Elimination of waste actinides by recycling them to nuclear reactors

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After a few centuries of radioactive decay the long-lived actinides, the elements of atomic numbers 89–103, may constitute the main potential radiological health hazard in nuclear wastes. This is because all but a very few fission products (principally technetium-99 and iodine-129) have by then decayed to insignificant levels, leaving the actinides as the principal hazardous species remaining. It is therefore at first sight an attractive idea to recycle the actinides in nuclear reactors, so as to eliminate them by nuclear fission.

Nuclear physics calculations show that the process would occur at a sufficient rate and that the neutron economy of the reactor would not be unduly affected by the presence of additional actinides in the fuel elements Hence there are good reasons for examining the idea in detail, and studies have been carried out in a number of countries. These have culminated recently in international conferences at the European Joint Research Centre at Ispra in Italy [1] and at Austin, Texas in the USA [2], as well as in the issue of an IAEA Technical Report entitled *An Evaluation of Actinide Partitioning and Transmutation,* a product of a four-year IAEA Co-ordinated Research Programme [2a], on which the present article is based. The term *partitioning* refers to the separation of the actinides from nuclear fuel cycle wastes, a necessary preliminary step to their introduction into reactors for *transmutation* by nuclear fission. The complete scheme will be referred to as P-T, i.e. partitioning-transmutation.

The actinides mainly involved are neptunium, americium, and curium, which form about 0.1% of typical nuclear fuel on its discharge from the reactor. (In the thorium fuel cycle, which we shall not consider in this short account, protactinium would also be involved.) These species find their way into waste streams when the fuel is reprocessed. They may be called *waste* actinides, whereas uranium and plutonium may be described as *fuel* actinides.

It would make little sense to carry out P-T of the waste actinides if the much larger quantities of plutonium in the fuel cycle were stockpiled indefinitely. Waste actinide recycling must therefore be considered in the context of recycling plutonium for use as a fuel, either in light-water reactors or fast breeder reactors.

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All waste streams containing actinides must be included in the scheme. The most important is high-level waste (HLW) from reprocessing, which contains the bulk of both the fission products and the waste actinides, but a number of other streams also contain significant amounts of actinides, e.g. the discarded fuel cladding and rejected material from fuel fabrication. Efforts are indeed made to decontaminate all these latter streams of both fuel and waste actinides, and to return the recovered actinides to appropriate points in the fuel cycle. These efforts will continue whether or not the waste actinides are recycled to reactors for transmutation, and it would leave the HLW as the sole waste stream containing waste actinides. The P-T concept would then require the further step of partitioning the HLW.

Outline of the scheme

The steps required to achieve P-T of the waste actinides are the following:
- Partitioning of the nuclear wastes to separate the waste actinides in sufficiently pure form.
- Fabrication of reactor fuel elements containing the waste actinides. In principle, the waste actinides might either be incorporated uniformly throughout the fuel (homogeneous recycling) or concentrated in special target elements (heterogeneous recycling), though the latter may involve almost insuperable difficulties, as described below.
- Introduction of the fuel elements containing the waste actinides into reactors for irradiation. Probably any of the principal types of power reactors could be used. The scale of operation is such that large numbers of reactors would be involved.
- Reprocessing of the irradiated fuel elements and repeated recycling of the waste actinides through the whole series of operations.

The main steps in the P-T scheme are shown in the diagram, which also indicates the principal differences from the normal fuel cycle. One consequence of the scheme may be mentioned immediately. As in all instances of recycling, the continued re-introduction of the recycled species into the system leads to a build-up from cycle to cycle in the quantities of these species. Usually the amounts tend to level off after a number of cycles. In the present case this occurs after 5–10 cycles, when the proportions of neptunium and americium have roughly doubled and that of curium (with different production and destruction rates) has risen about ten-fold.

