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WHICH ELEMENTS SHOULD BE RECYCLED FOR A COMPREHENSIVE FUEL CYCLE?

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Uranium recovery can reduce the mass of waste and possibly the number of waste packages that require geologic disposal. Separated uranium can be managed with the same method (near-surface burial) as used for the larger quantities of depleted uranium or recycled into new fuel. Recycle of all transuranics reduces long-term environmental burden, reduces heat load to repositories, extracts more energy from the original uranium ore, and may have significant proliferation resistance and physical security advantages.

Recovery of short-lived fission products cesium and strontium can allow them to decay to low-level waste in facilities tailored to that need, rather than geologic disposal. This could also reduce the number and cost of waste packages requiring geologic disposal. These savings are offset by costs for separation, recycle, and storage systems. Recovery of technetium-99 and iodine-129 can allow them to be sent to geologic disposal in improved waste forms. Such separation avoids contamination of the other products (uranium) and waste (cesium-strontium) streams with long-lived radioisotopes so the material might be disposed as low-level waste. Transmutation of technetium and iodine is a possible future alternative.

I. INTRODUCTION

This paper summarizes how the selection of elements to be recycled (which transuranics) impacts a broad range of recycle-related metrics. The main goals of the Advanced Fuel Cycle Initiative are to:[1,2,3]

- Recycle used nuclear fuel to minimize waste and control weapons-usable inventories.
- Reuse valuable parts of used nuclear fuel to maximize the energy derived from uranium ore.
- Enable the expansion of nuclear power while only requiring a single repository through the end of the century.
- Enable the global expansion of nuclear energy while also reducing proliferation risk and enhancing nuclear security.

The AFCI addresses the needs associated with increasing the sustainability of nuclear energy. First, the AFCI investigates fuel cycles that would convert current waste liabilities into energy source assets, helping ensure that availability of uranium ore resources does not become a constraint on the expansion of nuclear energy. Second, all of the AFCI fuel cycles would incorporate more proliferation-resistant technologies and designs than employed in current international practice, would reduce the inventory of weapons-usable material by consuming plutonium more quickly, and would reduce the need for uranium enrichment. Third, AFCI is developing technologies that should allow in the future more efficient disposition of used fuel and high-level waste, helping to delay the need for additional geologic repositories into the next century. While accomplishing these objectives, AFCI seeks to ensure competitive economics and maintain the excellent safety record for the entire nuclear fuel cycle.

Which chemical elements should be recovered to achieve various potential objectives? Section II explains that the original focus of recycling was energy recovery, which led to separation of plutonium and uranium only. Section III describes how the focus has changed to add waste management and proliferation risk management. Sections IV and V address fundamental properties of used nuclear fuel, radiotoxicity and heat generation, which impact long-term waste management in any country, independent of waste sites. Section VI considers Yucca Mountain parameters as a specific example site to ensure that the site-independent analyses remain valid. Section VI considers parameters that may influence economics and proliferation resistance.

II. ORIGINAL FOCUS OF RECYCLING WAS ENERGY RECOVERY

Figure 1 shows that about 95% of used nuclear fuel mass is useful material, uranium (94%) and the transuranic elements of neptunium, plutonium, americium, and curium (1%). The other 5% of used fuel comprises fission products.

Figure 2 shows that uranium and plutonium constitute 99.8% of the energy content in used fuel. This is why current nuclear fuel recycling efforts in France, the United Kingdom, Japan, and Russia use separation technologies such as PUREX that only separate uranium and plutonium. That is essentially all that is useful for energy recovery.

