Analogies of experience in the United States Transuranium Element Production Program with partitioning and transmutation of transuranic actinides in commercial used fuels

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Abstract

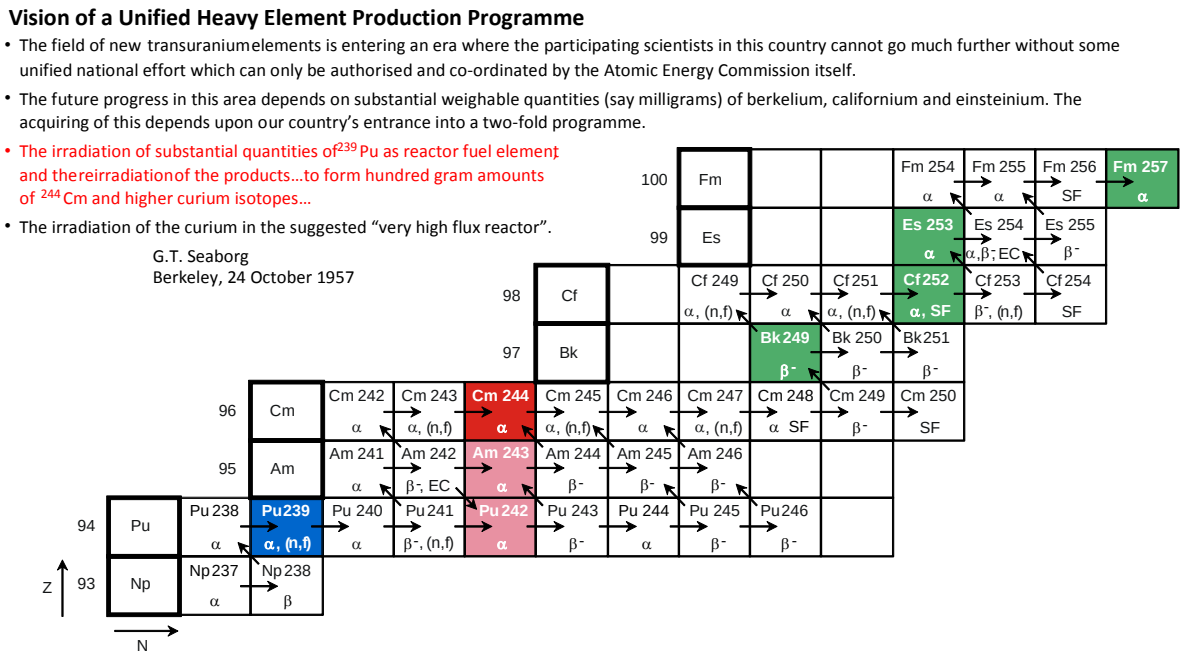
Average cross-section data have been used to compare relative capabilities for transmutation of transuranium element (TRU) isotopes in thermal and fast spectrum irradiations. The comparisons often conclude that fast spectrum irradiation is necessary to consume the actinide elements. However, a different conclusion can be made by comparison with experiences gained during the United States Transuranium Element Production Program, which has occurred at Oak Ridge National Laboratory and at the Savannah River Site (SRS) since the mid-1960s. This experience shows the extremely low production efficiency of converting ^{239}Pu to the heavy actinides (americium, curium, berkelium, californium, einsteinium and fermium) in either a thermal or fast spectrum irradiation. This is because many of the precursor isotopes in the sequential neutron capture steps necessary to produce the heavier actinides are predominately transmuted to fission products. About 98.5% of the plutonium isotopes undergo fission in a fast spectrum irradiation, while ~90% are fissioned in a thermal spectrum irradiation. However, ~92% of the curium isotopes produced undergo fission, such that the transcurium isotopes produced are <1% of the original ^{239}Pu. Thus, the conclusion is that both thermal and fast spectrum irradiations consume >99% of the TRU actinides to fission products. If the goal is actinide burning, then either thermal or fast spectrum irradiations are efficient. However, the rate of actinide destruction may be higher in the thermal spectrum, since the average cross-sections are much larger. This is especially true in high-flux thermal neutron irradiations, such as those used in special configuration production reactors at SRS.