The properties of some of the waste actinides make them difficult to handle. The chief problems are their high emission of heat and of neutrons. Compared with spent nuclear fuel, which is of course also a difficult material to handle, the emission of heat due to radio-active decay is greater by a factor of around 30, and the emission of neutrons is greater by a factor of around 10 000. The latter necessitates substantial shielding and remote operation of all processes in which the waste actinides are present. Dissipation of the radioactive decay heat can be a very severe problem when the waste actinides are present in bulk in concentrated form.
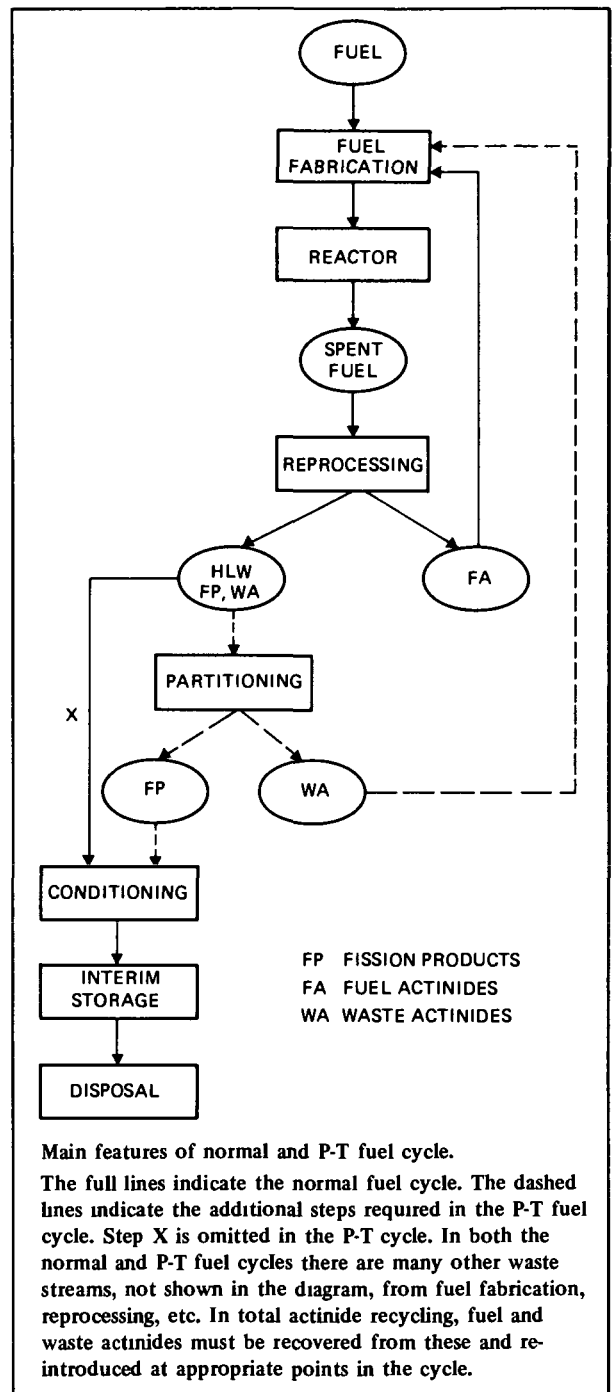
Partitioning

As noted above, it may be presumed that future practice will be to remove actinides from all wastes other than the HLW in order to minimize disposal problems. The further step required for P-T is to remove them from the HLW. There is no special difficulty about this in the case of uranium, neptunium, and plutonium, whose chemistry has unique features which can be exploited to separate them from other species. Americium and curium (and higher actinides) are, however, chemically exceedingly similar to certain fission products, viz the lanthanides, the elements of atomic numbers 57—71. Their separation is notoriously hard to achieve, and the high activity level of the HLW greatly increases the difficulty.

The recovery of the waste actinides is usually envisaged as starting with recovery of residual fuel actinides and neptunium, followed by separation of a combined americium/curium/lanthanide fraction. These steps can be carried out in much the same way as ordinary reprocessing operations. Several alternative methods are available, some of which have been fully tested in the laboratory and to some extent on a plant scale [3]. The feasibility of this part of the P-T scheme has been well established.

The final step is the separation of americium and curium from the lanthanides. Virtually only one reagent has been found which is suitable for the task, a complicated organic compound by the name of di-ethylenetriamine-penta-acetic acid or DTPA for short. This may be used in conjunction with either solvent extraction or cation exchange. For example, if added to the aqueous phase in extraction by a suitable solvent, it will hold back the americium and curium in the aqueous phase, while permitting extraction of the lanthanides into the solvent phase, thus effecting a separation of the species concerned. There is large-scale experience with the use of DTPA in conjunction with cation exchange (though not specifically for actinide P-T), which is regarded as sufficient to establish its technical feasibility [4].

