



# Proliferation Relevance and Safeguards Implications of Partitioning and Transmutation Nuclear Fuel Cycles

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## ABSTRACT

Over the last 2 decades there has been renewed interest in developing advanced nuclear reactors and fuel cycles. Many of these advanced design concepts require or can use fuel elements that contain actinides recycled from light water reactor spent fuel. Irradiation of these elements in fast nuclear reactors is supposed to transmute them into less toxic isotopes and reduce their mass. Since transmutation is not efficient, recycling into new advanced reactor fuel must occur repeatedly to achieve a substantial reduction in mass. The introduction of this technology will create long-term proliferation risks and require safeguarding not only of plutonium, but also of the other target actinides: neptunium, americium, and curium. These elements will be present in isotopic mixtures for which information on their critical masses is unavailable. This paper provides data on critical masses and spontaneous fission neutron background for the isotopic compositions of the actinides of interest and their evolution in light water reactor fuel of various burnups and during spent fuel storage. These data are complemented by generic estimates of total inventories of these elements present in full-scale partitioning and transmutation fuel cycles being considered for commercial scale and of the time periods required for significantly reducing their proliferation potential.

## ARTICLE HISTORY

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## Introduction

In 2000, the Generation IV International Forum was created to coordinate research and development of an advanced generation of nuclear energy systems.<sup>1</sup> The Generation IV International Forum selected six reactor concepts (Generation IV reactors) for its consideration, all of which have the potential to incorporate nuclear fuel cycles that partition plutonium and minor actinides (neptunium, americium, and curium) from spent fuel, followed by transmutation.<sup>2</sup> These advanced fuel cycles include multi-step separation, fuel fabrication, and irradiation of considerable masses of fissile

actinides and will therefore require dedicated safeguards. Partitioning and transmutation of these actinides is of interest because the processes potentially reduce the toxicity and mass of the high-active wastes, as well as reduce the perceived period of greatest risk for geologic repositories to a few centuries.<sup>3</sup> The latter benefit may be of limited value because long term safety assessments show that plutonium and other actinides are virtually immobile in the geosphere and biosphere and are at low risk of significantly contributing to potential radiation exposures in the future.<sup>4</sup>

Generation IV reactor research largely focuses on developing an advanced sodium-cooled fast reactor. Sodium cooled fast reactor designs have the most operational experience of the six reactor concepts selected by the Forum and include some commercial reactors.<sup>5</sup> Sodium cooled fast reactors are promising for the transmutation of actinides, since isotopes of these elements generally show higher elimination to build-up ratios in fast rather than in thermalized neutron spectra. However, sodium-cooled fast reactors are characterized by a positive reactivity coefficient which could cause a power excursion with high energy release and core destruction, as experienced in the Chernobyl accident in 1986.<sup>6</sup> Additionally, partitioning and transmutation fuel cycles have increased proliferation risk for two major reasons. First, a homogeneous core of actinide-bearing fuel rods increases the inherent positive reactivity coefficient for these reactors. The solution to this problem is to heterogeneously position the minor actinides in the outer core region only. This requires the separation of plutonium from the minor actinides and prevents the use of a more proliferation resistant reprocessing strategy of extracting a mixture of plutonium and actinides, if spiked with lanthanides.<sup>7</sup> Second, the use of uranium as the fuel matrix is mandatory because of the large negative Doppler reactivity coefficient of uranium-238. Its production of plutonium, however, limits the net transmutation rate per irradiation cycle, thus creating the need of multiple recycling to effectively reduce the mass of plutonium and the minor actinides. Consequently, inventories of these elements will remain in circulation for long periods at the various stages of the fuel cycle. These constraints significantly affect the proliferation sensitivity of the partition and transmutation concept.

Whereas some data are available on critical masses of pure transuranium isotopes, they are unavailable for the isotopic mixtures of those actinides intended for transmutation when current generation nuclear spent fuel is reprocessed.<sup>8</sup> These data will be provided in the following analysis, together with some generic estimates of their total masses that need to be safeguarded in the event a full-scale partitioning and transmutation fuel cycle is realized.

## Materials and methods

### *The partitioning and transmutation scenario*

It is generally expected that the actinides to be partitioned and transmuted are generated by commercial light water reactors. When this technology will become available on an industrial scale is uncertain. The isotopic composition of the spent fuel at the time of partitioning is a function of the storage period. Therefore, several different interim storage periods for irradiated fuel prior to partitioning were assumed, resulting in quite different isotopic compositions depending on the length of storage. It is assumed here that partitioning will be achieved by aqueous reprocessing with separation of uranium, plutonium, neptunium, americium, and curium.<sup>9</sup> Two reprocessing options are considered. Under option one the minor actinides americium and curium are co-extracted, since their separation is difficult. In the other, if recycling of the curium isotopes is not intended, americium is separated, avoiding the excessive shielding requirements caused by the high spontaneous fission rate of curium-244.<sup>10</sup>

Recent fuel cycle simulations have calculated the transmutation efficiencies of a 1200 MWt sodium-cooled fast reactor, which is optimized for plutonium and minor actinide transmutation, respectively.<sup>11</sup> These data are used in the following analysis, modified to reflect a much longer storage period. Previous simulations assumed five cycles of irradiation, each of 365 equivalent full power days, followed by two years of storage and another year for reprocessing and fuel fabrication. However, such a short out-of-core period is unrealistic. The introduction of high burnup fuel in light water reactors has already increased decay times, which are required before fuel reprocessing to limit radiolytic degradation of the solvents used by the PUREX process. In this study, therefore, seven years was assumed for interim storage of the spent fuel resulting in an out-of-core period of 15 years between successive irradiations of the recycled actinides.<sup>12</sup> The correction of the cycle length may slightly modify the transmutation efficiencies per partitioning and transmutation cycle calculated by Vezzoni et al. due to the radioactive decay and build-up processes<sup>13</sup>; however, this effect has not been quantified in this analysis. This generic scenario permits the assessment of the relative proliferation risk of partitioning and transmutation technology for both the phasing out or the continuation of commercial nuclear power programs.