For transmutation of the transplutonium actinides, sometimes called "minor actinides" when produced in commercial reactor used fuels, average cross-section data show that the efficiency of conversion to fission products does not differ greatly between thermal and fast spectrum irradiations. Again, modelling calculations indicate that the rate of conversion may be greater in thermal spectrum irradiation because of the larger cross-sections.

Introduction

Establishment of a unified heavy element production programme in the United States to produce substantial, milligram quantities of newly discovered elements (berkelium, californium and einsteinium), plus picogram amounts of fermium, was the vision of Dr. Glenn Seaborg in October 1957. The production of these elements would begin with the irradiation of substantial quantities (multi-kilograms) of ^{239}Pu and proceed via a series of neutron capture and beta decay reactions as illustrated in the partial chart of the elements shown in Figure 1.

Figure 1: Vision of a unified heavy element production programme



Subsequently, after he was appointed Chairman of the United States Atomic Energy Commission in 1961 by President John F. Kennedy, Dr. Seaborg oversaw the implementation of the United States' heavy element production programme, using the large production reactors and separations canyons at the Savannah River Plant (SRP), and a newly constructed, very high flux reactor and specially designed hot cells at the Oak Ridge National Laboratory.

Transplutonium element yields

The difficulty in producing large amounts of the transplutonium elements is illustrated by the chart of successive neutron captures and beta decays that are required and the huge portion of losses due to fissioning as shown in Figure 2 [1]. For example, in the three neutron captures required for transmutation of ^{239}Pu to ^{242}Pu, 90% of the mass is lost to fission reactions; then, another 9.7% fission loss occurs during the ten additional neutron captures required to transmute ^{242}Pu to ^{252}Cf; and finally, essentially all of the remaining mass is fissioned during the five additional neutron captures required to transmute ^{252}Cf to ^{257}Fm [2].

Heavy element production

Heavy element production was begun by irradiation of substantial quantities of ^{239}Pu in the large production heavy-water reactors at SRP, using a special configuration of the reactor core to enable a peak thermal flux of ~6×10^{15} neutrons per second per square centimetre [3]. Recovery

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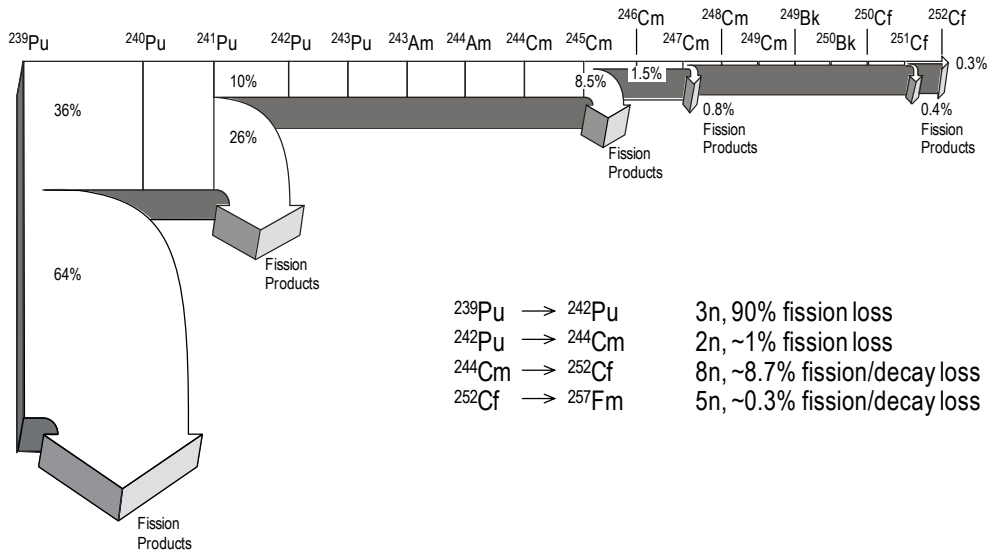
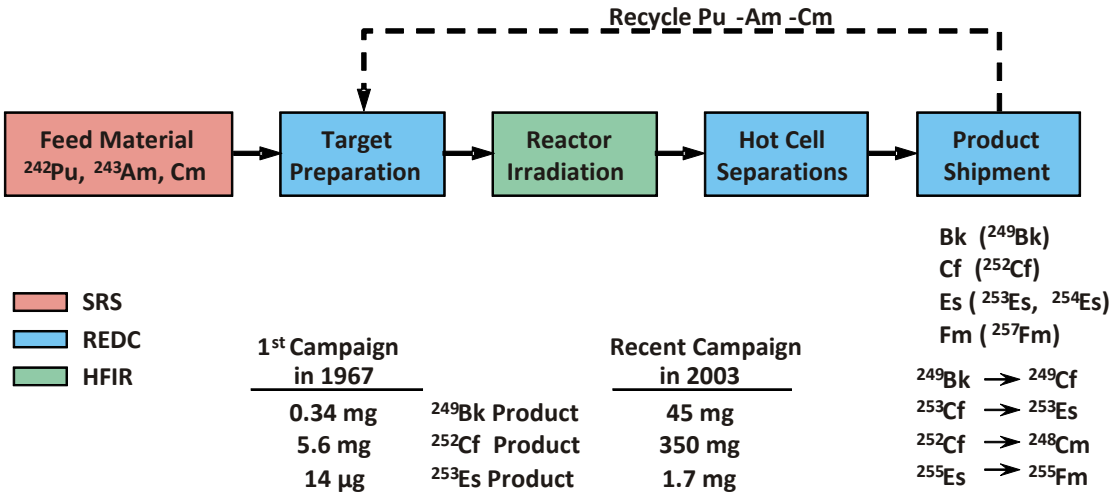
Figure 2: Transplutonium-element yield and fission loss during thermal neutron irradiation of plutonium



