The effect of burn-up and separation efficiency on uranium utilisation and radiotoxicity

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Abstract

This paper addresses two fundamental cradle-to-grave issues of fuel cycle sustainability. The two primary issues of interest are efficient use of the natural uranium resource (cradle), and management of nuclear waste radiotoxicity (grave). Both uranium utilisation and radiotoxicity are directly influenced by the burn-up achieved during irradiation (transmutation-related) and where applicable the separation efficiency (partitioning-related). Burn-up influences the in-growth of transuranics by breeding them into the fuel cycle. Breeding of transuranic elements is essential to resource sustainability because it increases utilisation of naturally abundant fertile ^{238}U. However, the direct consequence of this build-up is the in-growth of transuranic isotopes which generally increase the source of future geologically committed radiotoxicity. For scenarios involving recycle, separation efficiency influences the degree to which this transuranic source term is removed from active service in the fuel stream and made a disposal legacy of human activity.

Introduction

Uranium utilisation is an issue of fissile conservation within the fuel cycle. Fissile conservation stems from: *i)* increasing fissile breeding from ^{238}U; *ii)* decreasing uranium disposed of as enrichment tails. The uranium utilisation efficiency is defined as the rate that uranium is consumed by fission divided by the rate that uranium is mined from the ground, on an energy normalised basis. The rate of uranium consumption by fission is essentially the average fission Q-value for the system and is approximately 950 GWth-day per tonne of uranium. Hence, complete fission of one tonne of uranium generates ~950 GWth-days of fission energy. Mined uranium mass that is not fissioned, depleted uranium tails, disposed used nuclear fuel, and separation losses represent the extent of under-utilised resource. The ability of a fuel cycle to utilise uranium depends on the ability to convert the entire mined fertile isotope into fission products. We found that greater than one percent uranium utilisation is difficult for most fuel cycles, particularly those involving thermal reactors. This is due to the fact that >99% of all natural uranium is fertile and most fuel cycles require more fissile material than can be converted from this fertile material.

Furthermore, it is important to note that decreases in the mass, heat and radiotoxicity characteristics of nuclear waste are closely related to increases in uranium utilisation. The amount of mass requiring a long-term disposal path is tightly coupled to the quantity of actinides in the "highly radioactive" waste stream. Here we purposely avoid the term "high-level waste" as to avoid any implication that we know the legal definitions of this nomenclature going into the future. In this paper we use "highly radioactive" to mean dispositioned spent nuclear fuel and separation losses of used nuclear fuel. Complete uranium utilisation by definition means that zero actinide mass is present in the highly radioactive waste stream; thus producing a fission product only waste stream. Therefore, fuel cycles with complete (uranium and transuranic) recycle discharge predominately fission products with some actinide process losses. Fuel cycles without complete recycle discharge a much more massive and radiotoxic waste stream because only a fraction of the initial actinide mass is burned prior to disposal.

Removal of transuranics from the fuel cycle into waste also constitutes poor fissile conservation. Thus, poor transuranic separation efficiency results in both decreased uranium utilisation and increased radiotoxic legacy. This paper contrasts a range of fuel cycle concepts to ascertain which combinations of reactor, fuel and recycle technologies use uranium more efficiently with less radiotoxic consequences.