### *Neutronic simulations*

To estimate the masses of actinide elements entering an industrial partitioning and transmutation fuel cycle and their proliferation risk,

information on their formation during burnup of the fuel in a power reactor and on their decay (or build-up) during interim storage is required. These estimates were calculated using the SCALE, version 6.1, code system and their critical masses, accounting for their isotopic compositions, which are dependent on their irradiation and storage history.<sup>14</sup>

### *Irradiation in light water reactors*

Formation of the fissile elements in power reactors was simulated for high burnup uranium fuel with Gd<sub>2</sub>O<sub>3</sub> burnable poison irradiated in a pressurized water reactor. Specifications of the fuel element and operating conditions are given in Table 1. The radially heterogeneous fuel loading/burnup pattern of light water power reactors was considered generically, since this modifies the build-up of americium and curium isotopes compared to the more commonly used simplified assumption of a homogeneous core.<sup>15</sup> Simulations were performed using the TRITON code of SCALE,<sup>16</sup> which allows for a two-dimensional representation of the fuel assembly's geometry, and its ENDF/B-VII nuclear data libraries.<sup>17</sup>

### *Interim storage*

Storage times up to 50 years were considered, since it seems to be reasonable that it may be 50 years until a full commercial scale partitioning and

**Table 1.** Fuel element data and irradiation history used for simulating fissile element formation in a pressurized water reactor.

|  |                 |
|--|-----------------|
| <i>Fuel rod data</i>                                     |                 |
| Fuel material  | UO <sub>2</sub> |
| Fuel density (g cm <sup>-3</sup> )                       | 10.4            |
| Initial <sup>235</sup> U enrichment (wt.-%) <sup>a</sup> | 4.0 (2.6)       |
| Fuel pellet diameter (cm)                                | 0.805           |
| Clad inner diameter (cm)                                 | 0.822           |
| Clad outer diameter (cm)                                 | 0.95            |
| Clad material  | Zircaloy-4      |
| Initial Gd <sub>2</sub> O <sub>3</sub> content (wt.-%)   | 7.0             |
| <i>Guide tube data</i>                                   |                 |
| Guide tube inner diameter (cm)                           | 1.11            |
| Guide tube outer diameter (cm)                           | 1.232           |
| Guide tube material                                      | Zircaloy-4      |
| <i>Fuel assembly data</i>                                |                 |
| Lattice geometry   | 18 × 18         |
| Rod pitch (cm)   | 1.27            |
| Number of UO <sub>2</sub> rods                           | 288             |
| Number of rods with Gd <sub>2</sub> O <sub>3</sub>       | 12              |
| Number of guide tubes                                    | 24              |
| <i>Operating conditions</i>                              |                 |
| Power density (kW kg <sup>-1</sup> )                     | 37.5            |
| Full power days per cycle                                | 444.3           |
| Shut down time per cycle (days)                          | 30              |
| Number of cycles per assembly                            | 3               |
| Moderator density (g cm <sup>-3</sup> )                  | 0.711           |
| Mean soluble boron concentration (ppm)                   | 750             |

<sup>a</sup>Value given in brackets refers to gadolinium-bearing fuel rods.

transmutation fuel cycle is available. Modifications of masses and isotopic compositions of the transuranium elements during such an extended period of interim storage of the irradiated fuel were calculated as part of the burnup simulations with the TRITON code.

### ***Critical masses***

Critical masses were calculated for bare solid metal spheres,<sup>18</sup> using the Monte Carlo code KENO V.a in combination with the CSAS5 control module of SCALE.<sup>19</sup> This allows for an iterative search of a configuration with  $k_{eff}=1.0$  by systematic variation of the radius of the sphere. The 238 energy group ENDF/B-VI.8 nuclear data library available in SCALE was used. For plutonium metal, its  $\alpha$  phase has been assumed. For comparison, some additional simulations were performed assuming a beryllium reflector surrounding the fissile metal sphere.

### ***Spontaneous fission rates***

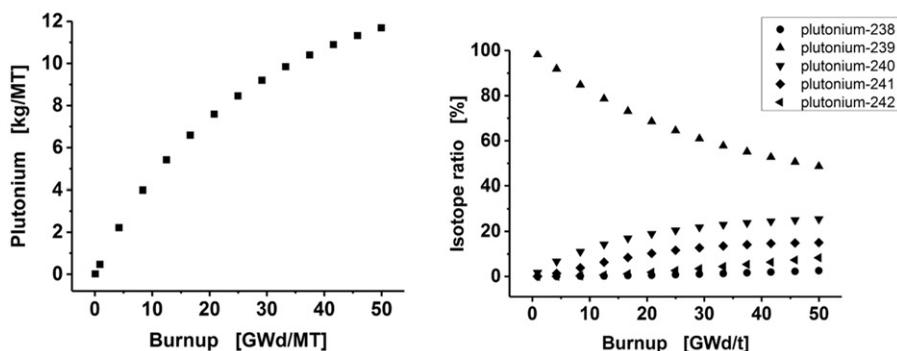
Since the neutron flux density generated by spontaneous fission events has a major impact both on the technical effort required to manufacture a high yield nuclear explosive device and on the measures needed for limiting radiation exposure during handling, this quantity has been determined for the calculated bare critical masses using the spontaneous fission probabilities and numbers of neutrons per fission given in the ENDF/B-VII nuclear data libraries of SCALE.<sup>20</sup>

## **Results and discussion**

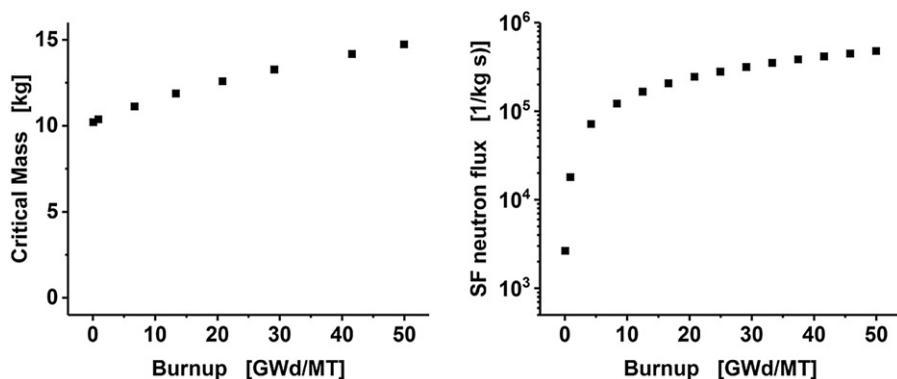
### ***Plutonium***

#### ***Fuel irradiation***

The formation of plutonium with increasing burnup and its isotopic composition in a pressurized water reactor are shown in [Figure 1](#). At high burnups its mass asymptotically approaches saturation, since with depleting uranium-235 the share of power produced by fission of plutonium increases. Its isotopic composition is characterized by a monotone decreasing fraction of plutonium-239. In contrast, the plutonium-240 almost reaches equilibrium at about 30 GWd/MT burnup due to its high near-thermal neutron absorption cross section. This temporal development of the isotopic composition is reflected by their critical masses and spontaneous fission source strengths given in [Figure 2](#). The critical mass of a solid metal sphere only slightly increases with burnup, whereas its spontaneous fission neutron flux, which is mainly produced by plutonium-240, steeply increases at low burnup, but attains almost constant levels at high fuel burnups. These results expand previous assessments that a nuclear



**Figure 1.** Mass of plutonium produced in pressurized water reactor fuel during irradiation (left) and its isotopic composition (right).



