Some Calculations Regarding the Management of the UK's Plutonium Stocks

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1.1 Plutonium Waste Composition and Decay

Following irradiation within a thermal nuclear reactor, spent reactor fuel will typically comprise of the following isotopic fractions: 3.5% ^{238}Pu, 45% ^{239}Pu, 26% ^{240}Pu, 16% ^{241}Pu, 9% ^{242}Pu (variations in the isotopic composition can exist subject to reactor type and operating regime). Most of the plutonium isotopes have long half lives and decay slowly with time, in such circumstances the composition of a given isotope will change little over a storage period in the order of 40 years, however in the case of ^{241}Pu the decay half life is relatively short at just 14 years and over this period it will decay substantially. The decay mechanism is predominantly that of β decay and as such it will result in a significant conversion to ^{241}Am.

Table 1.0:
Decay data for isotopes of Z = 94

Nuclide	Decay Type	Half-life	Decay Constant s^{-1}
^{238}Pu	α 100%, SF 1.9×10^{-7}%, Si+Mg 2×10^{-14}%	87.7 y	2.51×10^{-10}
^{239}Pu	α 100%, SF 3.1×10^{-10}%	24110 y	9.11×10^{-13}
^{240}Pu	α 100%, SF 5.7×10^{-16}%	6561 y	3.35×10^{-12}
^{241}Pu	β 99.998%, α 2.45×10^{-13}%, SF 2.4×10^{-14}%	14.29 y	1.54×10^{-09}
^{242}Pu	α 100%, SF 5.5×10^{-4}%	3.75×10^{5} y	5.86×10^{-14}
^{241}Am	α 100%, SF 3.6×10^{-10}%	432.6 y	5.08×10^{-11}

Following a 40 year storage period the the 16% ^{241}Pu fraction will have decayed to just 2.4% whilst the ^{241}Am component will have increased from zero to 13.6%. The presence of this increased ^{241}Am content can lead to difficulties when used as MOX fuel in critical thermal reactors. The fission cross-section for ^{241}Am is much smaller than the capture cross-section and hence it will remove neutrons from the system whilst at the same time generating heavier actinides within the fuel. The fission and capture cross-sections for selected plutonium and americium nuclides are shown in Fig. 1.1.

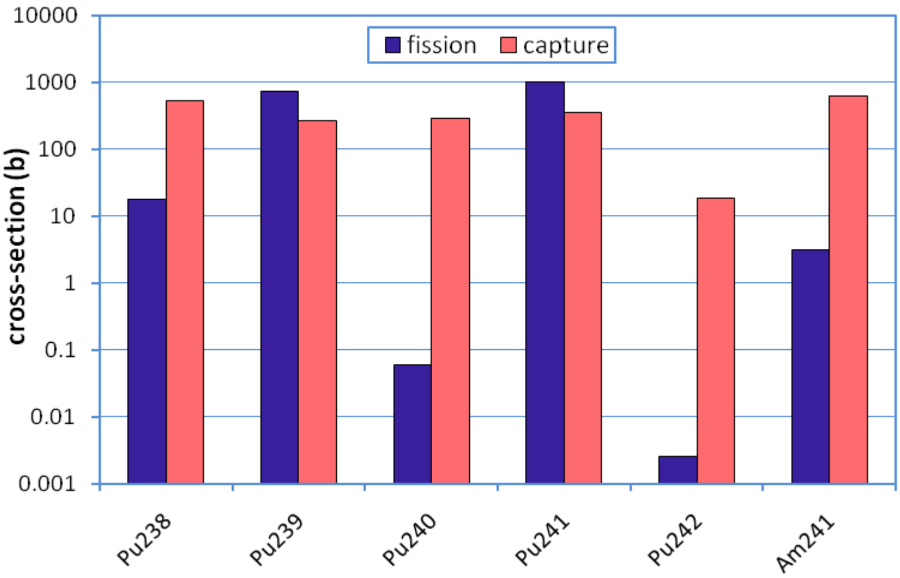

Fig. 1.1: One-group effective capture and fission cross-sections for selected nuclides in the thermal spectrum