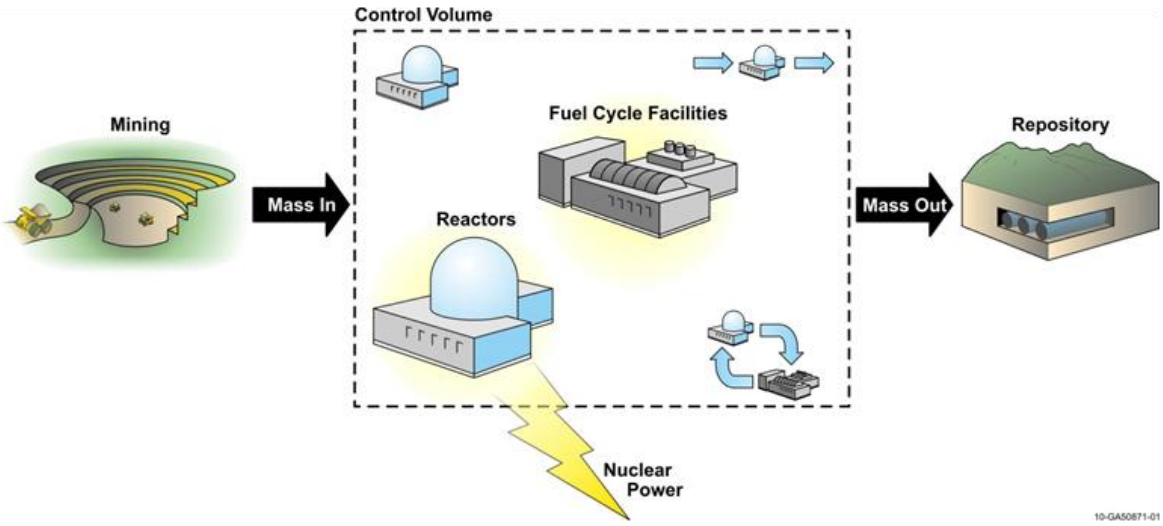
Uranium utilisation

The resource utilisation efficiency (%) is defined as the fraction of the original natural uranium ore that is converted into fission energy. This formulation of uranium utilisation is time-period and reactor independent as it assumes no credit for availability of current fissile stocks.

$$Uranium\ utilasation\,(\%) = \frac{\sum_{all\ reactors} Mass\ of\ heavy\ metal\ \ consumed\,by\ \ fission}{\sum_{all\ mines} Mass\ of\ natural uranium\ (or thorium) mined} \times 100 \qquad (1)$$

Generally, the above definition is simply mass utilised/mass consumed. However, the sums over mining and reactors emphasise uranium utilisation must be calculated for a fuel cycle, including all reactors included in that fuel cycle and all mining operations. Consider a static control volume (Figure 1) placed around a fuel cycle's entire enrichment, partitioning and transmutation infrastructure. In the control volume analogy, the numerator of Eq. (1) is defined for all nuclear energy facilities inside the control volume. Similarly, the denominator describes the material input to the control volume. The generating capacity of the set of nuclear energy facilities within the control volume (sometimes called a nuclear park) is considered static. This means that to the extent feasible in a fuel cycle option, there is no net growth or decrease in the overall generating capacity, or in fuel material storage, etc. It should be noted that in reality fuel

Figure 1: Control volume analogy used in calculating resource utilisation



cycles are dynamic instead of static, as new nuclear energy facilities are brought on or taken off-line continuously. However, the static analyses to follow are useful in gauging how current and future fuel cycles will perform given a set of fixed presumption about material flows.