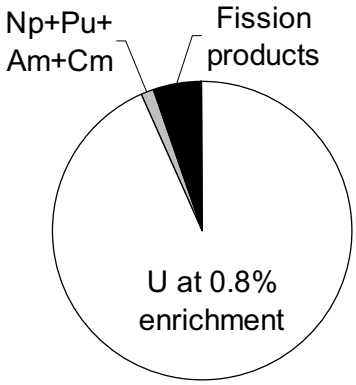

Figure 1. Composition of used nuclear fuel at 50 MW-day/kg burnup

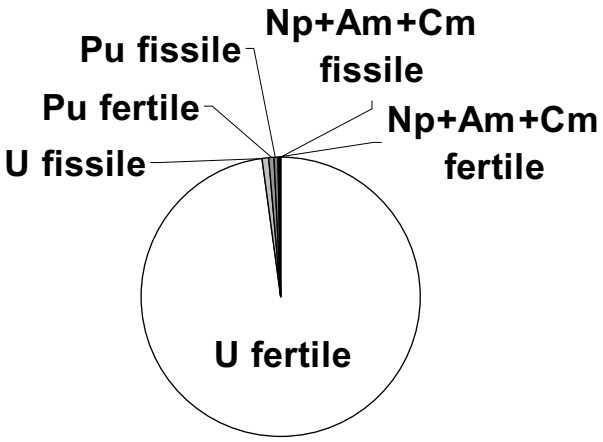

Figure 2. Energy content of used nuclear fuel at 50 MW-day/kg burnup

These facts have been known for decades; recycling used fuel for its energy content only requires recovery of uranium and plutonium. The plutonium is mostly fissile and can be readily recycled in thermal or fast reactors; the uranium is mostly fertile and best recycled in fast reactors. To use more of the energy content in the original uranium ore, the unused uranium and transuranic elements in used fuel must be recovered. Sustained recycling is needed to substantially improve energy recovery.

III. NEW FOCUS

Trends in other industries suggest that recycling will continue to grow in acceptance, with better economics, whereas waste disposal will continue to drop in acceptance, with worse economic and social costs. The U.S. census bureau statistics indicate that about a quarter of municipal waste is now burned for energy and another quarter is recovered for reuse. Recycling can and should address constraints other than energy recovery, in particular waste management. What can recycling of used nuclear fuel do for waste management?

IV. SITE INDEPENDENT RADIOTOXICITY

To answer this question, it is necessary to look at the composition of used nuclear fuel from other perspectives than energy recovery. Like other wastes, used nuclear fuel is toxic, primarily because of radioactive isotopes. Internationally, a common way to describe the hazard of used fuel is radiotoxicity, which is the inventory of radioisotopes divided by their relative hazard to humans. Figure 3 shows the radiotoxicity of used nuclear fuel relative to the uranium ore that started the process. It remains more radiotoxic than uranium ore for about a million years. The "actinides" plotted in the figure include uranium and transuranic elements.

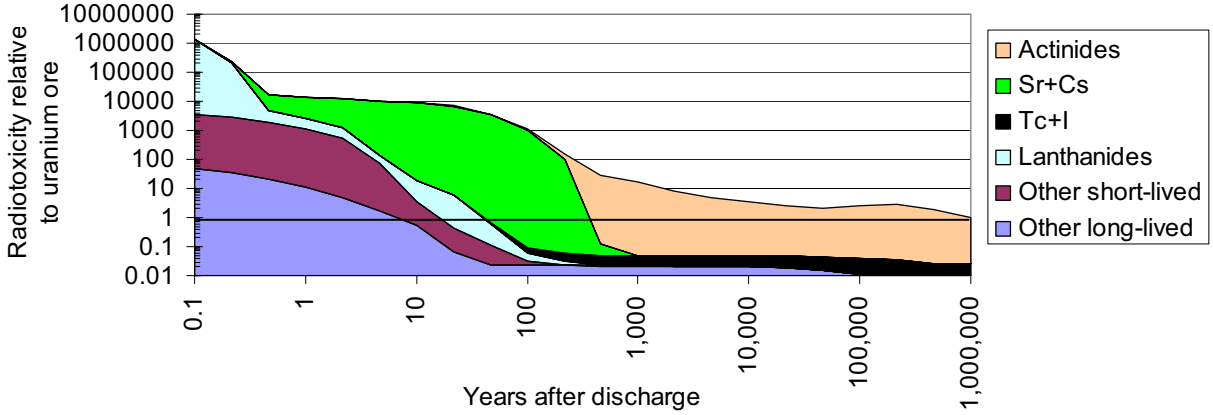

Figure 3. Radiotoxicity of used fuel relative to uranium ore. Actinides are uranium, transuranic elements, and their decay products. Sr+Cs is strontium and cesium. Tc+I is technetium and iodine. Other long-lived elements include selenium, palladium, tin, and antimony. All other fission products are grouped into "other short-lived" elements. Calculations for used LWR fuel with 50 MW-day/kg burnup.