The presence of the ^{241}Am comes at the expense of the very high fission cross-section which is present in ^{241}Pu, hence one of the side effects of storing plutonium is that the subsequent increase in its americium content results in a decrease in the reactivity of the plutonium. This effect can be seen in Fig. 1.2 which shows the available neutron surplus for both fresh and 40 year old plutonium enriched MOX PWR's. When the ratio of the neutrons produced from fission versus the number of neutrons absorbed falls below 1.0 insufficient neutrons are available to sustain a critical fission chain reaction. In all cases where the reactor neutron surplus falls below 1.0 critical reactor operation is not possible. The two profiles shown in Fig. 1.2 show that when fresh plutonium is used as the fissile enrichment, critical reactor operation is possible and a $43\,\mathrm{GWdt}^{-1}$ burn-up of the fuel is possible. However in the case of the plutonium which has been stored for 40 years, the conversion of ^{241}Pu to ^{241}Am has resulted in a significant and highly detrimental reduction in the available neutron surplus. Critical reactor operation with this fuel enrichment would not be possible, although the fuel mixture does achieve a brief neutron surplus between $15\,\mathrm{GWdt}^{-1}$ and $30\,\mathrm{GWdt}^{-1}$ the shortage of neutrons at the start of the burn-up would mean that this point is can never be reached without the addition of more neutrons through fissile enrichment or by means of an accelerator.

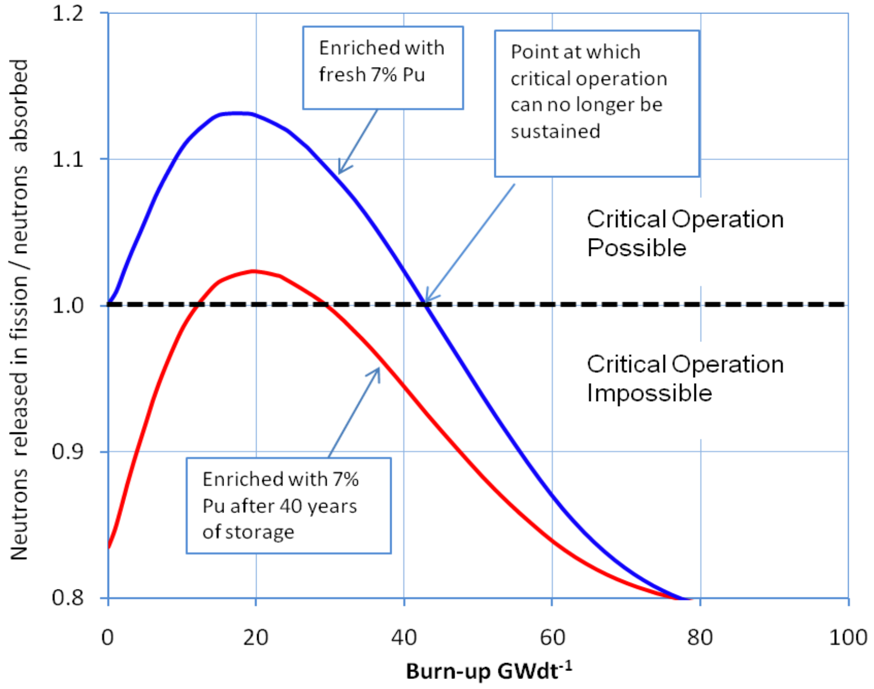

Fig. 1.2: Neutron economy versus burn-up for a 7% plutonium enriched MOX PWR using fresh plutonium and plutonium stored for 40 years

Clearly any proposal to use old plutonium stocks contaminated with americium will need to address the problems associated with the very poor neutron economy if such a proposal is to work.

1.2 Thorium Fuel

A possible alternative to the use of plutonium enriched ^{238}U MOX fuel is that of plutonium enriched thorium MOX fuel. One of the advantages of a thorium fuel platform in the thermal spectrum is its enhanced neutron surplus. This can be seen in Fig. 1.3 which shows the neutron surplus available for 100% ^{232}Th and 100% ^{238}U fuel sources. Although, due to the absence of an initial fissile enrichment, these fuel configurations cannot in practice sustain a fission chain reaction over the early part of the cycle, they do give a mathematical representation of the best position that can be achieved in the longer term.