**Figure 2.** Minimum critical mass (left) and spontaneous neutron fission flux (right) of plutonium produced in pressurized water reactor fuel.

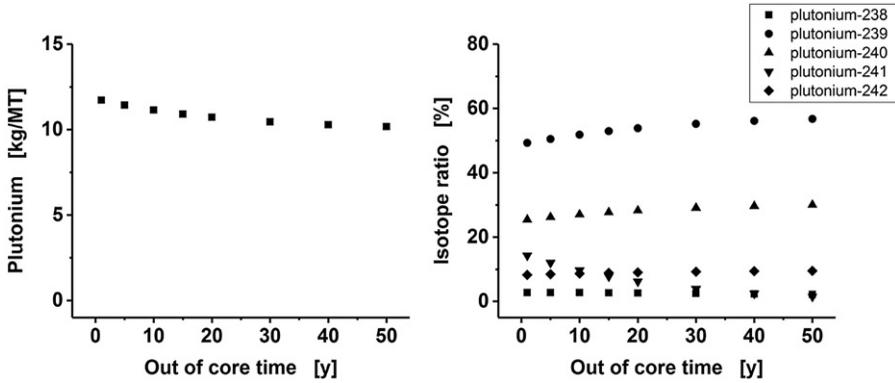
explosive device may be produced using plutonium originating from high burnup fuel of commercial power reactors.<sup>21</sup>

### Interim storage

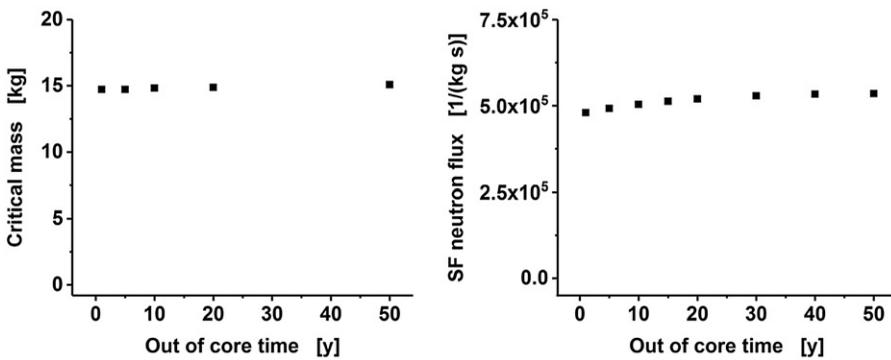
The decay of plutonium-241 with its 14.4-year half-life has a marked effect on the isotopic composition of the plutonium during extended storage of the irradiated fuel and will reduce its mass by up to 13% within 50 years (Figure 3). Its effect on the critical mass of a bare solid metal sphere and on the spontaneous fission neutron flux remains marginal (Figure 4). Alpha decay of the curium isotopes does not significantly contribute to the plutonium inventory due to their low mass ratio even in high burnup fuel (see Figure 10).

### Neptunium

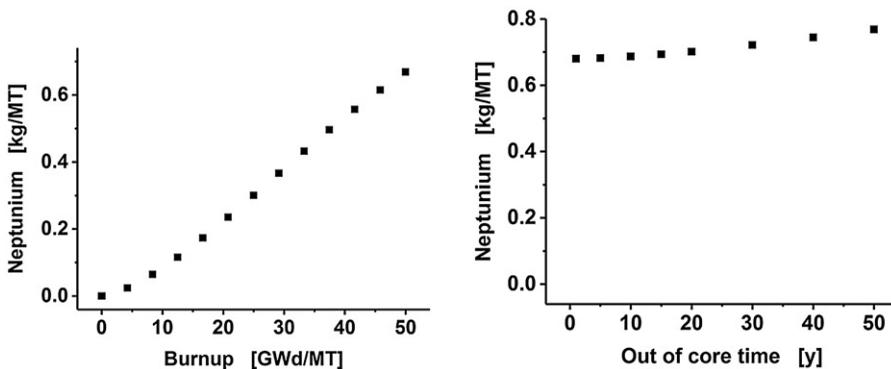
The only relevant isotope is neptunium-237 with its  $2.1 \times 10^6$ -year half-life. During irradiation its formation rate increases with burnup (Figure 5, left).



**Figure 3.** Effect of interim storage on the plutonium mass present in the irradiated fuel (left) and its isotopic composition (right).



**Figure 4.** Modification of critical mass (left) and spontaneous fission neutron flux (right) of plutonium present in the irradiated fuel during interim storage.



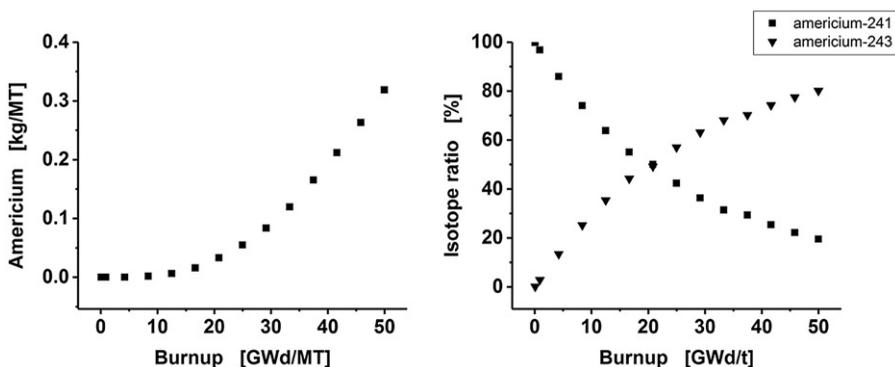
**Figure 5.** Mass of neptunium present in pressurized water reactor fuel during irradiation (left) and interim storage of the spent fuel (right).

In stored fuel, more neptunium is produced by alpha decay of americium-241 (Figure 5, right). Neptunium's isotopic composition does not change over time. The critical mass of a bare solid neptunium metal sphere therefore remains constant, at a calculated value of 65.0 kg. Its spontaneous fission source strength is marginal with  $0.11\ \text{neutrons}\ \text{kg}^{-1}\cdot\text{s}^{-1}$ .