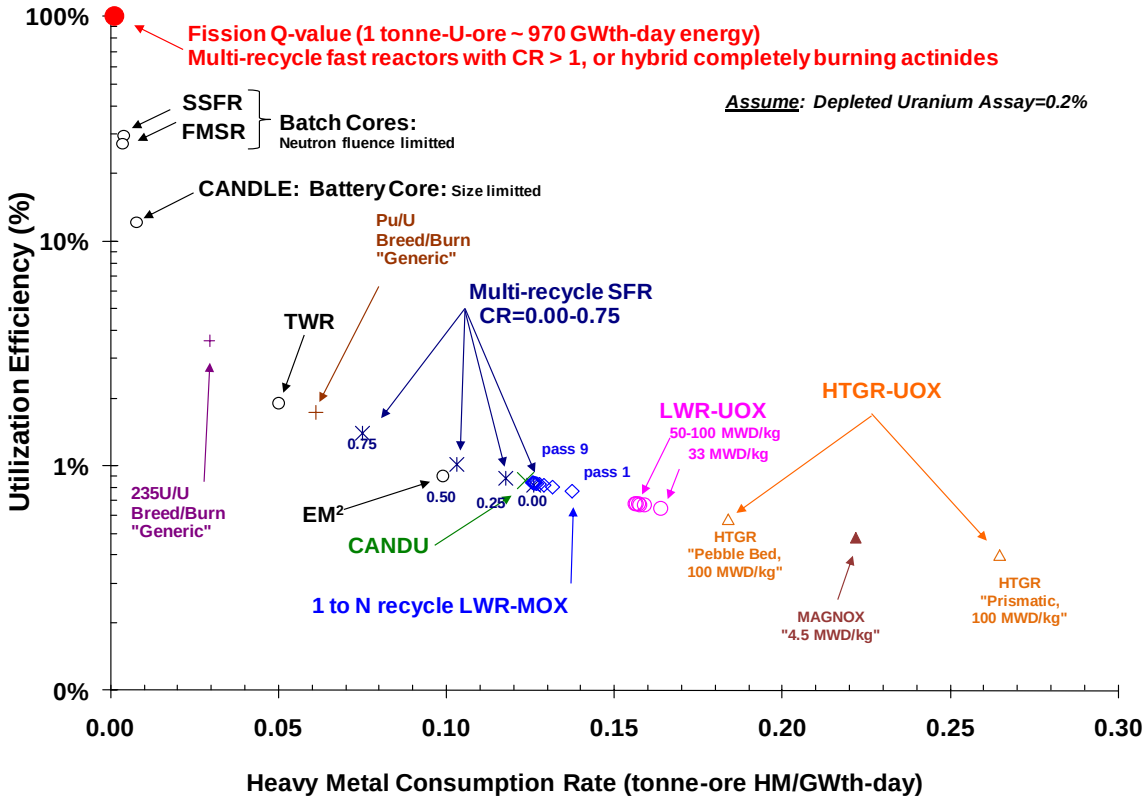
For once-through fuel cycles where no uranium enrichment is used to concentrate ^{235}U, the utilisation efficiency is equivalent to the fraction of initial heavy metal atoms that underwent fission during the single time in the reactor; essentially the burn-up. Also, for such a fuel cycle, the rate of uranium resource consumption (tonne-HM per GWth-day) is inversely proportional to uranium utilisation (GWth-day/tonne of natural uranium). For a closed fuel cycle with no uranium or transuranics discharged as waste, the only mass entering the control volume is uranium. Thus for a perfectly closed fuel cycle, the ore consumption rate perfectly matches the Q-value limited fission rate giving 100% uranium utilisation.

The uranium utilisation efficiency is plotted against the rate of heavy metal consumption (taken at the point where it is mined) per unit fission energy liberated (Figure 2). Perfect utilisation efficiency (100%) corresponds to no heavy metal mass leaving the control volume. The only mass leaving the fuel cycle in this situation are fission products. The absolute minimum ore consumption, occurring at 100% utilisation efficiency, is directly proportional to the energy released per fission (~200 MeV/fission). Converting units, this becomes ~950 GWth-day per tonne of uranium or thorium fissioned.

Fuel cycles that do not require uranium enrichment such as heavy water reactors (i.e. CANDU) and graphite-moderated MAGNOX reactors in Figure 2 exhibit low utilisation [1,2]. One original motivation for non-enrichment reactor technologies was plutonium production directly from natural uranium. However, this does not necessarily imply a plutonium self-sustaining fuel cycle because the production of plutonium is far lower than the consumption of the ^{235}U in the natural uranium fuel. It is also important to note that high-temperature gas reactor (HTGR) technologies evaluated had smaller uranium utilisation than that of light water reactors (LWR) [3-5]. This is because the HTGR designs that we analysed did not yield as much fuel burn-up per initial uranium enrichment invested. Therefore, despite their higher burn-up than the LWR cases, the uranium utilisation was still less because of the depleted uranium created during enrichment.