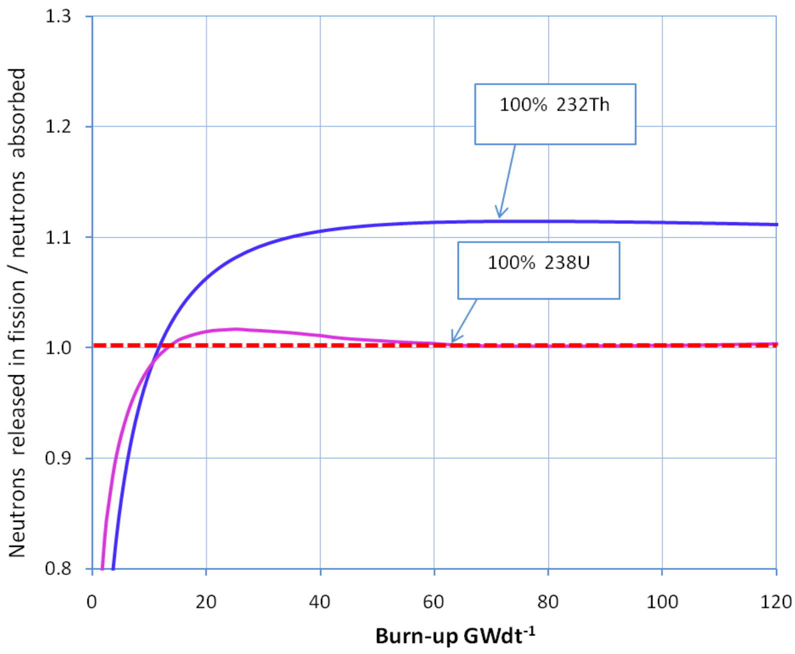

Fig. 1.3: Neutron economy ratio versus burn-up for 100% ^{232}Th and 100% ^{238}U fuel platforms in the thermal spectrum

Clearly the thorium fuel platform produces a significantly higher neutron surplus than the ^{238}U fuel source. By enriching a thorium fuel platform with plutonium, the shortfall in neutrons in the early part of the burn-up can be compensated for allowing a critical fission reaction to be established with fresh plutonium enrichment. Fig. 1.4 shows the neutron economy for a thorium fuelled PWR enriched to 7% with both fresh and 40 year old plutonium. Over the early part of the burn-up good neutron surplus exists for the fresh plutonium enrichment however when 40 year old plutonium is adopted the neutron surplus falls significantly below 1.0 and is unable to sustain critical operation. For both of the plutonium enrichments shown the neutron surplus produced with the thorium fuel is better than that from ^{238}U fuel over the early stages of the burn-up however over the $10\,\mathrm{GWdt}^{-1}$ to $50\,\mathrm{GWdt}^{-1}$ range a superior neutron economy is present with the ^{238}U fuel source. For reactor operation to be established the system will need the capacity to overcome all of the shortfalls in the neutron population over the intended burn-up range. In this respect the aged plutonium enrichment with ^{238}U fuel produces the biggest maximum shortfall in neutrons and hence it will require the largest fissile enrichment or accelerator installation to enable operation to take place. In this respect the thorium fuel platform has an advantage, however where fresh plutonium is used as the fissile enrichment both the thorium and plutonium fuel platforms are capable of sustaining critical operation.