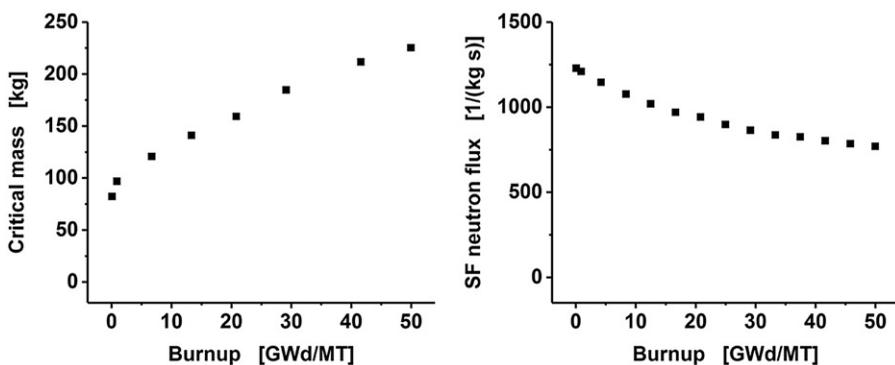
## Americium

### Fuel irradiation

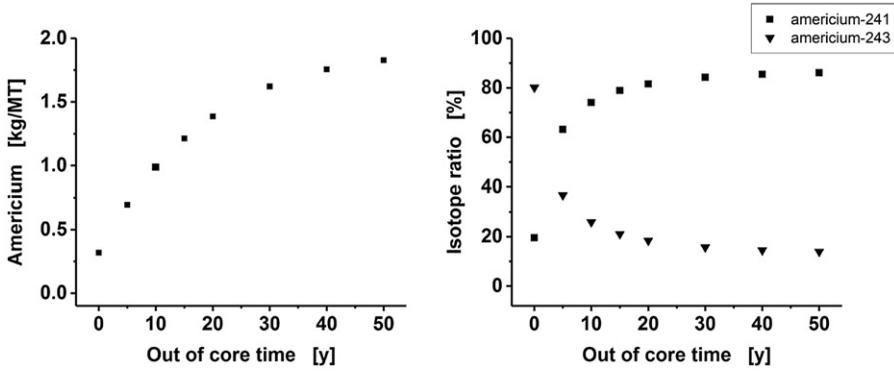
Compared to plutonium, the mass of americium generated in a light water reactor remains low with most of it being produced at high burnup (Figure 6, left). Its isotopic composition is dominated by americium-241 at low burnup, but gradually shifts to americium-243 during irradiation (Figure 6, right). The isotopes americium-242m, americium-242 and americium-244 do not accumulate in the fuel due to their high fission cross sections at thermal neutron energies. The evolution of the isotopic composition of the americium is reflected by the increase of its mass of a critical bare solid metal sphere with burnup (Figure 7, left), which results from the lower fission cross section of americium-243 than of americium-241. The masses calculated correspond to metal spheres with radii between 13.5 cm at 10 GWd/MT burnup and 16.5 cm at fuel unloading. Since spontaneous fission rates of both americium isotopes are low, the neutron fluxes given in Figure 7 (right) are considerably smaller than those of weapons-grade plutonium ( $5.18 \times 10^4$  neutrons  $\text{kg}^{-1} \cdot \text{s}^{-1}$  at 5% plutonium-240).



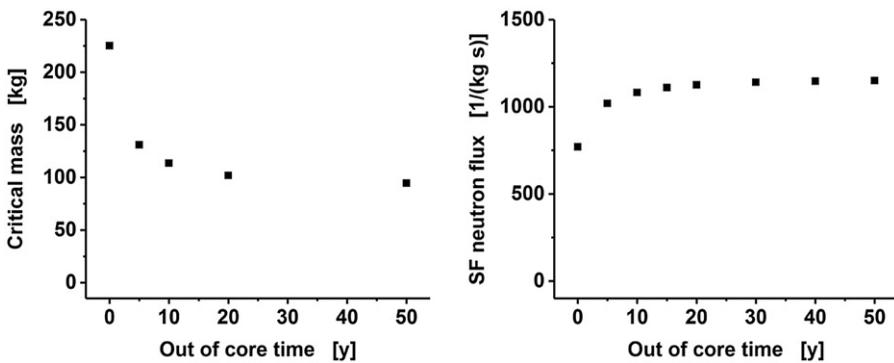
**Figure 6.** Mass of americium generated during irradiation of pressurized water reactor fuel (left) and its isotopic composition (right).



**Figure 7.** Critical mass of a bare metal sphere (left) and spontaneous neutron fission flux (right) of americium produced in pressurized water reactor fuel.



**Figure 8.** Effect of interim storage on the mass of americium present in the irradiated fuel (left) and its isotopic composition (right).



**Figure 9.** Variation of critical mass (left) and spontaneous fission neutron flux (right) of americium present in the spent fuel during interim storage.

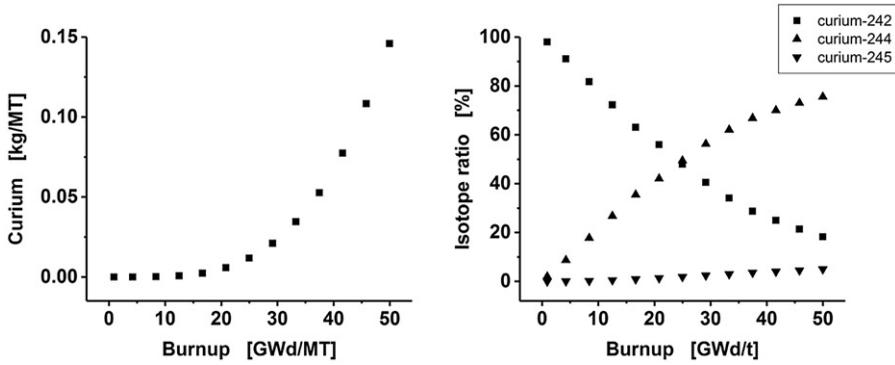
### *Interim storage*

Although both americium-241 and americium-243 have long half-lives of 433 and 7,370 years, respectively, even moderate storage periods strongly increase the mass of americium present in the spent fuel (Figure 8, left). This effect results from the beta decay of plutonium-241 (14.4-year half-life) into americium-241. This ingrowth shifts the isotope ratio (Figure 8, right) and decreases the critical mass of a bare metal sphere (Figure 9, left) to values like those of americium in low burnup fuel. The production of americium-241 during storage is also reflected by a slight increase of the spontaneous fission neutron flux of americium with time (Figure 9, right), which is caused by twice the SF neutron source strength of this isotope compared to americium-243.