Next, consider how to "peel back the onion," to reduce the amount and longevity of radiotoxicity. The first step is to remove the uranium and transuranic elements (actinides); these can be fissioned to produce energy. Thus, proper use of technology can turn them from waste liabilities into energy assets. If the energy content elements are indeed removed from waste, the waste is less radiotoxic than uranium ore in less than 1,000 years. Humans have experience with successful engineering constructs with a time scale of a thousand years. Humans lack engineering experience for hundreds of thousands of years.

In the first few decades, the radiotoxicity is dominated by cesium and strontium (cesium-137 and strontium-90), which have about 30-yr half-lives. Most AFCI recycle strategies call for these elements to be removed from the other fission products and managed separately due to their high heat production as they decay. The radiotoxicity of the residual long-term waste falls below uranium ore in less than 100 years. Basically, separation of cesium and strontium divides the waste into a short-term component (cesium-strontium) that may only have to be managed for a few hundred years and a residual long-term component that has relatively low long-term radiotoxicity.

Further separation of waste streams accomplishes little. Within one year after discharge from a nuclear power plant, the most radiotoxic elements are the lanthanides, also called the "rare earths." But, these have decayed in the first few decades, before waste would be emplaced in the repository. Two dominant long-lived elements - technetium and iodine - are inherently separated from other materials in the UREX+ process and would be made into appropriate waste forms.

V. SITE INDEPENDENT HEAT GENERATION

Like radiotoxicity, heat generation rates are important to almost any waste disposal concept. Excessive heat can lead to excessive temperatures because of the difficulty of providing for heat removal for long time periods. Only natural methods such as heat conduction or convection can be depended on for thousands of years. This subsection shows that the same basic pattern seen for radiotoxicity – transuranics, cesium-strontium, technetium-iodine, lanthanides, other – occurs for the heat generation rate.

There is an obvious benchmark for radiotoxicity, the radiotoxicity of natural uranium ore that starts the process. There is not such an obvious benchmark for heat generation rate because the rate of heat removal depends on how the waste disposal site is arranged and the heat conductivity of surrounding material.

Figure 4 shows the heat generation for used fuel. As with radiotoxicity, the most important material to remove from used fuel are the actinides, followed by cesium-strontium. Figure 4 shows the heat generation rate after the energy content (uranium, transuranic) and high-heat elements (cesium-strontium) are removed from residual waste. One could go further and remove the lanthanides, technetium, and iodine, but figure 4 shows that the heat generation rate would not be greatly reduced.

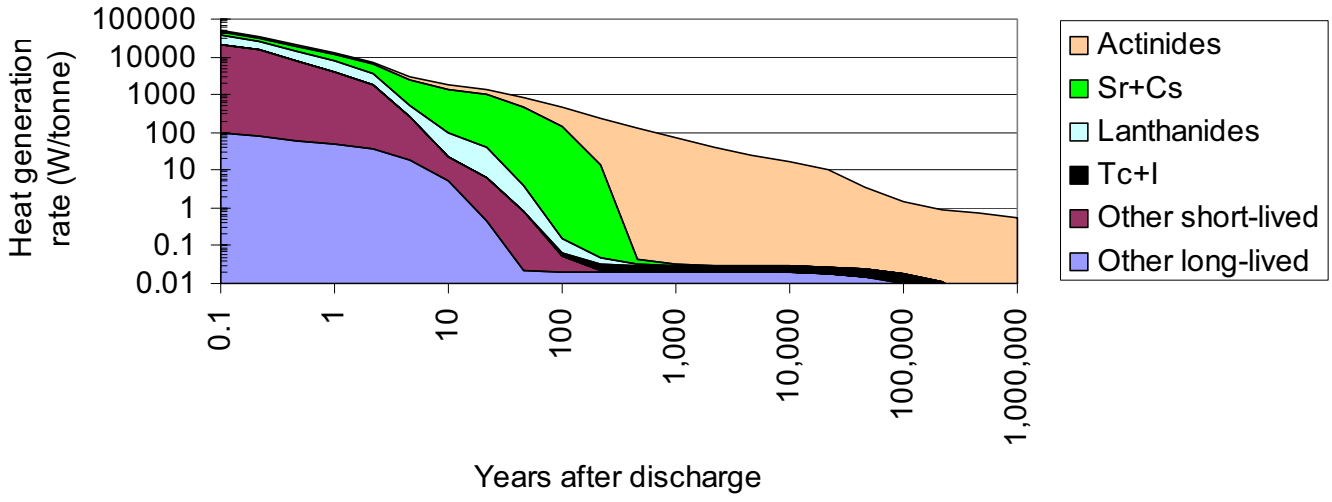

Figure 4. Heat generation rate of used fuel, at 50 MW-day/kg-HM burnup.