of multi-kilogram amounts of ^{242}Pu, ^{243}Am and ^{244}Cm was accomplished in the shielded canyons at SRP and the hot cell facilities at the Savannah River Laboratory. Portions of these products were shipped to Oak Ridge National Laboratory for production of berkelium, californium, einsteinium and fermium in the High Flux Isotope Reactor (HFIR) and the heavily shielded hot cells in the Radiochemical Engineering Development Center (REDC). The production sequence is illustrated in Figure 3.

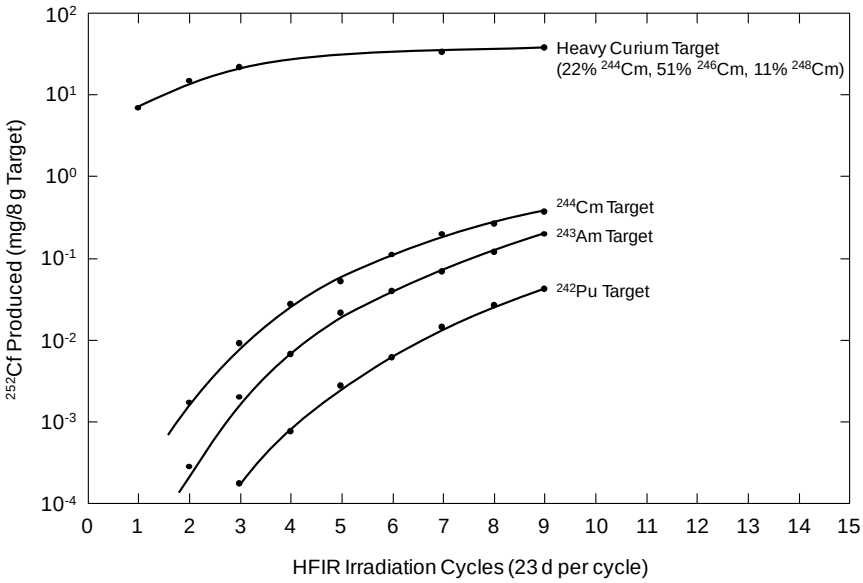
Figure 3: Heavy element production for DOE research programmes and ^{252}Cf neutron sources



The feed materials, initially produced at the Savannah River Site, are fabricated into target rods, irradiated in the HFIR, and processed in the REDC to recover the berkelium, californium, einsteinium and fermium. The residual plutonium, americium and curium are also recovered at the REDC and remotely fabricated into recycle targets. By this means, the isotopic composition of the curium isotopes is altered during several partitioning-transmutation cycles to "near-equilibrium" conditions containing predominantly the heavier isotopes, ^{246}Cm and ^{248}Cm.