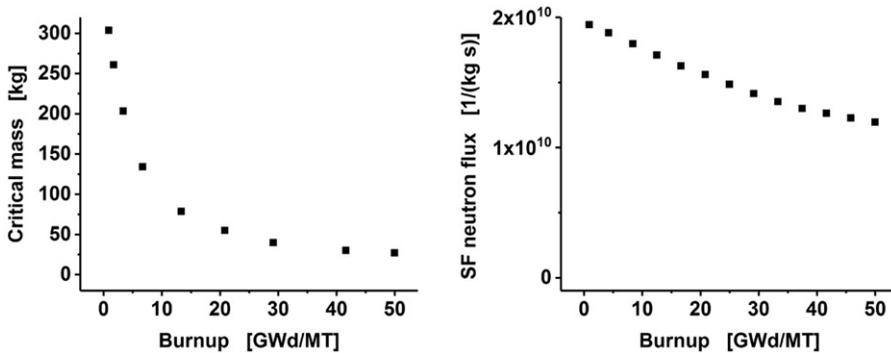
### *Curium*

#### *Burnup*

As the curium isotopes are produced by neutron absorption in americium followed by beta decay, their formation requires long irradiation times of



**Figure 10.** Mass of curium generated during irradiation of pressurized water reactor fuel (left) and its isotopic composition (right).

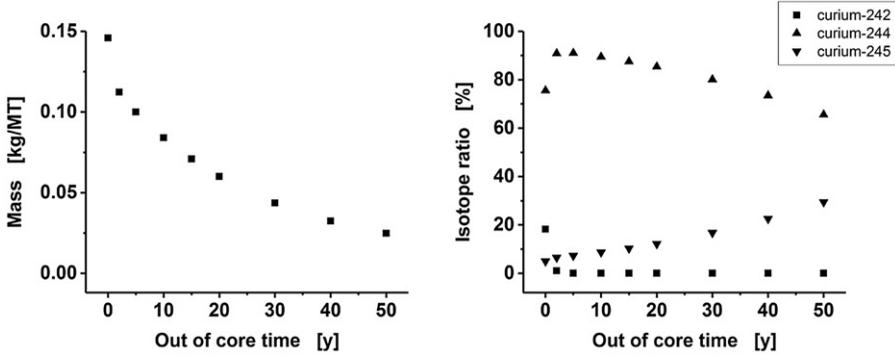


**Figure 11.** Critical mass of a bare metal sphere (left) and spontaneous neutron fission flux (right) of curium produced during burnup of pressurized water reactor fuel.

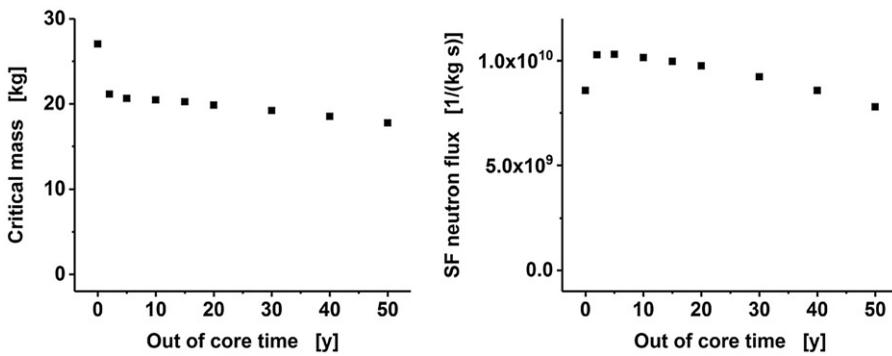
the fuel, but then proceeds exponentially, although masses remain comparatively small (Figure 10, left). First, curium-242 prevails, but with increasing burnup curium-244 dominates its isotopic composition (Figure 10, right). The variation of the isotopic composition is reflected by the critical masses of a bare sphere of curium metal, which, due to the formation of curium-244, decreases by an order of magnitude during burnup, reaching a value of only 27 kg at fuel discharge (Figure 11, left). However, the attractiveness of curium for a nuclear explosive device is severely hampered by its excessively high neutron flux (Figure 11, right), which originates primarily from spontaneous fission of the even-numbered curium isotopes.

### *Interim storage*

Due to the rapid disintegration of the major curium isotopes with half-lives of 163 days (curium-242) and 18.1 years (curium-244), the mass of curium present in spent fuel decreases by a factor of 10 within 50 years



**Figure 12.** Effect of interim storage on the curium mass present in the irradiated fuel (left) and its isotopic composition (right).

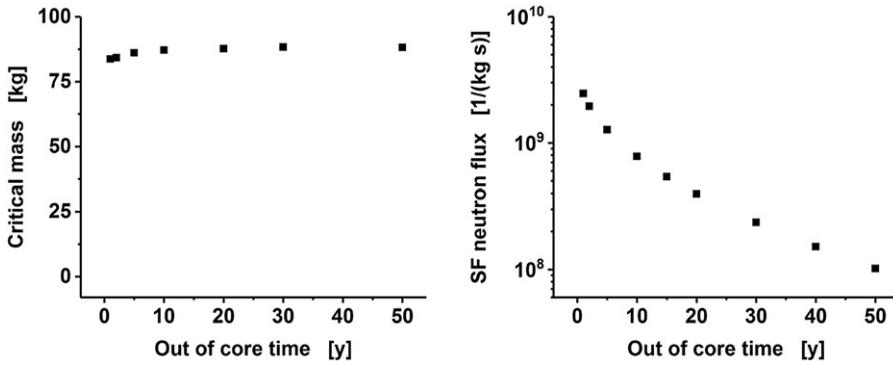


**Figure 13.** Variation of critical mass (left) and spontaneous fission neutron flux (right) of curium present in the spent fuel during interim storage.

after removal from the reactor core (Figure 12). Whereas the critical mass of a bare sphere of curium drops by about 25%, its excessive spontaneous fission neutron source strength varies only slightly during storage (Figure 13).