VI. POTENTIAL REPOSITORY CONSTRAINTS

Radiotoxicity and heat generation rates are useful metrics, but do not, by themselves, indicate the risk and cost of disposal of used nuclear fuel or residual wastes. These require looking at specific proposals for waste disposal in actual candidate sites. No country yet has a repository in operation for used nuclear fuel, and therefore there are no hard numbers for the risk and cost of used fuel disposal. The preceding discussion does suggest that the sequence of removing material from residual waste should be the energy content elements (uranium and transuranics), cesium-strontium, and then technetium-iodine. Here, the analysis goes deeper, from site independent to site dependent, to ensure that the preceding site-independent analyses do not lead to misleading conclusions.

The U.S. proposes to dispose of used fuel in a geologic repository at Yucca Mountain, Nevada. Examination of some of the constraints on such disposal helps look us deeper at how recycling can address waste management challenges. It is stressed that Yucca Mountain parameters are used only as a benchmark; the principles and approaches are generic to geologic repositories.

There are three potential constraints on geologic repository capacity – statutory capacity limits, dose limits, and space capacity limits. The two technical limits are described in the following subsections.

VI.A. Dose Limits

Regulations limit the hypothetical dose to the public from postulated release of waste in future centuries and millennia. These dose limits are independent of the amount of waste emplaced or the amount of electricity generated from the fuel that results in the waste. For example, the currently proposed dose limits for the proposed geologic repository are 15 mrem/yr from when waste is emplaced in the repository to 10,000 years and 350 mrem/yr from 10,000 years to 1,000,000 years. For the 10,000-year limit, the estimated dose tends to peak at 10,000 years, meaning that is the key time period for assessment of compliance. For the longer-term dose limit, the dose generally peaks between 100,000 and 1,000,000 years - increased time means more time for waste to escape and migrate to the public but also more time for radioactivity to decay.

Often, the hypothetical dose via ground water exposure is a dominant contributor to estimated dose. It depends on a host of factors and complex modeling of the repository site and waste form behavior. To see if the conclusions on the importance of different elements is significantly different than those based on radiotoxicity, consider past calculations for the Yucca Mountain repository.[4]

Figure 5 shows the relative contribution to hypothetical peak dose at 10,000 years. The fission products technetium and iodine dominate, even though in figure 3 their radiotoxicity is an order of magnitude lower than the transuranics; this is because they are more mobile. Fortunately, the current calculations indicate several orders of magnitude margin between the calculated hypothetical dose and the dose limit. Thus, even if the long-lived fission products from substantially more years of waste were emplaced in such a repository, the dose limit would likely still be met. If future calculations do show an issue, one approach would be to assure that the durability (slower leaching) of the technetium and iodine waste forms was longer than UOX, if the technetium and iodine were released from the waste form slower than from UOX, the calculated dose would decrease per unit of source term. Another approach would be to transmute these long-lived fission products.[5]

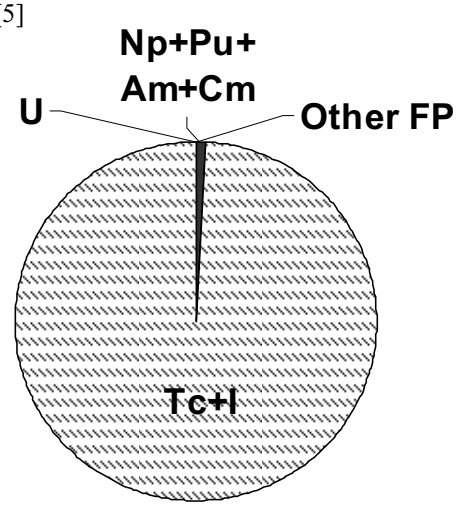

Figure 5. Contributions to hypothetical dose from waste in an oxidizing environment at 10,000 years from elements in 5-yr old LWR fuel.