The yield of transcurium elements produced by irradiation of these near-equilibrium compositions of curium isotopes is significantly increased over the yields from transmutation of the initial feed materials, as shown in Figure 4. The large difference occurs because ~92% of the mass of the initial feed materials is lost to fissioning of ^{245}Cm and ^{247}Cm.

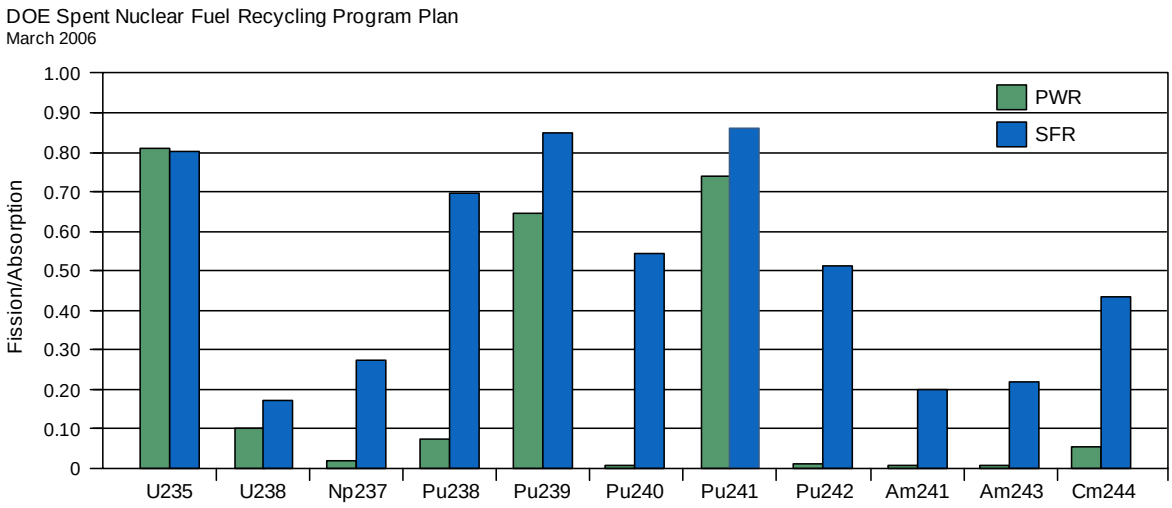
Figure 4: Production rate for ^{252}Cf



Comparison of actinide consumption in thermal and fast spectrum irradiations

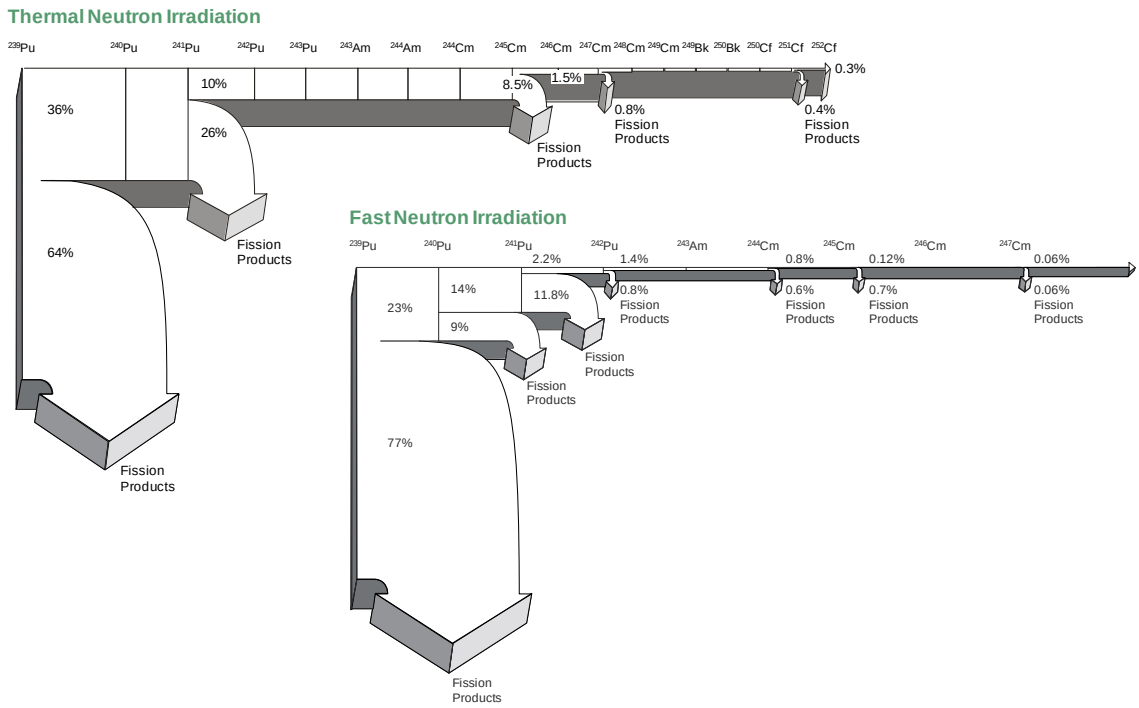
Average cross-section data have been used to compare relative capabilities for transmutation of transuranium element (TRU) isotopes in thermal and fast spectrum irradiations. The comparisons, illustrated in bar chart form in Figure 5, often conclude that fast spectrum irradiation is necessary to consume the actinide elements produced in commercial used fuels in order to minimise the difficulties of high-level waste disposal [4].

Figure 5: Comparison of fission versus absorption in a pressurised water reactor (PWR) and a sodium fast reactor (SFR)



However, by illustrating the same average cross-section data in the form used in the description of difficulties of heavy element production (Figure 2), a more accurate conclusion can be made regarding actinide consumption by fission. Figure 6 shows the fission losses and yields of heavier actinides for both thermal and fast neutron transmutations.

Figure 6: Transplutonium-element yield and fission loss during irradiation of plutonium



About 98.5% of the plutonium isotopes undergo fission in fast spectrum irradiation, while ~90% are fissioned in thermal spectrum irradiation. However, ~92% of the curium isotopes produced undergo fission, such that the transcurium isotopes produced are <1% of the original ^{239}Pu. Thus, the conclusion is that both thermal and fast spectrum irradiations consume >99% of the TRU actinides to fission products. If the goal is actinide burning, then either thermal or fast spectrum irradiation is efficient. However, the rate of actinide destruction may be higher in the thermal spectrum, since the average cross-sections are much larger.