The reactors with on-line refuelling strategies, CANDU and pebble bed HTGR, have greater uranium utilisation compared to similar reactor types with a batch process, i.e. MAGNOX and prismatic HTGR. For example, CANDU reactors achieve nearly twice the burn-up as MAGNOX reactors though both reactors begin with natural uranium. Also, pebble bed HTGR achieves approximately the same burn-up as prismatic HTGR, but only requires two-thirds the initial ^{235}U/U enrichment and therefore have higher natural uranium utilisation. This is because the

Figure 2: Uranium resource utilisation efficiency as a function of consumed natural uranium "mass-in"[1]



excess reactivity of fresh fuel is better matched with reactivity-deficient fuel that is nearly spent. Thus, more excess neutrons that otherwise would need to be absorbed by control mechanisms such as control rods, are instead invested into fuel, thus extending their burn-up.

All of the once-through enriched UO_2 fuel cycles (i.e. current LWR fuel cycle) exhibit utilisation in the range of 0.6 to 0.8%, regardless of their discharge burn-up [5]. Higher burn-up is offset by higher uranium enrichment, which produces more front-end depleted uranium. The higher burn-up in these systems is not attained via higher conversion of ^{238}U to fissile ^{239}Pu. Here again, burn-up (and hence uranium utilisation) can be slightly extended by increasing the number of batches that reside in the core at any given time for the same reason that on-line refuelling extends burn-up as explained above. For LWR recycle scenarios such as mixed-oxide (MOX), uranium utilisation generally increases with the first recycling and then subsequently either increases or decreases slightly with subsequent recycles as they continue to require uranium fissile support (uranium enrichment) to counter the effect of plutonium fissile degradation [6]. The added enrichment for the subsequent reactor passes creates further depleted uranium, thus decreasing utilisation.

The sodium-cooled fast reactor (SFR) cases in Figure 2 are transuranic burners with fissile conversion ratios less than unity [7]. Because, these reactors purposely destroy the fissile material (namely ^{239}Pu) generated by a supporting fleet of LWR, the separative work performed during enrichment as well as the neutronic investment made in creating the plutonium during LWR irradiation is purposely destroyed in the SFR. Intentionally burning transuranic wastes has

1. For the MOX-UE cases shown in this paper, it was assumed that no recycled uranium is recovered for the purpose of re-enrichment and use in subsequent cycles. However, the concentration of ^{235}U in recycled UOX uranium is an interesting possibility for increasing uranium utilisation.

obvious waste minimisation advantages as will be discussed in the next section. However, without sustainably propagating investments by the fuel cycle to create and concentrate fissile material, the uranium utilisation will be significantly penalised.

In general, uranium utilisation increases the more the fuel can be burned without reliance on uranium enrichment. Two generalised fast reactor breed-and-burn approaches are given that approximate the ideal maximum burn-up achievable starting with the lowest possible enrichment (^{235}U/U or Pu/HM) required to achieve first criticality in the reactor. This maximum burn-up corresponds to the time in the irradiation when positive reactivity of the fuel is balanced by the negative reactivity drag of fertile isotopes and fission products. The calculated maximum burn-up for this simple zero-dimensional calculation yielded 35-40% of all initial heavy metal destroyed. The reason that the "Pu/U Breed/Burner" has a smaller utilisation than the "^{235}U/U Breed/Burner" point (see Figure 2) is that starting with an initial fissile load of Pu requires that it first be generated in a previous reactor, assumed to be an LWR in this case. The unused depleted uranium created during fuel enrichment in the LWR causes the utilisation to go down.

The extent of this depleted uranium wastage is the dominating factor in differences between the entire breed-and-burn concept technologies plotted in Figure 2. The General Atomics Energy Multiplier Module (EM^2) concept that we analysed is a plutonium-neutral concept [8]. That is, the reactor does not make any more or less plutonium than is fed into the core as fresh fuel, thus it is effectively a fissile converter or break-even system. However, the concept is intended to be operated without sustained plutonium recycle. Without subsequent recycling, any reactor option using plutonium as the initiating fissile source is not afforded the ability to burn stored depleted (or recycle recovered) uranium that was amassed in an earlier prerequisite phase of the fuel cycle.