Nevertheless, the process would be very difficult to operate because it has to deal with the lanthanides and waste actinides at high concentration. Their radiations

Main features of normal and P-T fuel cycle.

The full lines indicate the normal fuel cycle. The dashed lines indicate the additional steps required in the P-T fuel cycle. Step X is omitted in the P-T cycle. In both the normal and P-T fuel cycles there are many other waste streams, not shown in the diagram, from fuel fabrication, reprocessing, etc. In total actinide recycling, fuel and waste actinides must be recovered from these and re-introduced at appropriate points in the cycle.

would cause gassing, damage to the ion-exchange beds, and self-heating of the solutions and ion-exchange beds, resulting in complicated plant design and operation and hence considerable maintenance work and exposure of workers to radiation. It is doubtful whether the process could be operated routinely under industrial conditions.

Fuel fabrication

Only recently has much attention been paid to the problems of manufacturing fuel, and fuel elements containing waste actinides [5,6]. In homogeneous recycling it has generally been felt that no great problems

would arise, because spent fuel already tolerates around 0.1% of waste actinides, and the increase envisaged in P-T is only to, say, 0.5%. Nevertheless, a substantial programme would be required to develop fuel fabrication methods and to design and test the fuel elements.

In heterogeneous recycling, when the waste actinides are concentrated in special target fuel elements, which are distributed among the normal fuel elements in the reactor, the problems are much more severe. Heat and radiation emitted by the concentrated waste actinides would interfere with all the operations from preparation of the desired chemical form (presumably oxide) to assembly of the fuel elements from individual fuel pins. The problems would be somewhat mitigated by the need to dilute the waste actinides with, say, magnesia for reasons mentioned below, but would remain very acute. It might indeed not be feasible at all to carry out the assembly operation for target fuel elements for a fast breeder reactor; so far as can be seen the process would have to be carried out remotely in a bath of liquid sodium to provide the requisite cooling, and these seem almost impossible conditions. In the light-water reactor case, with a greater degree of dilution, and water instead of sodium cooling, the problems might be more tractable, though still beyond existing technology.

Transmutation

It seems likely that the transmutation step would be carried out in fast reactors, since they may be the only reactors available in sufficient numbers by the time P-T could be implemented in a major way, several decades ahead. The choice is reinforced by detailed technical considerations, which indicate that they are probably marginally superior to light-water reactors for the purpose [4,7].

The transmutation rates, which of course differ from actinide to actinide and from reactor to reactor, correspond typically to transmutation of half of the quantity of actinides present in 2–3 years; but this time could be doubled if reactor load-factors, out-of-reactor time, etc., are taken into account. To reduce the amount of waste actinides by a factor of 1000 (which would require about ten successive halvings) might thus take about 50 years. This means that the waste actinides must be recycled through the reactors a large number of times in any P-T scheme, since the fuel elements can only remain in the reactor for about 2 years before they begin to deteriorate and must be discharged.

There is then a further consequence, which has to do with actinide losses [7]. In the conventional fuel cycle the waste actinides pass through the cycle once only, and there are small losses to waste steams other than the HLW. Efforts will be made to reduce these, but some loss is inevitable in practice. In the P-T fuel cycle the waste actinides pass repeatedly through the cycle, with losses on each pass, and the total loss is several times that in the

conventional cycle. It would indeed be difficult to avoid 1–2% loss of waste actinides in a P-T scheme.

It appears that the presence of waste actinides would actually be beneficial to the neutron economy of a fast reactor, and would make very little difference in a light-water reactor.

Only preliminary consideration has been given to such topics as fuel/cladding compatibility and the possibility of the central temperature of the fuel pins rising above the melting-point of the fuel. The provisional conclusions [5,6] are reassuring, but further study would be necessary before P-T could be implemented, expecially in the case of heterogeneous recycling. In heterogeneous recycling there would, moreover, be a need to match the power output of the target to the normal fuel elements. This is probably best achieved by mixing the waste actinides with a neutronically inert diluent such as magnesia [5]; the dilution factor required would be about 2 in a fast reactor and about 7 in a light-water reactor.