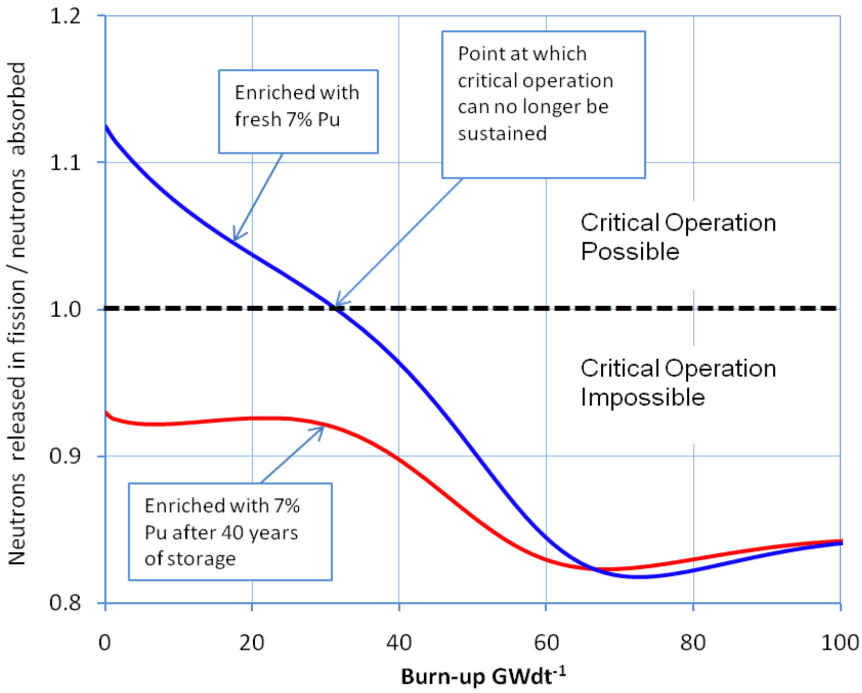

Fig. 1.4: Neutron economy versus burn-up for a thorium fuelled, 7% plutonium enriched MOX PWR using fresh plutonium and plutonium stored for 40 years

1.3 Nuclide Equilibrium

The presence of the higher ^{241}Am population within the initial fuel load generates short term transient equilibrium positions for the actinides within the reactor. This can be seen in Fig. 1.5 which shows the relative atom populations for selected curium and americium isotopes which are generated using aged and fresh plutonium enrichments.

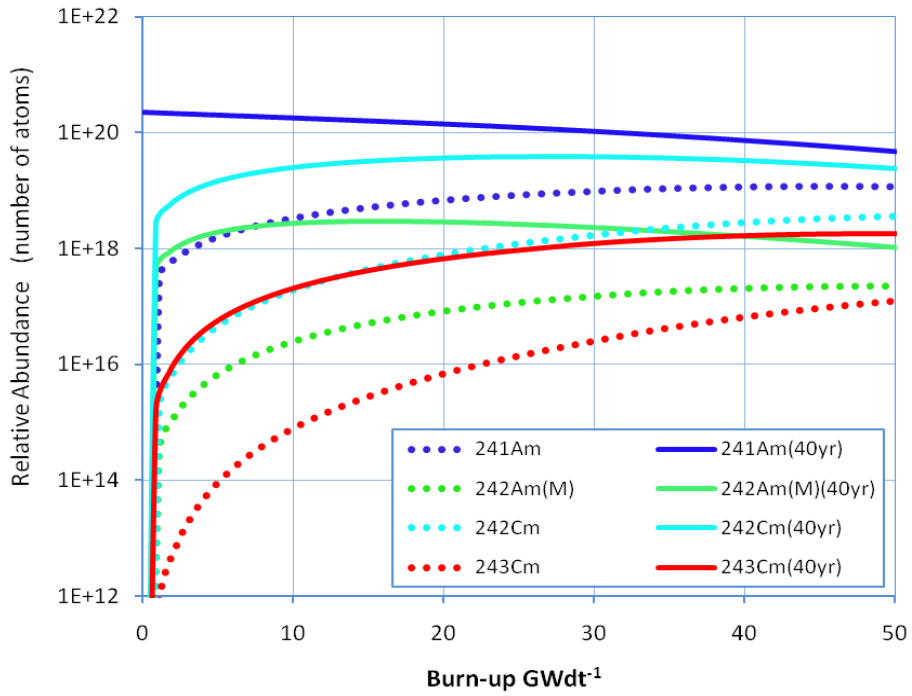

Fig. 1.5: Relative atom populations for a 7% plutonium enriched MOX PWR using fresh plutonium and plutonium stored for 40 years