Figure 6 shows the relative contribution to hypothetical peak dose, which occurs about a half-million years after discharge from the reactor. The actinides (uranium plus the transuranic elements (Np, Pu, Am) and their daughters comprise about 99% of the potential dose at 500,000 years;[1] next is technetium and iodine at about

[1] To better understand figure 6, note that the single most important isotope is Np237, which is a daughter of both Pu241 and Am241. Thus, the contribution to future Np237 dose is reflected in three slices of figure 6, neptunium itself, the amount of plutonium that decays into Np237, and the amount of americium that decays into Np237. The relative contributions are graphed this

1%. This is consistent with figure 3, which shows the contribution of technetium (Tc) and iodine (I) to total radiotoxicity between 10,000 and 1,000,000 years to be about two orders of magnitude lower than the actinides. Other long-lived fission product (FP) isotopes make an even smaller contribution. Thus, the site-specific example and the site-independent analysis point in the same direction: recycle the transuranics and uranium and ensure technetium and iodine hazards are properly managed. The total dose commitment from the transuranic elements comes to about 99%, so that removal and recycle of these elements may reduce the potential long-term dose from the repository by about a factor of 100.[2] If future calculations show an increase in the relative contribution from technetium and iodine, options would be higher durability (slower leaching) waste forms or transmutation.

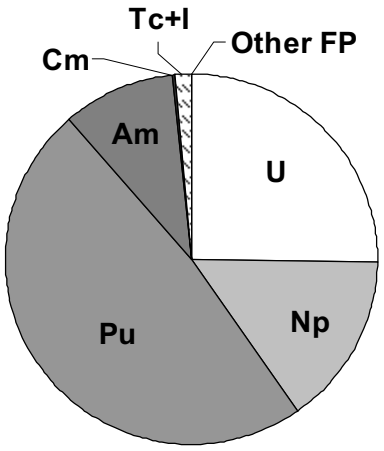

Figure 6. Contributions to hypothetical dose from waste in an oxidizing environment at 500,000 years from elements in 5-yr old LWR fuel.

VI.B. Space Capacity Limits

Several factors are involved in estimating how much waste can fit into the allowable space such as the mass of waste, the volume of packaged waste, the tunnel wall temperature, and temperatures in the rock between tunnels. The peak tunnel wall temperature can occur when the waste is emplaced or just after the repository is closed (and thus the tunnels are no longer being cooled by ventilation) - both are within several decades of when waste is discharged from the reactor. The peak temperature in the rock between tunnels can occur centuries after waste is discharged from the reactor. The technical capacity can depend on the mass, volume, short-term heat (decades), long-term heat (centuries), and long-term hypothetical dose (millennia) of emplaced waste.

VI.B.1. Unprocessed waste mass

The unprocessed waste mass is determined by how much mass in used fuel is removed and recovered for future use. Uranium is about 94% of discharged fuel; transuranics are about 1% (previous figure 4). Thus, there is potential to reduce the mass of waste by a factor of 20 if all of this 95% is recycled.

VI.B.2. Packaged waste volume

The volume of waste is determined by the mass of material to be disposed times the concentration of waste in the final waste form, adjusted to reflect the volume of surrounding waste packing. For example, one potential waste form is borosilicate glass. For each material to be put into such a waste form, there is a maximum concentration that will dissolve into the glass, which determines the maximum waste loading. The glass is then put into some sort of package. Work continues on appropriate waste forms, waste loadings, and packaging for specific waste streams and compositions. For example, which waste streams should be combined into a single waste form or kept separate.

Removal of uranium from the residual high-level waste both recovers energy content and helps reduce the mass and volume of residual high-level waste. The high-level waste volume is reduced by keeping long-lived contaminants such as technetium and iodine out of products (such as uranium and transuranic elements) and short-lived wastes (such as cesium-strontium) so that those products and short-lived wastes can be consumed or disposed in ways other than high-level waste.