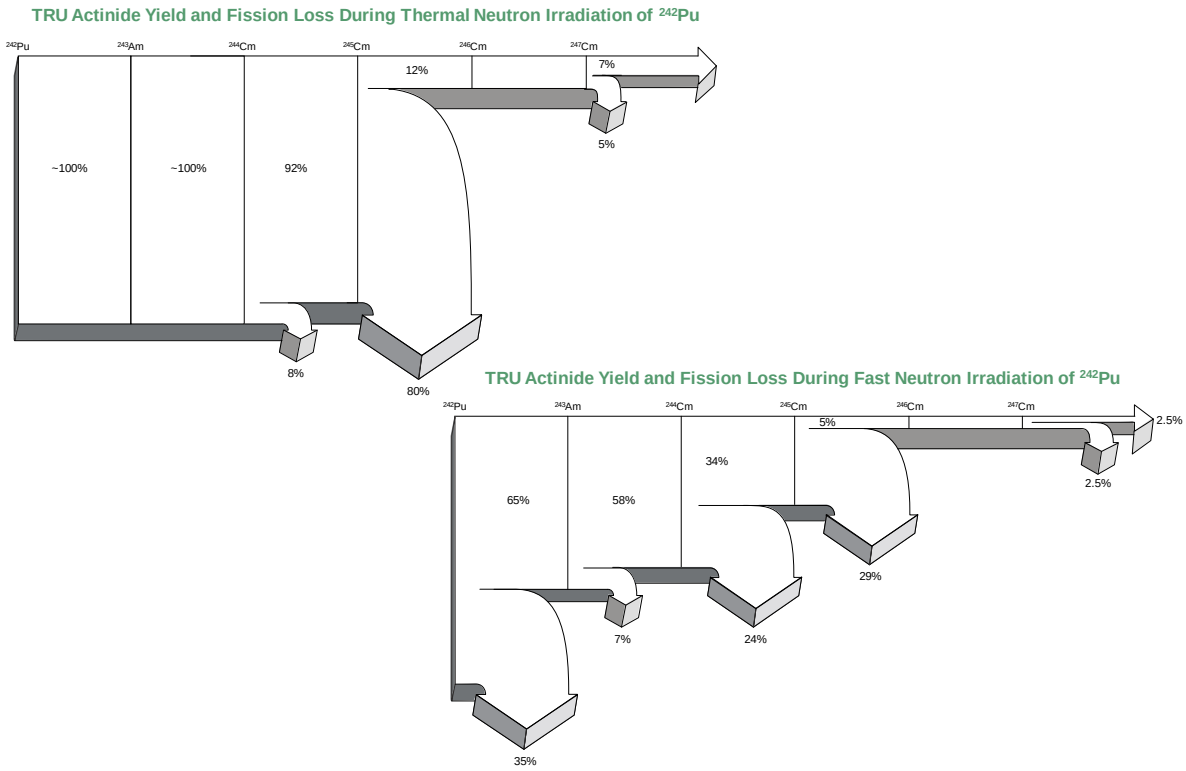
Moreover, the average cross-section data comparison shown in Figure 7 indicates that, if the ^{242}Pu, ^{243}Am and ^{244}Cm isotopes produced in thermal spectrum irradiations are transmuted further, the consumption of actinides is not significantly better in fast spectrum irradiations than in thermal spectrum irradiations. Again, the actual rate of destruction may be higher in the thermal spectrum since the average cross-sections are much larger. This is particularly relevant to "high burn-up" and "deep burn" concepts.

Actinide partitioning-transmutation modelling studies

Actinide partitioning-transmutation (P-T) modelling studies were made to compare performance of thermal and fast reactor irradiations, using the P-T scenarios shown in Figure 8, and beginning with pressurised-light-water reactor (LWR) UO_2 (5% ^{235}U) used fuel that had been irradiated for ~45 GWd/MT and had been decayed for 30 years [5].

The results of the modelling studies showed that, indeed, the destruction of plutonium and the two minor actinides, ^{241}Am and ^{237}Np, occurred to a greater extent during each reactor cycle in an LWR than in a fast reactor (Figure 9).

Figure 7: Transplutonium-element yield and fission loss during irradiation of ^{242}Pu