A similar situation exists for the Constant Axial shape of Neutron Flux, Nuclide Density and Power Shape During Life of Energy Production (CANDLE) and the TerraPower Traveling Wave Reactor (TWR) [8]. The current version of these reactor concepts (at the time of this writing), assumes a lifetime core of enriched uranium with no sustained recycle of the irradiated fuel. Therefore, the achievable uranium utilisation is dictated by the volume of core that can be set aside to accommodate incorporation of depleted uranium. Even if all the depleted uranium can be incorporated into the reactor volume, or if depleted uranium were fed into the reactor in a batch process, the uranium utilisation then becomes limited by the attainable single reactor-pass burn-up. This is the case for the Sustainable Sodium-cooled Fast Reactor (SSFR) and Fast Mixed-Spectrum Reactor (FMSR) that we analysed [8]. In these two concepts depleted uranium assemblies are loaded in a large outer radial blanket to achieve initial breeding and then gradually shuffled inboard towards the chain reaction driving active region which then burns the fissile investment. In the FMSR case, the blanket region is moderated. In the SSFR case, the blanket is un-moderated.

One possible means of extending the uranium utilisation without using full recycle is the use of limited separation technologies. This class of recycle technologies typically capitalise on oxidation and reduction reactions to extract volatile fission products, thus partially cleansing the fuel of fission product neutron poisons. This partial cleansing in turn enables the chain reaction to be restarted for subsequent irradiations. Thus, if limited separation recycle concepts are used it can be assumed that the uranium utilisation can further be extended. However, 100% uranium utilisation cannot be achieved with limited separations due to the eventual squelching of available reactivity in the fuel to restart the chain reaction caused by accumulated non-separated non-volatile fission products.

Radioactive waste

Radioactive materials are generated in essentially all parts of a nuclear fuel cycle. If they are not re-used, they become wastes that need to be managed and disposed safely. The analyses reported here focuses on used nuclear fuel assemblies that contain unutilised uranium, transuranics and fission products associated with fuel that has been irradiated (i.e. the fuel meat associated with a

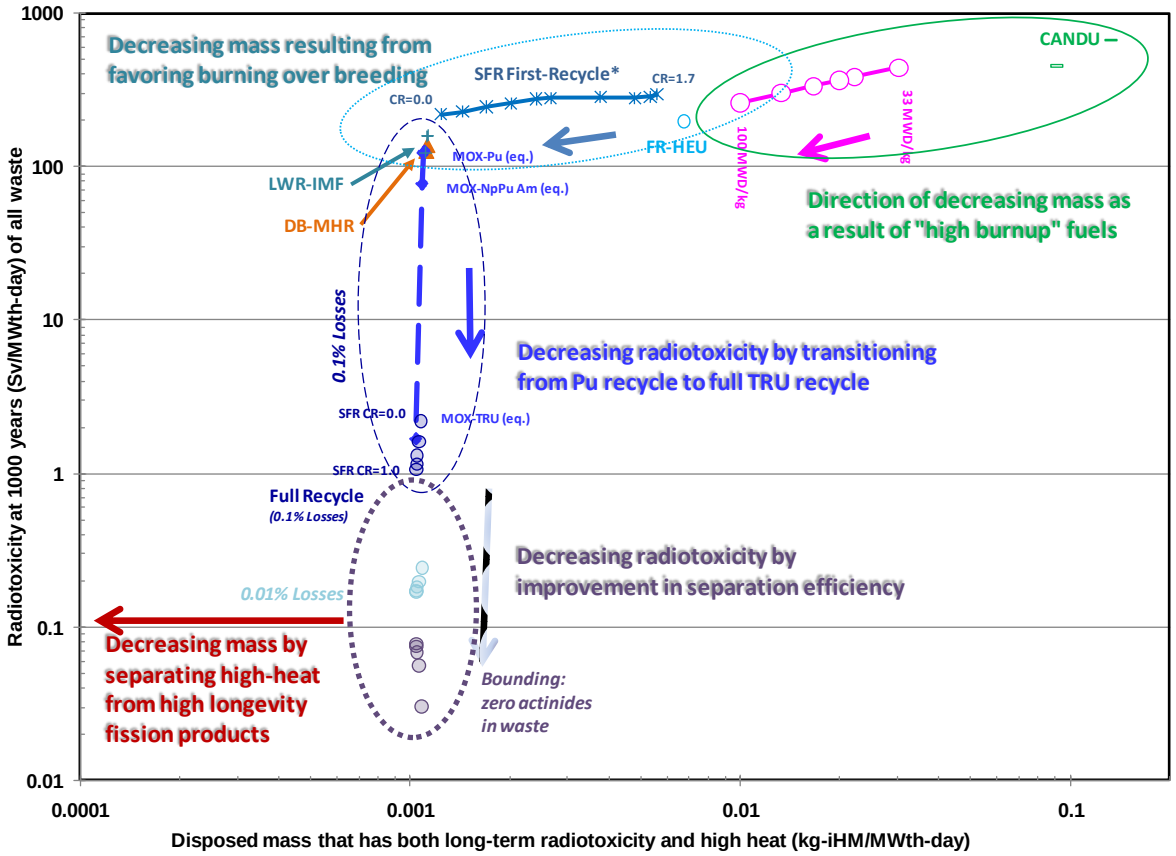
pellet or kernel). In other words, depleted uranium and separated uranium mass outputs from reprocessing are not considered a "highly radioactive" waste stream for the purpose of the following analyses.