The presence of the higher ^{241}Am atom population clearly affects the population of other actinides within the reactor and results in the generation of higher ^{242}Am(M), ^{242}Cm and ^{243}Cm populations over the short term. If reactor operation was to continue over an extended period with repeated recycling of the actinides but with no further enrichment, identical actinide populations would result regardless of the initial fuel enrichment. The significance of the presence of the elevated ^{241}Am population in this respect is in the short term early recycling scenarios or in a situation where once through reactor fuel cycle is employed. In this respect the influence of the ^{241}Am population is significant in its effect on the plutonium composition following a single batch operation. The evolution of the plutonium isotopes with the reactors is shown in Fig. 1.6 and 1.7 for fresh and aged plutonium compositions. Whilst most of the plutonium isotopes are largely unaffected by the presence of the ^{241}Am population, its effect on ^{238}Pu is significant, resulting in a 300% increase in its population after a $50\,\mathrm{GWdt}^{-1}$ burn-up. This is due to the elevated ^{242}Cm population which generates ^{238}Pu through it's α decay mechanism with a half life of 235 days.

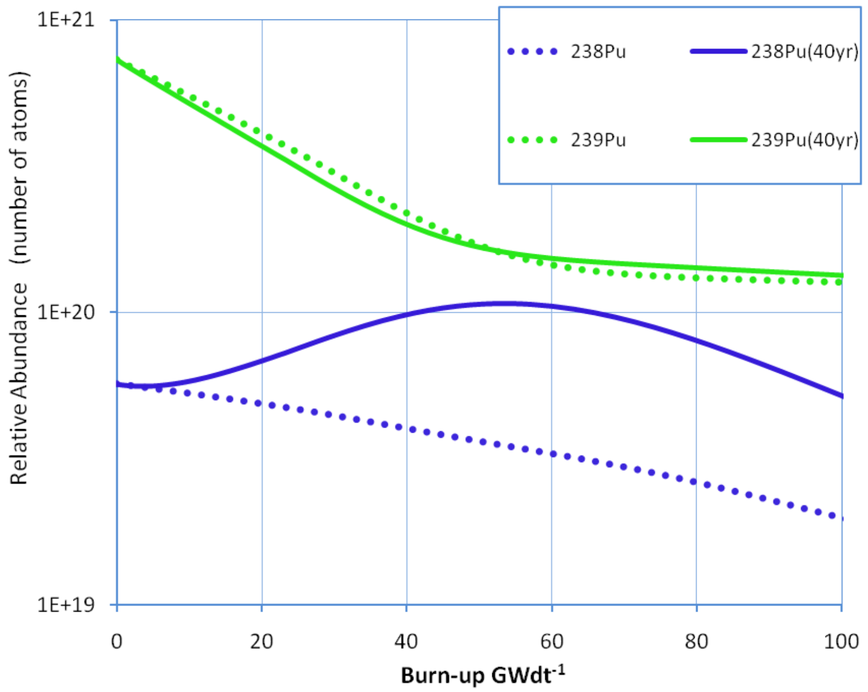

Fig. 1.6: Relative atom populations for ^{238}Pu and ^{239}Pu using a 7% plutonium enriched MOX PWR for fresh plutonium and plutonium stored for 40 years