VI.B.3. Waste heat generation

The heat generated by waste must be factored into the selection and design of the waste form, waste packaging, and waste site. The temperature of wastes may increase depending on the detailed design, yet waste forms and packages must maintain their integrity and the performance of the waste site must not be compromised. For a repository similar to the Yucca Mountain site, analyses have shown that the heat generated from the time the repository is closed (ventilation stops) to about 1500 years is a metric that helps compare options. Figure 7 shows the contributors to this time-integrated "heat commitment" from used LWR fuel to a repository.

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way because it shows which elements have to be separated before disposal.

[2] There are non-linearities in the calculation of hypothetical dose from the repository. Thus, reducing the source term by two orders of magnitude may not result in the same reduction in estimated dose.

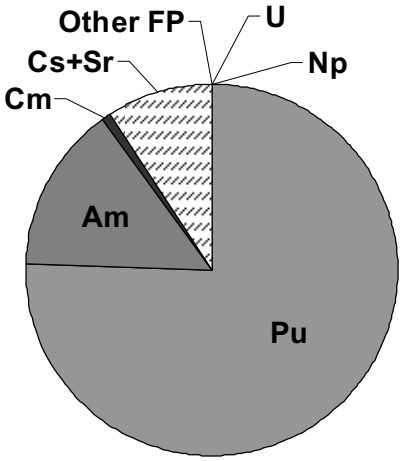

Figure 7. Heat to repository during key time period (50-1500 yr) from elements in 5-yr old fuel.

Plutonium and americium (and their decay daughters) comprise about 90% of the heat commitment; thus, to reduce the "heat commitment" by a factor of ten requires removal of plutonium and americium (and their decay daughters).[3] The next most important heat generators are cesium and strontium, bringing the total heat commitment to about 99%, so that removal of the transuranics, cesium, and strontium would reduce the heat commitment to the repository by a factor of about 100. Detailed calculations validate this these general trends.[6] And, this result is consistent with figure 4, which shows that in the decades to centuries after discharge from a reactor, the highest heat generators are the actinides (in this case virtually all due to the transuranics rather than uranium), followed by cesium and strontium. The site-specific example here and the site-independent analysis point in the same direction: recycle the transuranics and separately manage the heat from cesium and strontium.

7. PROLIFERATION RESISTANCE AND PHYSICAL SECURITY

[3] Note that the single most important isotope is Am241. Its contribution to heat in the time period from 50 to 1500 years is reflected in two slices in figure B-8, plutonium (which includes Pu241 that decays into Am241) and americium (which includes Am241). The partition between plutonium and americium depends on the age of the used fuel because of the decay of Pu241 (14.4-year halflife) into Am241. With 5-year old fuel, plutonium and its daughters (such as Am241) are about three-quarters of the heat commitment; material that starts as americium and its daughters is about 15%. As the fuel ages, the plutonium fraction decreases and the americium fraction increases.

Any program or project aimed at future nuclear energy technologies must properly address the issue of "proliferation resistance" of the overall system in which the advanced technologies would be deployed. Both institutional measures (which are not considered here, but have the potential to be the primary factor in reducing proliferation risk)) and technological measures must be considered. Technological measures to reduce proliferation risk include those that will reduce the attractiveness of materials and processes for weapons purposes, i.e., increasing proliferation technical difficulty, proliferation time and/or cost). Also, the technical proliferation risk reduction measures include a variety of steps to increase the efficacy of international safeguards such as safeguards by design and improved monitoring approaches. *The AFCI aims to develop a progressive fuel cycle approach that will set a high standard of proliferation resistance and nuclear security.* To provide a higher standard of proliferation resistance, AFCI technologies must reduce nuclear proliferation risk relative to current nuclear fuel cycle technologies such as plutonium separation technology (PUREX). Attention must be paid to proliferation resistance measures that include proliferation technical difficulty (the inherent technical difficulty, arising from the need for technical sophistication and materials handling capabilities, to overcome barriers to proliferation), fissile material type, time and cost to overcome proliferation barriers, and detection probability.[7]

The following figures show neutron emission, heat generation, gamma emission, and the bare sphere critical mass for potential recycle materials. Except as noted below, all compositions are for equilibrium recycle composition (i.e. after many recycles) based on reactor physics calculations.[8,9,10,11] Five years is assumed to elapse between reactor discharge and recycled fuel reinsertion into a reactor.

