Burning of actinides: A complementary waste management option?

Partitioning and transmutation of actinides and fission products may serve as an additional tool in waste management strategies

by L.H. Baetsle

Worldwide nuclear electric capacity amounts to nearly 340 gigawatts-electric (GWe) and produces spent fuel roughly amounting to 9000 tonnes heavy metal (tHM) per year. Each tonne of spent fuel contains about 10 kg of transuranium (TRU) actinides, of which 0.8 kg are minor actinides, and 30 kg of fission products, including 4 kg that are long-lived nuclei (having half-lives greater than 30 years).

The fate of the spent fuel depends to a great extent on the national fuel cycle policy. For one-half of the world's capacity of nuclear power plants, the policy calls for reprocessing, plutonium recovery, vitrification of residues, and disposal of wastes. Among the countries following this course are France, United Kingdom, Japan, Germany, Belgium, Switzerland, the Commonwealth of Independent States, and countries formerly in the Soviet alliance. Large reprocessing facilities have been constructed and are in operation in France and the United Kingdom while large plants are under construction in Japan and Russia.

The other half of the world's nuclear capacity produces spent fuel as a waste product. The so-called "once-through cycle" is being pursued in the United States, Canada, Sweden, Spain, and some other countries. The long-term storage of spent fuel in engineered facilities is the present trend. Storage is to be followed by disposal in suitable geologic formations.

The TRU actinides are building up at a rate of about 90 tHM per year. Approximately 45 tHM will remain occluded in the spent fuel structures, leaving about 45 tHM available; 92% as recycled plutonium and 8% as minor actinides (neptunium, americium, curium) immobilized in vitrified waste.

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The vitrified waste is stored in engineered facilities awaiting final disposal in underground repositories. Disposal of vitrified high-level waste (HLW) containing the minor actinides, or spent fuel with plutonium and minor actinides, are from the environmental point of view very similar at least during the first millennia. Beyond 10 000 years — the technical lifetime of an underground repository — spent fuel with its full load of plutonium becomes the dominant environmental hazard.

Interest in partitioning and transmutation

Two decades ago the question was already raised: Can we avoid the long-term hazard associated with TRU actinides and long-lived fission products? Important research and development (R&D) programmes were run in the European Community and in the USA to investigate this issue. The success of these efforts was very limited. It soon became apparent that it was practically impossible to eliminate all TRU actinides and that some fission products, e.g. technetium-99, caesium-135, and iodine-129, are equally important in the dose-to-man evaluation on the horizon of a million years.

However, there is renewed interest in partitioning and transmutation (P&T), largely because of difficulties encountered throughout the world in finding suitable geologic formations in locations which are acceptable to the public.

In 1988, the Japanese Atomic Energy Commission launched a very important and comprehensive R&D programme. *(See the following article.)* It is aimed at the elimination of long-term hazards resulting from nuclear power production and at the optimal use of resources. This initiative aroused interest from some other major nuclear countries, for example France, to set up research programmes to improve or adapt the currently used PUREX process. The objective

was to reduce the plutonium content in HLW and to eliminate minor actinides by installing new or additional extraction steps.

The P&T strategy can only be implemented by a fuel-cycle policy incorporating reprocessing as a key step through which all major actinides (uranium, plutonium) are recycled, and which is capable of isolating the minor actinides and some long-lived fission products from the effluent stream in order to prepare them for the subsequent transmutation steps.

General strategies and schemes

The general strategy of introducing P&T as an alternative waste management option is based on the radiological benefit which is expected from such a venture. The selection of the actinides and long-lived fission products which are beneficial to eliminate by transmutation depends upon a number of technical factors, including hazard and decontamination factors, and the effect of geological confinement.* Long-lived fission products are much less toxic than actinides on the basis of hazard indexes once strontium-90 and caesium-137 have decayed, that is after about 600 years. *(See tables on page 34 for a ranking of actinides and fission products by their hazard factors.)*

There are two ways to approach the separation of minor actinides and long-lived fission products from reprocessing streams: by modifying the current processes in order to reroute the critical nuclides into a single solution, for example high-level liquid waste, and use this as a source for partitioning processes; and by extension of the conventional PUREX process to all minor actinides and long-lived fission products in second generation reprocessing plants.

Prior to the implementation of one of these schemes, it seems obvious to improve the separation yield of plutonium from HLW within the presently running plants. Rerouting nuclei into single product or waste streams is very important for neptunium which occurs in many different process streams.

The implementation of a step for technetium recovery is not only important in a P&T strategy but is also a means to reduce the uranium-plutonium contamination.

There are a number of reagents for partitioning of actinides, the most promising of which up to now is known as CMPO, which can be used in conjunction with TBP in the so-called TRUEX process. While additional work is needed, it may be anticipated that this or a similar separation technology will, if sufficiently funded, come up with a dependable process compatible with the PUREX process. The partitioning step will produce single elements or groups of elements that are the material sources for transmutation processes (neptunium, americium, curium), or that may become strategic resources for the future (technetium and platinum group elements).

The ranking of fission products is not altered by the role of geologic confinement. Technetium-99 and iodine-129 remain high on the list of the fission products to be examined in a P&T option because of their mobility in the geosphere.

As a conclusion, one may postulate that a P&T strategy ought to provide as much radiological protection to humanity as geologic disposal does. The selection of nuclides to be separated and transmuted, and to what extent they have to be eliminated, will be determined by the trade-off between a decreasing confidence in the merits of geologic disposal as the time period becomes unimaginable on a human scale, and the increase in waste management costs from improved reprocessing and P&T.