Depending on handling and disposal approach, disposal of radioactive waste can be limited by three factors:

- Short and long-lived decay heat, to avoid overheating the waste form, the waste package, the structures that permit near-term retrievability. Overheating culminates in rock temperatures that could change the ability of the system to isolate the waste or changing the flow of water through the rock.
- Radiotoxicity, which is the source term for the dose that the repository system is designed to control.
- Waste mass and volume.

The following discussion and Figure 3 describes the relationship between: *i*) the radiotoxicity at 1 000 years after reactor discharge; *ii*) the amount of mass that is both high long-lived radiotoxicity and high heat, i.e. lacking disposal precedents. Both the mass and the radiotoxicity of the disposed material are normalised on a thermal power generation basis. (The trends are very similar if instead one graphs at 10 000 years. Analogous graphs at 100 000 or 1 000 000 years would show increasing impact of whether uranium is consumed or not.)

Figure 3:Long-lived radiotoxicity (taken at 1 000 years after irradiation) per mass of disposed actinide and fission product constituents of used nuclear fuel



As burn-up increases, the amount and the radiotoxicity at 1 000 years of disposed waste decreases. For example, the green oval (top right) contains all the once-through options. As burn-up increases from 7.5 to 150 MWth-day/kg-iHM across a range of reactor types (CANDU, LWR, HTGR, SFR), the disposed waste mass (which, normalised to thermal energy production, is proportional to 1/burn-up) decreases, and radiotoxicity decreases slightly [1,3-5,7].

The mass of disposed waste further decreases when used fuel is recycled at least once. The light blue oval (top centre) contains all the single recycle options, when all fission products separated from reprocessed fuel are disposed. The reduction of disposed mass is minimal for cases with high transuranic conversion ratio (i.e. high uranium content fuel, thus high uranium content disposed fuel). The reduction of disposed mass is an order of magnitude lower for low transuranic conversion ratios such as for IMF-type fuels (i.e. HTGR deep burn or LWR-IMF concepts).

Full recycle cases in the dark-blue oval (centre) can lower the radiotoxicity of the disposed waste by about two orders of magnitude when the transuranic isotopes are recycled. The full extent of the decrease in radiotoxicity is achieved only when all transuranic isotopes are recycled, rather than only Pu. The purple oval (bottom centre) contains all the full recycle options in which all the transuranics are recycled, and only the fission products separated during used fuel reprocessing are disposed.