Figure 8 shows neutron emission, higher values complicate weapon physics and may make detection of the presence of transuranics easier (hence improving proliferation resistance) but also increase fuel fabrication costs. For comparison, weapon-grade (WG-Pu) is shown, assuming that WG-Pu has 93.5% Pu239 and 6.5% Pu240. Weapon-grade uranium has even lower neutron emission than WG-Pu and is not shown. The figure shows that the neutron emission from first recycle transuranics (i.e., derived directly from UOX discharge) is about the same whether the material is Pu, NpPu, or NpPuAm. In those three cases, the neutron emission increases only modestly as the material evolves over many recycles toward equilibrium in a thermal reactor (either uranium based mixed oxide fuel (MOX) or uranium-free inert matrix fuel (IMF)). However, if all TRU are recycled (including Cm, Bk, Cf), the neutron emission is two orders of magnitude higher than if Pu, NpPu, or NpPuAm. If recycled strictly in thermal reactors, the neutron emission eventually

increases another two orders of magnitude. If recycled in only fast reactors, the neutron emission can evolve into one order of magnitude higher (than UOX discharge) for fast reactors with transuranic conversion ratio of 0.0 to a slight reduction in neutron emission for TRU CR of 1.1.

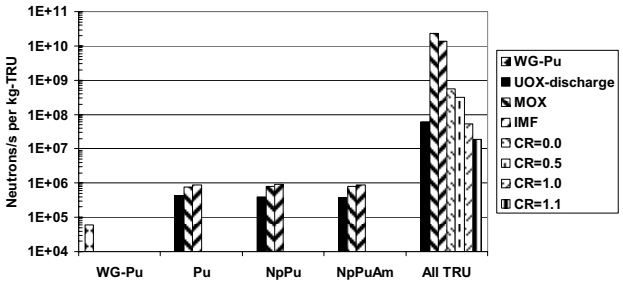

Figure 8. Neutron emission of several recycle options at equilibrium (except UOX-discharge), starting with 50 MW-day/kg-HM burnup UOX with 5 years between recycles.

Figure 9 shows the heat generation rate of recycled transuranics. The IAEA considers plutonium that is 80% Pu238 to be not directly weapon usable because its heat generation rate is too high; thus the figure shows the heat generation rate of 80% Pu238 and WG-Pu for comparison. Unlike neutron emision, recycle of Am incurs a heat generation increase (compare NpPuAm vs NpPu). The penalty in moving from NpPuAm to all-TRU is significant, but more modest than for neutron emission. The underlying reason is that neutron emitters are primarily the very high transuranic isotopes, whereas all of the transuranic isotopes emit heat (have significant decay energy). Thus, as the mix of isotopes shifts to higher transuranics in the all-TRU case, there is a substantial increase in neutron emission. The reduction in neutron and heat as the TRU CR increases is due (in part) to dilution of shorter-lived isotopes by increased production of long-lived Pu239.

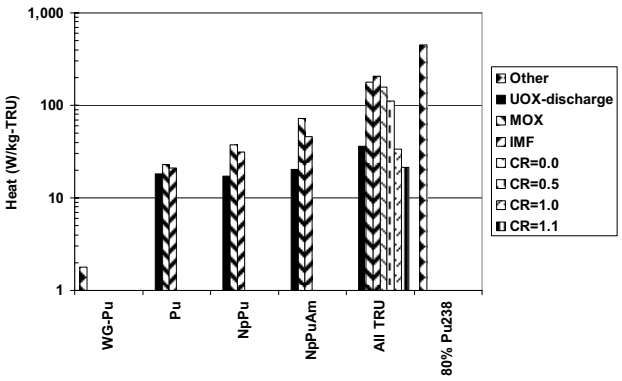

Figure 9. Heat generation rates of recycled materials at equilibrium (except UOX-discharge, WG-Pu, and 80%Pu239)

Figure 10 shows the gamma emission rate; higher gamma emission irradiates would-be proliferators and fuel fabricators. For comparison, the figure includes WG-Pu and UOX with fission products included. The trends are broadly the same as heat generation rates, although the there is generally less difference from first recycle (UOX discharge) to equilibrium for gamma energy than for heat generation. Note that the increase in either heat or gamma emission in progressing from Pu to NpPu to NpPuAm to all-TRU is far more modest than the increase in neutron emission.