Figures 8(a) and 8(b): P-T scenarios evaluated

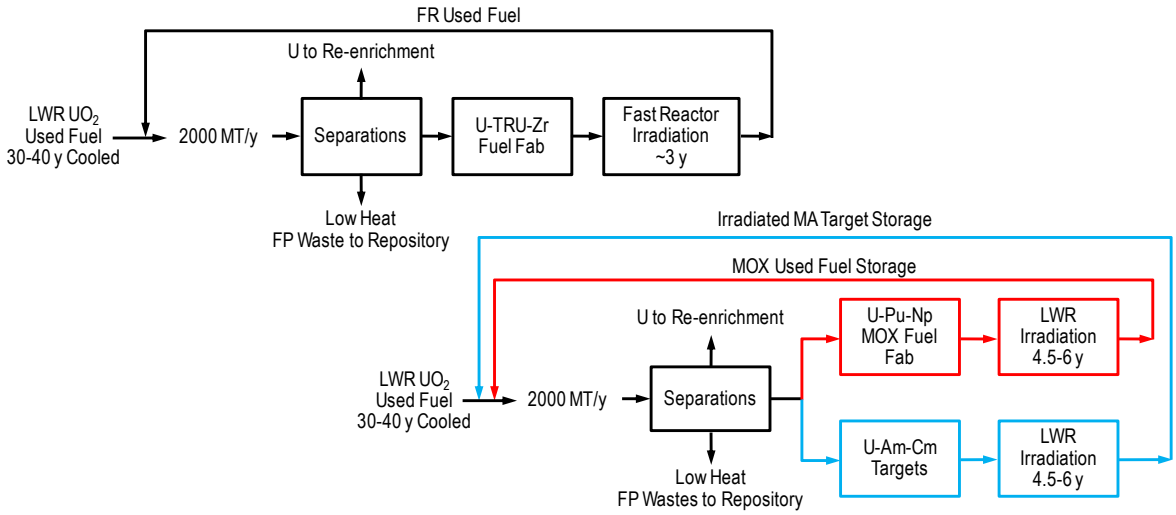
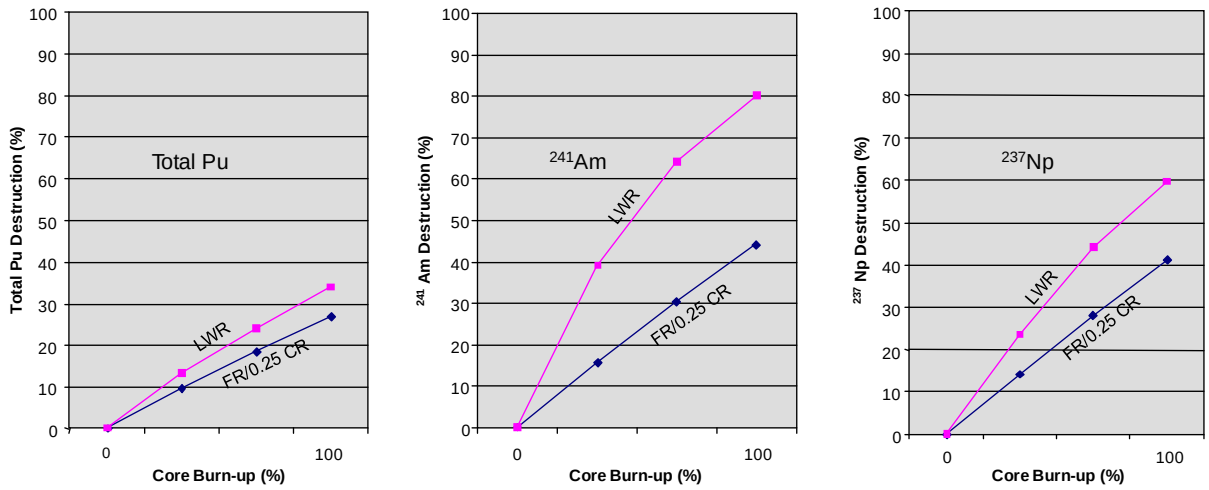


Figure 9: Burn-up of TRU actinides in one P-T cycle irradiation



The results of the modelling studies for actinide partitioning showed that for multi-cycle operation, the rate of production of the heavier actinides occurs to a greater extent in thermal spectrum irradiations. However, the rate of heavy actinide accumulation can be minimised by storing the used fuel for >30 years between P-T cycles. This allows the ^{241}Pu (half-life = 14.3 years) to decay predominantly to ^{241}Am and change the transmutation pathway from:

$$^{241}\text{Pu} \rightarrow {}^{242}\text{Pu} \rightarrow {}^{243}\text{Am} \rightarrow {}^{244m}\text{Am} \rightarrow {}^{244}\text{Cm}$$

to:

$$^{241}\text{Am} \rightarrow {}^{242}\text{Am} \begin{array}{l} \nearrow {}^{242}\text{Pu} \\ \searrow {}^{242}\text{Cm} \rightarrow {}^{238}\text{Pu} \rightarrow {}^{239}\text{Pu} \end{array}$$