The stair-step shape of Figure 3 stems from the fact that at the point where nearly all of the transuranics are recycled, the only truly highly radioactive waste stream is dominated by the mass of fission products and the long-lived radiotoxicity associated with transuranics. Going back to the control volume analogy, if fission products are the only waste being produced in "mass-out", then the waste production rate per unit energy is the inverse of the Q-value. Thus, if 950 GWth-day is generated per one tonne of uranium consumed then 1/950~0.001 tonnes of fission products are produced per GWth-day. This is the break-over point of the stair-step in Figure 3.

At this point further decreases in the "mass" dimension would require isolating low-heat with high, long-lived radiotoxicity isotopes from low-heat with low, long-lived radiotoxicity isotopes. Such hybrid scenarios would require matching waste characteristics with the repository characteristics. Another consideration is that some low-heat, low-radiotoxicity elements could have economic value in future metal markets; and some other isotopes may have economic value in other markets; thus not requiring disposal. Still other options include isotopic dilution of long-lived radiotoxic fission products with stable isotopes of the same element prior to disposal. These options are considered transformational disposal options but their consideration would greatly expand the fuel cycle option space beyond the scope of this report.

Conclusions

The analyses summarised here provide an insightful contrast of uranium utilisation and waste impacts for a wide array of nuclear fuel cycle options. Uranium utilisation is principally dictated by the method of generating and preserving fissile inventories in the fuel cycle. Less than 1% of all natural uranium is the fissile isotope, ^{235}U. Generally this requires that some portion of the fertile ^{238}U be immediately discarded in the beginning phase of the fuel cycle in order to concentrate ^{235}U enrichment step or ^{239}Pu during a first-recycle step. If uranium enrichment is used, much of the ^{238}U is discarded (temporarily or permanently) as depleted uranium. If a natural uranium fuel technology is used as the starting phase, such as in CANDU or MAGNOX, ^{238}U must be removed from the used nuclear fuel (at least temporarily) in order to concentrate the bred plutonium. Without at least some limited recycling capacity, this un-utilised ^{238}U forces the uranium resource utilisation to be less than 1%. Even in fast spectrum breed-and-burn concepts (i.e. very high burn-up with *in situ* consumption of bred material), the uranium utilisation is much less than 100%. The only fuel cycle scenario that can fundamentally reach 100% uranium utilisation is a fuel cycle employing fast reactors in a plutonium break-even or breeder mode in conjunction with multi-recycling. This is because of the abundance of extra neutrons that these reactors can use for converting ^{238}U into fissile plutonium. Full recycle of both transuranics and uranium is necessary because it allows the reactor fuel to be continuously

purged of fission product neutron poisons thus allowing an infinite number of opportunities for the ^{238}U to be converted into fissile plutonium and ultimately burned. At this point all natural uranium consumed from the earth is converted into fission products giving the maximum amount of fission energy that can be liberated from the mined uranium resource, equivalently the fission reaction Q-value (i.e. ~950 GWth-day per tonne of natural uranium).

The mass of highly radioactive nuclear waste, requiring a long-term disposal strategy, is also tightly dependent on the recycle strategy. For once-through fuel cycles increases in burn-up equate to decreases in the mass requiring disposal. However, for fuel cycles involving recycling, the amount of mass requiring disposal can be further deceased by reprocessing at least a limited number of times. This has the effect of removing much of the low radiotoxic used fuel uranium from the disposition destined transuranics and actinides. In full recycle strategies, even for reactors that are not breeders, the long-lived radiotoxicity contribution from the transuranics is kept in the fuel cycle, thus greatly reducing both radiotoxicity and mass of the disposal stream. Ultimately, the minimum mass requiring disposal is limited by the mass of fission products produced per unit of fission energy, equivalently the inverse of the nuclear reaction Q-value (i.e. ~0.001 tonne fission products per GWth-day).

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