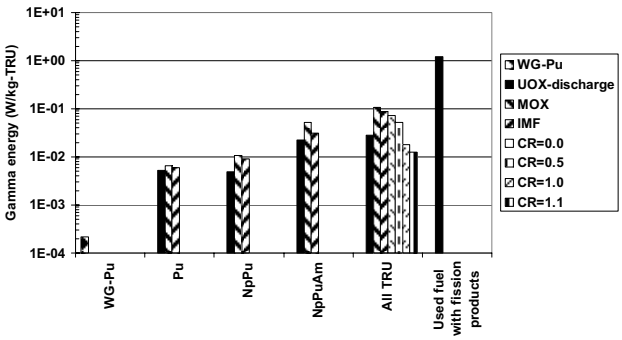

Figure 10. Gamma energy emission of recycled materials at equilibrium (except UOX-discharge, WG-Pu, and used UOX fuel).

Figure 11 shows the bare sphere critical mass of recycle options. With the exception of pure U235 included for comparison, all cases have no uranium. The calculations were done by the author using an estimator of bare sphere critical mass from E. Schneider. All of the recycle cases (Pu, NpPu, NpPuAm, and all-TRU) lie between the bare sphere critical mass values for pure U235 and WG-Pu; there is little variation.

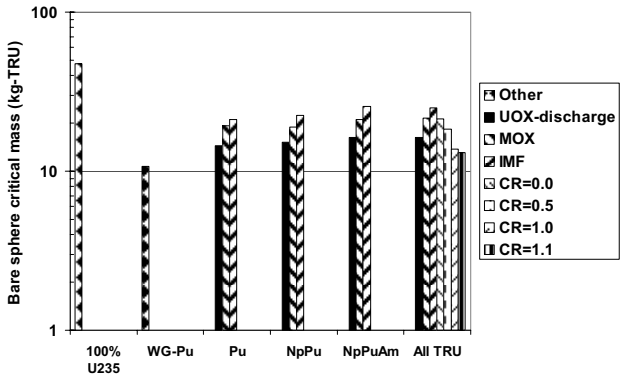

Figure 11. Estimates of the bare sphere critical mass of recycled materials at equilibrium (except UOX-discharge, WG-Pu, and 100% U235)

If U238 is kept with any of the recycle compositions shown in figure 11, the bare sphere critical mass will increase substantially (well over 100 kg-TRU), just as it does for downblending of U235 or WG-Pu.

VIII. CONCLUSIONS

Fuel cycle objectives must simultaneously address waste management, proliferation resistance and nuclear security, energy recovery, economics, and safety. This paper only addresses certain topics; it ignores for example the critical issues of the cost of doing different chemical separations or making different types of fuels.

Disposition paths for all recycle and waste materials must be included, addressing material forms, storage, transportation, and reuse or disposal. It is not sufficient to only recover and recycle uranium and plutonium, as is done with the separation technology PUREX that was developed over 50 years ago.

To improve physical security against threats from sub-national groups, transuranics should be kept together as much as practical. This significantly reduces the material attractiveness because of higher heat load, higher gamma emission, and especially higher neutron emission.

Inclusion of uranium with recycled transuranics increases the bare sphere critical mass meaning that more material would have to be stolen to make a weapon and then either the weapon would have to be larger, or the uranium would have to be separated from the transuranics.

Recycling for U.S. waste management could include the following:

- Recovery and consumption of transuranic elements, turning waste management liabilities into energy assets. This reduces waste heat generation and long-term radiotoxicity and dose.
- Eventual reuse of uranium, when uranium ore prices warrant. This reduces waste mass, probably volume, and long-term radiotoxicity and dose.
- Separate management of the heat from short-lived cesium and strontium to simplify the design of disposal facilities for the residual long-lived wastes.
- Careful attention to the residual hazards posed by long-lived fission products (especially technetium and iodine). As long-lived transuranic isotopes are destroyed isotopes such as Tc-99 and I-129 become dominant and constrain long-term environmental benefits. It may be useful to put these isotopes in especially low-leach-rate waste forms, geochemical environments that inhibit their transport, or transmute them.

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