83%

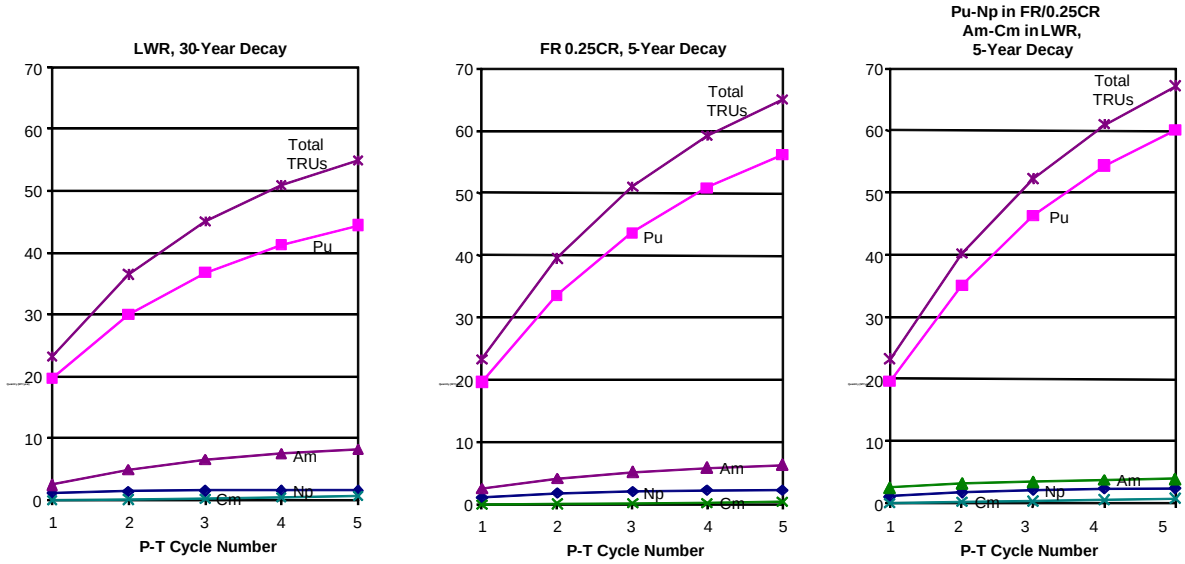
The latter pathway produces primarily lighter isotopes of plutonium (^{238}Pu, ^{239}Pu) rather than the heavy actinides (^{243}Am, ^{244}Cm, ^{252}Cf, etc.). Some (17%) branching of the decay of ^{242}Am (half-life = 16 hours) to ^{242}Pu occurs and allows a slow build-up of heavy elements, but even that will be mitigated by decay of ^{244}Cm (half life = 18 years) during the >35 year storage period between P-T cycles.

Comparative compositions of the actinide elements during five successive P-T cycles are shown in Figure 10 for recycling the actinides in LWR used fuel, beginning in each case with used fuel that had been irradiated for 45 GWd/MT and decayed for 30 years. The performance is shown for: *i)* recycling in LWR with a 30-year decay period of the used fuel between cycles; *ii)* recycling in fast reactors with a five-year decay period between cycles; *iii)* recycling of Pu-Np in fast reactors and Am-Cm in LWR with a five-year decay period between cycles. The studies concluded that, overall, there is not a great difference in actinide transmutation performance in the scenarios evaluated. The optimum performance was concluded to be to irradiate Pu-Np in fast breeder reactors when they are available or, until then, to utilise existing thermal spectrum reactors with a >30 year decay period between P-T cycles.

Summary

Lessons learned from experiences and research during the United States heavy element production programme show the difficulty and low yields of production of the heaviest transuranium element (TRU) actinides. These lessons were the basis for recent analyses of advanced fuel cycle partitioning-transmutation (P-T) of the TRU actinides produced in commercial used nuclear fuels. The recent analyses showed that previous interpretation of the average cross-section data and

Figure 10: Comparison of total TRU and element inventories in the total recycle from each cycle during multiple P-T cycles



the contention that fast spectrum transmutation is essential for consumption of the TRU actinides are not correct. The accurate conclusion is that both thermal and fast spectrum irradiation consume >99% of the TRU actinides and that consumption is not significantly different for multiple P-T cycles – even though the pathway for conversion of the actinides to fission products proceeds through different nuclides.

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