Actinide P&T is not an alternative long-term waste management option. Rather, it is a complementary technique to geologic disposal capable of further decreasing the radiological impact of the fuel cycle over the very long term.

Recycling of minor actinides

If partitioning were to be implemented in the large reprocessing units (La Hague, Sellafield, Rokkashomura, etc.) a total output of about 1700 kg of neptunium-237 and 1500 kg of americium (including some kg of curium) would become available each year. This corresponds to roughly 44% of the total world output.

According to the transmutation method used, the minor actinides would be conditioned as oxides or metals in specially equipped fuel fabrication facilities which have to be built for that purpose. Depending on the type of recycling (homogeneous or heterogeneous) the dedicated fabrication capacity ought to be in the range of 68 to 85 tonnes of mixed oxide (MOX) per year. These facilities would be closely associated with the reprocessing plant activities.

The production of metal fuel is based on a pyrometallurgical refining process which is presently at the bench-scale development stage in the USA and Japan. A total capacity of 80 tHM per year of dry reprocessing-fuel fabrication would have to be built in order to treat the output of minor actinides from the USA and some other countries.

* Comprehensive technical details are available from the author.

Actinides in high-level waste

	ICRP-61	1/tHM
1	Am-241	$3.3 - 12.3 \times 10^{13}$
2	Am-243	$1.8 - 2 \times 10^{12}$
3 (1%)	Pu-240	$7.6 - 8 \times 10^{11}$
4 (1%)	Pu-239	$3.2 - 3.5 \times 10^{11}$
5	Np-237	$4.7 - 6.4 \times 10^{10}$
6	Cm-246	$2.4 - 2.7 \times 10^{10}$

Fission products

	Nuclide	1/tHM
1	Sr-90	$3.9 \times 10^{12} - 2.13 \times 10^{4}$
2	Cs-137	$3.8 \times 10^{12} - 3.66 \times 10^{4}$
3	Tc-99	1.6×10^{9}
4	Sn-126	6.6×10^{8}
5	I-129	5.8×10^{8}
6	Cs-135	1.3×10^{8}
7	Zr-93	9.4×10^{7}

Notes Rankings are on the basis of criterion in ICRP-61 of the International Commission on Radiological Protection They are based on the volume of drinking water required to dilute a mixture of radionuclides to acceptable drinking water limits The hazard rankings for actinides are as they would occur in HLW cooled for 200 to 1000 years

Ranking of long-lived fission products and actinides by radiological hazard

However, these facilities would not be connected to the conventional reprocessing activities. They would be an integral part of fast-reactor complexes for burning actinides with a dedicated capacity depending on the reactor power of each site.

Transmutation of minor actinides

In principle it is possible to transmute the minor actinides in existing nuclear plants. However, the transmutation is slow and produces essentially heavier nuclides, and repeated reuse of spent fuel materials is difficult if homogeneous recycling is adopted. The heterogeneous recycling of minor actinides in specially designed fuel elements has not yet been developed sufficiently.Transmutation of americium is possible under certain conditions.

The most efficient transmutation of minor actinides occurs in fast neutron reactors capable of transforming the actinides into fission products. A very important point to note is the large inventory of minor actinides of a fast reactor. This permits the transfer of almost the entire annual output to an "incineration" reactor. However, the net annual incineration throughput is rather limited to approximately 5%.

Specially designed reactors are under study to burn plutonium and minor actinides produced at a light-water reactor station. The Integral Fast Reactor (IFR) concept developed in the USA and the Minor Actinide Burner Reactor (MABR) studied in Japan are two new concepts which aim at the nuclear incineration of actinides.

Approaches and alternatives

In conclusion, P&T is becoming an additional tool in the overall waste management strategy to reduce the radiological impact of actinides and long-lived fission products. However, it is not a full alternative to geological disposal.

Improved reprocessing can significantly reduce the plutonium content in HLW. Some partitioning techniques for americium and curium are promising and rerouting of neptunium in conventional reprocessing is beneficial.

A comprehensive scheme of partitioning, which includes all minor actinides and long-lived fission products, is still a remote aim and needs intensive R&D efforts.

Transmutation of separated nuclides into fuel pins or irradiation targets is a very important step in the overall P&T strategy. This further needs a thorough technological and economical analysis in the frame of a comprehensive transmutation technology.

Transmutation is a time-consuming process which requires reactors with high and/or energetic neutron fluxes. Such neutron fluxes are available in high-flux reactors and fast reactors. The transmutation yield in tailor-made reactor types, such as the IFR or MABR, is relatively high. Nonetheless, the time period to destroy the nuclides is very long (20-30 years) depending on cooling- and fuel-cycle times. Accelerator driven reactors provide irradiation facilities with fluxes which are roughly one order of magnitude higher and could shorten the irradiation periods significantly.

The economics of the P&T approach needs a very thorough analysis. This is important since improved reprocessing, chemical partitioning, target fabrication and fuel development, fast reactor development, and recycle operations for minor-actinide fuel require the construction and operation of important nuclear fuel cycle facilities. They further require the development of mature partitioning and transmutation technologies applicable to industrial quantities.

These efforts have to be compared with the problems associated with public acceptance of geologic repositories and the delayed hazards from the migration of very long-lived nuclei in geological strata. Efforts to reduce costs for geologic disposal ought to be the economic motor behind P&T, and steps to minimize human radiation exposures should be the driving force for decreasing the long-term radiological impact of nuclear electricity production. ❐