### **Americium plus curium**

If the option of avoiding the chemically difficult stripping of curium from americium after co-extracting both trivalent actinides from the waste stream is taken, a mixture of americium and curium must be dealt with within a partitioning and transmutation fuel cycle. Its mass and isotopic composition as a function of fuel burnup and time after unloading the spent fuel from the reactor are easily deduced from the data given above for the pure elements. Compared to pure americium, the critical mass of the mixture is moderately reduced by the presence of curium (Figure 14, left). However, its spontaneous fission neutron flux is increased by a factor of about  $10^5$  (Figure 14, right), although the curium mass fraction within



**Figure 14.** Variation of critical mass (left) and spontaneous fission neutron flux (right) of americium plus curium present in the spent fuel during interim storage.

**Table 2.** Critical masses of bare metal spheres of transuranium elements produced in light water reactors and their spontaneous fission (SF) induced neutron flux.

| Element            | Critical mass [kg]      |       |      | SF neutron flux [ $\text{kg}^{-1} \text{s}^{-1}$ ] |                      |                   |
|--------------------|-------------------------|-------|------|--|----------------------|-------------------|
|                    | Out of core time of [y] |       |      | Out of core time of [y]                            |                      |                   |
|                    | 1                       | 10    | 50   | 1  | 10                   | 50                |
| Plutonium          | 14.7                    | 14.8  | 15.1 | $4.8 \times 10^5$                                  | $5.0 \times 10^5$    | $5.3 \times 10^5$ |
| Neptunium          | 65.1                    | 65.1  | 65.1 | 0.1  | 0.1                  | 0.1               |
| Americium          | 184.2                   | 113.6 | 94.6 | 865  | 1080                 | 1150              |
| Curium             | 22.2                    | 20.4  | 17.8 | $9.9 \times 10^9$                                  | $1.0 \times 10^{10}$ | $7.8 \times 10^9$ |
| Americium + curium | 83.7                    | 87.1  | 88.2 | $2.5 \times 10^9$                                  | $7.8 \times 10^8$    | $1.0 \times 10^8$ |
| For comparison     |                         |       |      |  |                      |                   |
| HEU <sup>a</sup>   | 69.5                    | 69.5  | 69.5 | 5.5  | 5.5                  | 5.5               |
| WGPu <sup>b</sup>  | 10.3                    | 10.3  | 10.3 | $1.1 \times 10^5$                                  | $1.1 \times 10^5$    | $1.1 \times 10^5$ |

<sup>a</sup>80% uranium-235.

<sup>b</sup>Weapon-grade plutonium. 95% plutonium-239, 5% plutonium-240.

50 years after fuel irradiation declines from 25% to 1%. Even after extended interim storage of the spent fuel, handling of a mixture of americium and curium will hence require massive shielding.

### Fuel cycle implications

The proliferation risks of the various elements and element mixtures that are intended to be isolated and handled within a partitioning and transmutation fuel cycle can be deduced from Table 2. It shows critical masses and spontaneous neutron fluxes at various times after fuel irradiation and, for comparison, of typical weapons grade uranium and plutonium. These data reveal that both neptunium and americium produced in light water reactors may be highly proliferation sensitive, as its nuclear properties (critical mass, spontaneous fission neutron source strength) resemble those of highly enriched uranium.

Estimates of the masses of plutonium, neptunium, and americium that need to be processed if the spent fuel originating from light water reactors

is destined to feed a partitioning and transmutation fuel cycle are given in [Table 3](#). They are based on an amount of 10,000 MT of accumulated spent irradiated fuel produced by light water reactors.<sup>22</sup> Masses discharged annually from a commercial light water reactor are included for comparison. [Table 3](#) additionally provides the corresponding numbers of critical masses. These were calculated for bare metal spheres as well as spheres surrounded by a beryllium reflector to indicate their variability. These data document that introduction of a full-scale industrial partitioning and transmutation fuel cycle irrespective of fuel burnups and out-of-core times will handle large amounts of proliferation relevant masses not only of plutonium, but also of neptunium and americium. For these elements complete safeguards will be required at all fuel cycle facilities and operations, including storage, transport, reprocessing, fuel manufacture and irradiation in a nuclear reactor.<sup>23</sup>

Finally, the time periods necessary to diminish proliferation sensitive elements through transmutation are considered. These generally will differ between reactor core designs optimized for transmutating plutonium and minor actinides, respectively. For these two options, changes in plutonium and minor actinide masses, which result from operation of a 1200 MWt fast reactor for the considered time periods, are given in [Table 4](#). They assume that the plutonium and minor actinide stocks are high enough for providing actinide bearing fuel elements at each reloading. Variations of out-of-core times only slightly affect the simulations. The most striking

**Table 3.** Inventories of plutonium and proliferation sensitive minor actinides and numbers of their critical masses discharged annually per reactor and accumulated in 10,000 MT of spent light water reactor fuel.

| Element   | Discharge <sup>a</sup> [kg] | No. of critical masses |           | Inventory <sup>b</sup> [MT] | No. of critical masses |           |
|-----------|-----------------------------|------------------------|-----------|-----------------------------|------------------------|-----------|
|           |                             | Bare sphere            | Reflector |                             | Bare sphere            | Reflector |
| Plutonium | 233.6                       | 16                     | 4.2       | 111.4                       | 7530                   | 19820     |
| Neptunium | 13.4                        | 0.2                    | 0.3       | 6.86                        | 105                    | 148       |
| Americium | 6.4                         | 0.03                   | 0.04      | 9.86                        | 87                     | 128       |

<sup>a</sup>20 MT, 1 GWe, 50 GWd/MT burnup.

<sup>b</sup>10,000 MT, 50 GWd/MT burnup, 10 y out of core.

**Table 4.** Masses transmuted by a 1200 MWt fast reactor with cores designed for plutonium and minor actinide (MA) conversion, respectively.<sup>a</sup>

| Time of operation [y] | Plutonium converter |         | MA converter   |         |
|-----------------------|---------------------|---------|----------------|---------|
|                       | Plutonium [MT]      | MA [MT] | Plutonium [MT] | MA [MT] |
| 25                    | -6.6                | +3.6    | -4.7           | +0.9    |
| 50                    | -11.6               | +4.4    | -7.0           | -2.3    |
| 75                    | -15.8               | +4.8    | -9.3           | -5.6    |
| 100                   | -20.1               | +5.1    | -11.6          | -9.2    |
| 125                   | -24.7               | +5.4    | -13.9          | -12.6   |
| 150                   | -29.5               | +5.6    | -16.8          | -16.1   |

<sup>a</sup>Numerical values have been deduced from [Figure 2](#) of "Plutonium and Minor Actinides Incineration Options."