Hazard evaluation

The purpose of P-T is to reduce the long-term potential radiological hazard of high-level waste by removing the actinides. This is achieved at the expense of increases in other potential hazards and in the complexity of the fuel cycle. It is difficult to strike a balance, partly because the potential benefits are long-term ones affecting the general public, whereas the detrimental effects are mainly short-term ones affecting operators of nuclear plants.

Studies of different disposal methods have brought to light no overriding objection on radiological grounds to the disposal of solid wastes which do contain actinides, either in geological formations or in the deep ocean [4, 8,9,10]. When the most recent values recommended by the ICRP [11] for annual limits are used, neptunium emerges from these studies as the most significant actinide, generally by a large margin, and P-T to remove this species and its precursors could produce a limited benefit. However, such a benefit could usually be obtained much more simply and cheaply by changing the disposal method or conditions. In some circumstances very long-lived fission products (technetium-99 and iodine-129) may contribute significantly to the potential hazards, and may restrict what can be achieved by P-T.

Steady improvement in the reliability of these hazard evaluations is expected as new data become available. Meanwhile the conclusions are generally reassuring and indicate little or no incentive for actinide P-T.

Costs

Estimates of the costs of P-T can only be very approximate at the present stage of development. Nevertheless, there seems to be agreement [4,12] that

they would amount to some 5% of the total fuel cycle costs, which is certainly not prohibitive. However, the prospective saving in the long-term collective dose is also comparatively small, and present indications are that P-T is very unlikely to be cost-effective.

The various studies of P-T appear to converge towards a consensus along the following lines.

● The concept would be technically feasible, though it is doubtful whether the chemical separations required could be carried out routinely under industrial conditions.

● A large and lengthy development effort would be necessary.

● Initial implementation might be possible early next century; general implementation can hardly be envisaged until well into the century.

● Transmutation of the waste actinides would involve a high proportion of a country's nuclear power reactors.

● It would make little sense to transmute the waste actinides unless plutonium was also recycled to reactors and burnt as a nuclear fuel.

● The scheme would not be unduly expensive but would probably not be cost-effective.

● The long-term radiological benefits would be limited and might well be outweighed by the short-term detriment arising from P-T operations.

References

[1]   Proc Second Tech Mtg on *The nuclear transmutation of actinides* Ispra, Italy, April 1980 (CEC, Luxembourg, 1980).
[2]   Intern Conf on nuclear waste transmutation Austin, Texas, USA, July 1980
[2a]   E.R. Irish, *Waste Actinide Partitioning/Transmutation, An Assessment* IAEA-SM-246/63, Vienna, Austria, (1981)
[3]   See, for example, *An Evaluation of Actinide Partitioning and Transmutation,* Appendix C, IAEA Tech Rep to be published.
[4]   A.G Croff, J.O. Blomeke, B.C. Finney, *Actinide partitioning-transmutation programme, final report.* I. Overall Assessment, ORNL-5566 (1980).
[5]   E. Zamorani, J. Cametti, C. Ponti. E. Schmidt, F. Caligara, A. Moreno, *Design of fuel pins containing by-product actinides for irradiation in LWR and FBR power plants* EUR-6817 EN (1980).
[6]   A.G. Facchini, S. Gallone, E. Zamorani, *Considerations on a sol-gel type process as a fuel fabrication route for the actinide recycle* ref [1], p 331
[7]   H.A.C. McKay, M.G. Sowerby, M. Bustraan, J. Montizaan, A. van Dalen, B. Verkerk, The separation and recycling of actinides — a review of the state of the art EUR 5801e (1977)
[8]   Appendices E, F, and G of reference [3].
[9]   W.C. Camplin, P.D. Grimwood, I.F. White, *The effects of actinide separation on the radiological consequences of disposal of high-level radioactive waste on the ocean bed* NRPB-R94 (1980)
[10]   M.D. Hill, I.F. White, A.B. Fleishman, *The effects of actinide separation on the radiological consequences of geologic disposal of high-level waste* NRPB-R95 (1980)
[11]   ICRP publications 26 (1977) and 30 (1979).
[12]   E. Schmidt, *Possible recycle strategies for by-product actinides in a reference scenario of installed nuclear generating capacity in the EC and their valuation* ref [1], p 471

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