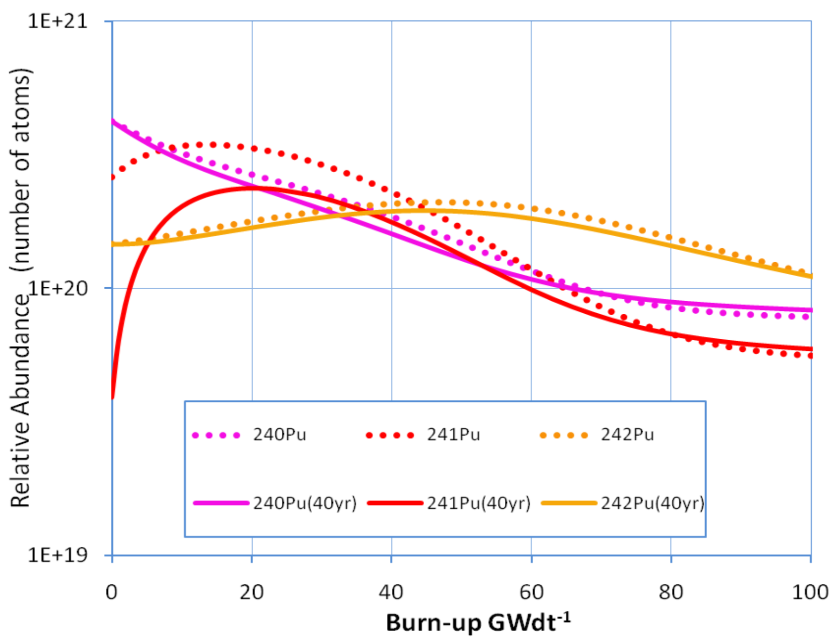

Fig. 1.6: Relative atom populations for ^{240}Pu, ^{241}Pu and ^{242}Pu using a 7% plutonium enriched MOX PWR for fresh plutonium and plutonium stored for 40 years

1.4 Thorium Equilibrium

Where a thorium fuel platform is employed a more rapid burn-up of the plutonium enrichment can be achieved. Furthermore the thorium fuel platform enables the plutonium enrichment to be burnt to a much lower atom population within the reactor. This can be seen in Fig. 1.7 which shows the change in atom populations for the plutonium isotopes within a 7% enriched reactor using both thorium and ^{238}U fuel platforms.

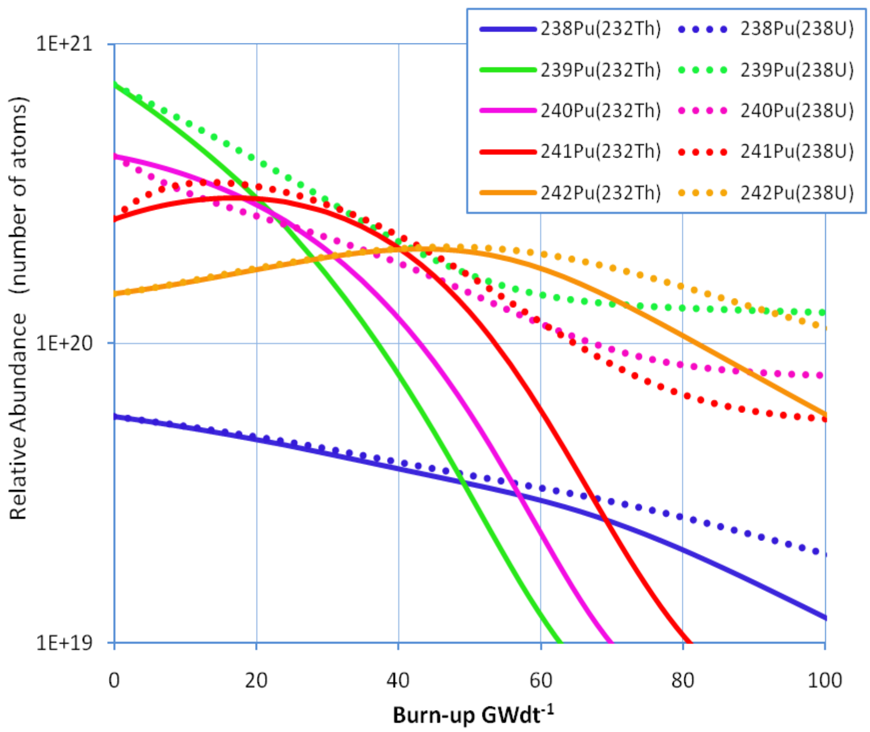

Fig. 1.7: Relative atom populations for plutonium isotopes in 7% plutonium enriched PWR using thorium and ^{238}U fuel platforms