result is that even in optimized reactors transmutation of the actinides is an inefficient process. This is primarily because high neutron absorption cross sections of uranium-238 and other actinides remain, resulting in the build-up of additional plutonium and minor actinides competing with their fission. If optimized for plutonium transmutation, such a reactor even increases the inventory of the minor actinides by a mass that will be fissioned by an actinide converter only after about 75 years of operation. Theoretically this actinide build-up could be decreased by replacing the uranium-238 by an inert fuel material. For safety reasons, however, the uranium-238 is indispensable, as its high negative Doppler reactivity coefficient limits the risk of severe power excursion accidents.<sup>24</sup>

Comparison with [Table 3](#) indicates that stocks of plutonium and minor actinides produced in a country which has generated a substantial fraction of its electricity by light water reactors will only be significantly reduced by operating transmutation reactors for more than a century. For example, Vezzoni et al. estimate that six to seven 1200 MWt fast reactors will need to run for 150 years to eliminate the plutonium and minor actinide inventories in the spent fuel that will have accumulated in Germany until shut-down of its last commercial reactor.<sup>25</sup> This result is still over-optimistic, as their simulations assume unrealistically short interim storage times between successive irradiations (see the Materials and Methods section). If it is assumed that the development and prototype testing of a full-scale industrial partitioning and transmutation fuel cycle will require some decades, the existence of a globally accepted treaty, including comprehensive safeguards agreements like the Non-Proliferation Treaty, will be required for the next 200 years if the risks created by introducing this technology are to be limited.

From [Table 4](#), a mean annual plutonium reduction rate of about 0.2 MT is deduced, if the reactor is optimized for plutonium conversion, and of about 0.1 MT in case of an MA converter. From the data given in [Table 3](#), it follows that such a reactor could annually eliminate the plutonium present in about 20 MT of spent fuel and the minor actinides in 60 MT, respectively. Noting that modern high burnup light water reactors will unload approximately 20 MT of spent fuel per GWe annually (see [Table 1](#)),<sup>26</sup> it becomes obvious that, in countries generating a substantial proportion of their electricity by light water reactors, even a fleet of transmutation reactors will only attenuate the accumulation of plutonium and the minor actinides. Stabilization or even gradual reduction of their inventories requires the shift to almost 100% electricity generation by fast reactors, some of which are optimized for plutonium and minor actinide conversion.<sup>27</sup> Economic analyses of such a nuclear energy generating system are not available. However, past experience has documented that sodium-

cooled fast reactors and actinide processing fuel cycle facilities may be economically uncompetitive compared to current light water reactor based once-through fuel cycles.<sup>28</sup>

## Conclusion

The realization of an industrial-scale partition and transmutation fuel cycle will induce potentially challenging proliferation issues. Current concepts envisage separating plutonium and the minor actinides neptunium, americium, and potentially curium from spent light water reactor fuel and reducing their masses by fission in fast reactors. In this paper, we provide data on critical masses and spontaneous fission neutron background for the isotopic compositions of the actinides of interest and their evolution in light water reactor fuel of various burnups and during spent fuel storage. These data are complemented by generic estimates of total inventories of these elements present in a full-scale partitioning and transmutation fuel cycle and of the time periods required for significantly reducing their proliferation potential.

The isotopic mixtures of plutonium, neptunium, and americium present in spent light water reactor fuel show low to moderate critical masses and spontaneous fission neutron fluxes for the considered burnups and up to storage times of 50 years. Thus, all three elements are highly proliferation sensitive and their separation and handling within a partitioning and transmutation nuclear fuel cycle requires complete safeguards. Coprocessing the americium with curium would effectively reduce its proliferation risk, but exaggerate shielding requirements and occupational radiation exposures during manufacture of actinide bearing fuel, transport, and handling.

The proliferation risk of the partitioning and transmutation concept is corroborated by the fact that even in a medium sized fuel cycle, multiples of the critical masses of the sensitive elements need to be separated, stored, transported, handled, and irradiated. Elimination rates are limited even in fast reactors designed and optimized for the transmutation of plutonium and minor actinides. Consequently, multiple recycling of these elements is necessary, requiring reactor operation well beyond a century for effectively reducing their initial stocks. Therefore, partitioning and transmutation would have to continue well past the time any globally accepted nonproliferation agreements, including comprehensive safeguards, will be in force, further increasing proliferation risk.

## Notes and References

1. Active Member are Canada, the People's Republic of China, Euratom, France, Japan, the Republic of Korea, the Russian Federation, the Republic of South Africa,

- Switzerland, and the United States, non-active members Argentina, Australia, Brazil, and the United Kingdom.
2. These are gas-cooled fast reactor, lead-cooled fast reactor, molten salt reactor, sodium-cooled fast reactor, supercritical-water-cooled reactor, and very-high-temperature reactor. For details see “Technology Road Map Update for Generation IV Nuclear Energy Systems,” Generation IV International Forum, accessed 1 August 2017, [https://www.gen-4.org/gif/jcms/c\\_60729/technology-roadmap-update-2013](https://www.gen-4.org/gif/jcms/c_60729/technology-roadmap-update-2013).
  3. International Atomic Energy Agency, *Implications of Partitioning and Transmutation in Radioactive Waste Management*, Technical Reports Series No. 435 (Vienna: IAEA, 2004), 4–7.
  4. For salt as host rock: Dieter Buhmann, “Relevance of Partitioning and Transmutation to Long-Term Safety of a Repository in Rock Salt under Realistic Disposal Conditions,” 7th International Conference on Radioactive Waste Management and Environmental Remediation, Nagoya, Japan, 26–30 September 1999. For clay as host rock: Nagra (Nationale Genossenschaft für die Lagerung radioaktiver Abfälle), *Project Opalinus Clay. Safety Report. Demonstration of Disposal Feasibility for Spent Fuel, Vitrified High-Level Waste and Long-Lived Intermediate-Level Waste (Entsorgungsnachweis)*, Technical Report 02-05 (Wettingen, Switzerland, 2004). For granite as host rock: SKB (Svensk Kärnbränslehantering AB), *Long-term Safety for the Final Repository for Spent Nuclear Fuel at Forsmark*, Technical Report TR-11-01. Stockholm, Sweden. In tuff as host rock, some contribution of plutonium to potential human radiation exposure have been predicted for pessimistic assumptions, see U.S. Department of Energy, Office of Civilian Radioactive Waste Management, *Yucca Mountain Repository License Application – SAFETY ANALYSIS REPORT*, Chapter 2: Repository Safety After Permanent Closure, Report DOE/RW-0573, Rev. 0, (Washington, DC, 2008).
  5. Sodium cooled fast reactors today are operated in Russia, India and Japan (although shut down after the Fukushima accident in 2011), whereas safety issues, proliferation concerns and low operational availabilities have resulted in the termination of sodium-cooled fast breeder programmes in France, Germany, the United Kingdom, and the United States.
  6. An example of an event sequence initiating such a reactivity excursion is leakage of oil from a bearing of a primary pump into the sodium of the primary loop, see Gerald Kirchner, “Reactivity Effects of Oil Contamination in the Primary Cooling Loop of a Sodium-cooled Fast Reactor,” Proceedings of the International Conference on Nuclear Criticality Safety, Oxford, U.K., 9–13 September 1991, pp. V-167 -V-173.
  7. Jungmin Kang and Frank von Hippel, “Limited Proliferation-Resistance Benefits from Recycling Unseparated Transuranics and Lanthanides from Light-Water Reactor Spent Fuel,” *Science and Global Security* 13 (2005): 169–181.
  8. E. D. Clayton, “Fissionability and Criticality: From Protactinium to Californium and Beyond,” *Nuclear Science and Engineering* 52 (1973): 417–420. R. M. Westfall, “Critical Masses for the Even-Neutron-Numbered Transuranium Actinides,” *Nuclear Science and Engineering* 79 (1981): 237–239. R. Q. Wright, W. C. Jordan, and R. M. Westfall, “Critical Masses of Bare Metal Spheres Using SCALE/XSDRN,” Proceedings of the Annual Meeting of the American Nuclear Society, San Diego, 4–8 June 2000, 167, cited by Kang and von Hippel, “Limited Proliferation-Resistance,” 172.
  9. Alternatively, pyrometallic separation provides a potentially promising technology. However, its development and successful demonstration at industrial scale will require high research and development efforts, “Status of Developments in the Back End of

