of global cooperation. http://www.iaea.or.at/worldatom/inforesource/bulletin/bull401/article3.html.

Salvatores, M., Slessarev, I., Uematsu, M., 1994. A global physics approach to transmutation of radioactive nuclei. Nucl. Sci. Eng. 116, 1.

Salvatores, M., Slessarev, I., Uematsu, M., 1995. Physics characteristics of nuclear power systems with reduced long-term radioactivity risk. Nucl. Sci. Eng. 120, 18.

Salvatores, M., Schapiro, J.P., Mouney, H., June 1996. French programs for advanced waste management option. In: Condé, H. (Ed.), Proceedings of the Second International Conference on Accelerator-Driven Transmutation Technologies and Applications, vol. I. Kalmar, Sweden, p. 60.

Salvatores, M., Slessarev, I., Tchistiakov, A., Ritter, G., 1997. The potential of advanced-driven systems for transmission or power production using thorium or uranium fuel cycles. Nucl. Sci. Eng. 126, 333.

Salvatores, M., Spiro, M., Barbier, F., Poitevin, Y., 1997. Les Systèmes Hybrides, CLEFS CEA No. 37. Hiver, p. 15, 1997–1998.

Schlosser, G.J., Krebs, W.D., Urban, P., 1999. Experience in PWR and BWR mixed-oxide fuel management. Nucl. Technol. 102, 54.

Spektrum der Wissenschaft, 1997. Dossier 1/97. Radioaktivität.

Steyn, J.J., 1993. Weapons material and the commercial fuel cycle. Trans. ANS 68, 72.

Strachan, D.M., Bakel, A.J., Buck, E.C., Chamberlain, D.B., Fortner, J.A., Mertz, C.J., Wolf, S.F., Bourcier, W.L., Ebbinghaus, B.B., Shaw, H.F., Van Konynenburg, R.A., McGrail, B.P., Vienna, J.D., Marra, J.C., Peeler, D.K., 1998. The characterization and testing of candidate immobilization forms for the disposal of plutonium. Proceedings of the WM '98 Conference On Waste Management. HWL, LLW, Mixed Wastes and Environmental Restoration — Working Towards a Cleaner Environment. Tucson, Arizona, USA, March 1–5, 1998. Am. Nucl. Soc. (1999). La Grange Park, IL, 1 CD-ROM.

Volf, V., 1989. Plutoniumrisiko: Abschätzung und Kontrolle. Biologische Grundlagen und organisatorische Maßnahmen, KfK 4592.

Weinberg, A.M., 1994. The First Nuclear Era: The Life and Times of a Technological Fixer. AIP Press, Woodbury, NY, p. 40.

Wiese, H.W., 1997. Actinide transmutation properties of thermal and fast fission reactors including multiple recycling. International Conference, 'Actinides '97', Baden-Baden, Germany. J. Alloys Compounds 271–273 (1998), 522–529.

Wiese, H.W., 1999. Equilibrium in pools of PWRs and burner reactors with homogeneous recycling of plutonium and minor actinides. Proceedings of Fréderic-Joliot/Otto-Hahn-Summerschool, held at Forschungszentrum Karlsruhe. August 18–27, 1999.