At a $40\,\text{GWdt}^{-1}$ burn-up the atom populations for ^{239}Pu and ^{240}Pu using thorium fuel are just 37% and 66% respectively compared with that for ^{238}U fuel. Although the populations for the other plutonium isotopes ^{238}Pu, ^{241}Pu and ^{242}Pu show little difference at this burn-up, significant divergence does occur with these isotopes at higher burn-ups with thorium producing the lower atom populations. This effect results from the lower steady-state equilibrium positions that are generated using thorium fuel, if the reactor is operated for a sufficient duration all nuclides will adjust in relative atom populations towards the equilibrium positions. In a thermal reactor the minimum relative atom populations that can be achieved through irradiation in enriched thorium and ^{238}U fuelled reactors are dictated largely (although not entirely) by the atom populations generated for the 100% pure fuel forms for these systems. This can be seen in Fig. 1.8 which shows the relative atom populations for pure and enriched fuel sources for plutonium isotopes with a ^{238}U fuel platform and for an enriched thorium system.

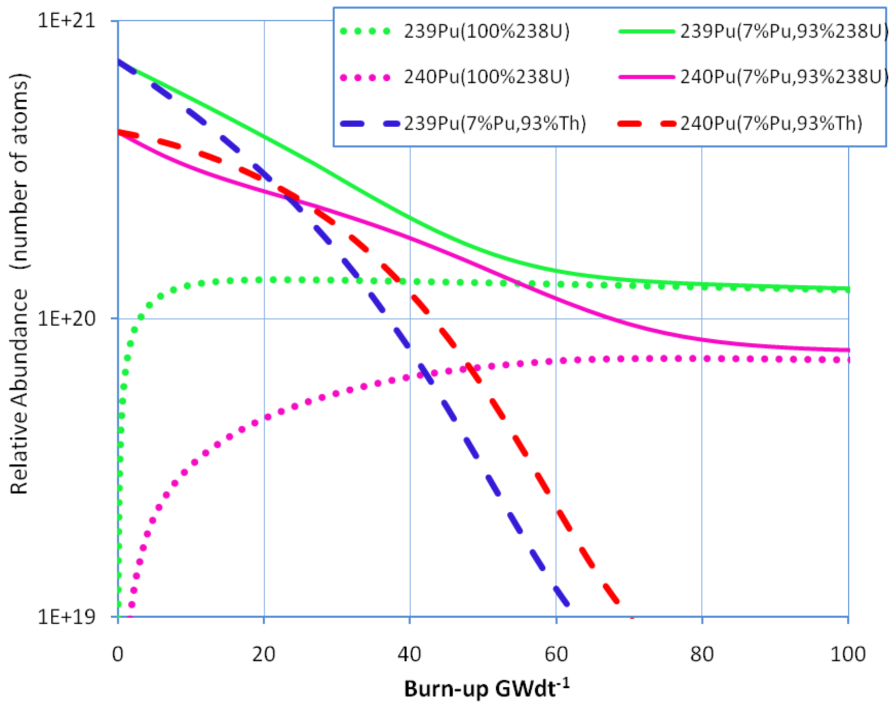

Fig. 1.8: Relative atom populations for plutonium isotopes in 7% plutonium enriched, pure ^{238}U and enriched thorium PWR's

Fig. 1.8 shows how the plutonium populations for the enriched system converge towards the 100% ^{238}U profiles, the 100% profiles largely represent the minimum populations that can be achieved through irradiation. The higher equilibrium values for the ^{238}U fuel limit the rate and extent of the plutonium burning. The much lower equilibrium positions which exist in a thorium system (not shown on this scale) enable a faster and greater level of plutonium destruction.

1.5 Fast Reactors

Reactors operating in the fast spectrum benefit from an excellent neutron surplus due to the beneficial fission to capture ratios which exists for the significant nuclides compared with that found in the thermal spectrum. This can be seen in Fig .1.9 which shows the fission fraction of the absorption cross-section for selected nuclides, with some notable exceptions, in the fast spectrum there is a greater likelihood of fission taking place for the majority of the nuclides. The very high neutron surplus enables critical operation to be established using plutonium enriched reactors with ^{238}U and thorium fuel platforms regardless of plutonium storage time.