- the Fast Reactor Fuel Cycle,” International Atomic Energy Agency, Nuclear Energy Series, No. NF-T-42 (Vienna, 2011).
10. G. Modolo, A. Wilden, A. Geist, D. Magnusson, and R. Malmbeck, “A Review of the Demonstration of Innovative Solvent Extraction Processes for the Recovery of Trivalent Minor Actinides from PUREX Raffinate,” *Radiochimica Acta* 100 (2012): 715–725.
  11. Barbara Vezzoni, Fabrizio Gabrielli, Andrei Rineiski, Alexandra Schwenk-Ferrero, Vincenzo Romanello, Werner Maschek, Concetta Fazio, and Massimo Salvatore, “Plutonium and Minor Actinides Incineration Options Using Innovative Na-Cooled Fast Reactors: Impacting on Phasing-out and On-going Fuel Cycles,” *Progress in Nuclear Energy* 82 (2015): 58–63.
  12. This value had been set as minimum interim storage period for the layout of the German commercial Wackersdorf fuel reprocessing plant construction of which was stopped in 1989 due to unexpected high cost increases.
  13. “Plutonium and Minor Actinides Incineration Options”.
  14. RSICC Computer Code Collection, CCC-732, *SCALE: A Comprehensive Modeling and Simulation Suite for Nuclear Safety Analysis and Design, Version 6.1*, Oak Ridge National Laboratory, Technical Report ORNL/TM-2005/39 (Oak Ridge, TN, 2011).
  15. During its first irradiation cycle, a fuel assembly was modeled to be surrounded by third cycle assemblies, during its second cycle by second cycle assemblies and during its third cycle by first cycle assemblies.
  16. Mark D. deHart, and Stephen M. Bowman, “Reactor Physics Methods and Capabilities in SCALE,” *Nuclear Technology* 174 (2011): 196–213.
  17. Germina Ilas, Ian C. Gauld, and Georgeta Radulescu, “Validation of New Depletion Capabilities and ENDF/B-VII data libraries in SCALE,” *Annals of Nuclear Energy* 46 (2012): 43–55.
  18. Densities were taken from David R. Lide (Editor), “CRC Handbook of Chemistry and Physics, 85<sup>th</sup> Edition,” CRC Press (Boca Raton, FL, 2004).
  19. Sedat Goluoglu, Lester M. Petrie, Jr., Michael E. Dunn, Daniel F. Hollenbach, and Bradley T. Rearden, “Monte Carlo Criticality Methods and Analysis Capabilities in SCALE,” *Nuclear Technology* 174 (2011): 214–235.
  20. “SCALE: A Comprehensive Modeling and Simulation Suite for Nuclear Safety Analysis and Design, Version 6.1”.
  21. J. Carson Mark, Frank von Hippel, and Edward Lyman, “Explosive Properties of Reactor-Grade Plutonium,” *Science and Global Security* 17 (2009): 170–185.
  22. For comparison, there were 62,683 tons of commercial spent fuel accumulated in the United States at the end of 2009 (U.S. Nuclear Regulatory Commission, accessed 27 October 2017, <https://www.nrc.gov/waste/spent-fuel-storage/faqs.html>) and 8,379 tons in Germany at the end of 2014 (Federal Ministry for the Environment, Nature Conservation, Building and Nuclear Safety, Inventory of Radioactive Wastes, 2015).
  23. This has been specified by DoE, see U.S. Department of Energy, “Nuclear Material Control and Accountability,” DOE O 474.2 Chg 4 (PgChg), (Washington, D.C, 13 September 2016).
  24. Generally fast reactors show an overall positive reactivity coefficient causing the risk of power excursions and severe core disruptive accidents, often called “Bethe-Tait” accident. Design of a fast reactor with plutonium and minor actinide fuel exhibiting a negative reactivity coefficient is still an unresolved problem.
  25. “Plutonium and Minor Actinides Incineration Options”.

26. For comparison, installed capacities are 13.6 GWe in Canada, 33.6 GWe in China, 63.1 GWe in France, 26.9 GWe in Russia, 99.6 GWWe in the United States, “World Nuclear Power Reactors & Uranium Requirements, August 2017”, World Nuclear Association, accessed 21 August 2017, <http://www.world-nuclear.org/information-library/facts-and-figures/world-nuclear-power-reactors-and-uranium-requireme.aspx>.
27. “Plutonium and Minor Actinides Incineration Options”.
28. This was the major reason for shutting down the only commercial sodium-cooled fast reactor, Super-Phenix in France, see “Liquid Metal Cooled Reactors: Experience in Design and Operation,” International Atomic Energy Agency, IAEA-TECDOC-1569 (Vienna, 2007). For economic challenges of building and operating fuel cycle installations, see “Mixed-Oxide Fuel Fabrication Plant and Plutonium Disposition: Management and Policy Issues,” Congressional Research Service, R43125 (14 December 2017).