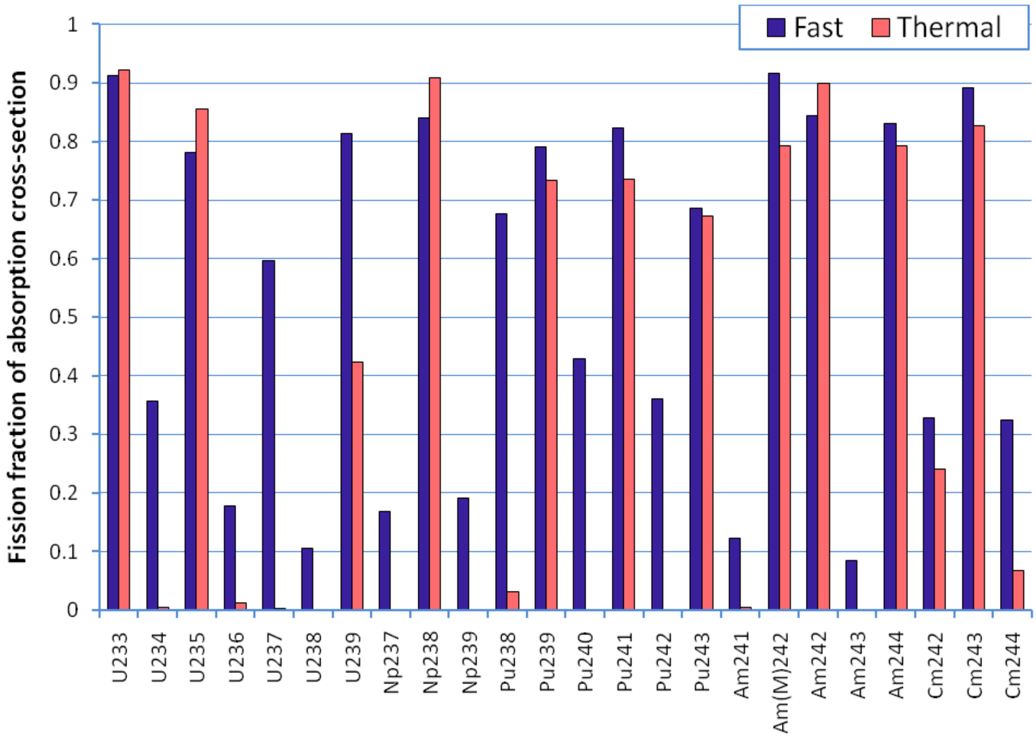

Fig. 1.9: Probability of fission as a fraction of the total neutron absorption cross-section for selected nuclides

Despite the clear advantage that fast reactors have over thermal systems in terms of enhanced fission cross-sections, thermal reactors do have an advantage in being able to achieve a more rapid destruction of the plutonium enrichment.

1.6 Conclusion

Plutonium which has been stored for just a relatively short period of time (tens of years) will become denatured as ^{241}Pu decays into ^{241}Am. The presence of elevated levels of ^{241}Am within the plutonium results in a significant deterioration in its fissile properties. If used as MOX fuel in thermal reactors this effect will need to be accounted for by some means if critical reactor operation is to be established.

The use of thorium fuel (in thermal reactors) provides an enhanced neutron surplus which can compensate to a limited extent for the effects of the americium contamination, however the effect is limited and critical reactor operation would not be possible with even modest levels of contamination.

Accelerator driven systems will enable reactor operation to be established for all reactor fuel mixtures, however less fissile fuels would generate less reactor power for a given fuel mass and accelerator size. In this respect the enhanced neutron surplus achieved by using thorium fuel in a thermal reactor would deliver the best return in power for a given fuel mass and accelerator size.

The low nuclide equilibrium positions which are generated from thorium enable a faster and greater reduction in the populations of heavy actinides such as plutonium compared with that in a ^{238}U fuelled reactor. This feature could be exploited to deliver a greater reduction in the plutonium stockpile through operation or repeated recycling of the actinides.

Fast reactors using thorium or ^{238}U fuel platforms are capable of establishing critical operation with plutonium enrichments of any age.