Implications of fast reactor transuranic conversion ratio

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Abstract

Theoretically, the transuranic conversion ratio (CR), i.e. the transuranic production divided by transuranic destruction, in a fast reactor can range from near zero to about 1.9, which is the average neutron yield from ^{239}Pu minus 1. In practice, the possible range will be somewhat less. We have studied the implications of transuranic conversion ratio of 0.0 to 1.7 using the fresh and discharge fuel compositions calculated elsewhere. The corresponding fissile breeding ratio ranges from 0.2 to 1.6.

The cases below CR = 1 ("burners") do not have blankets; the cases above CR = 1 ("breeders") have breeding blankets. The burn-up was allowed to float while holding the maximum fluence to the cladding constant.

We graph the fuel burn-up and composition. As a function of transuranic conversion ratio, we calculate and graph the heat, gamma, and neutron emission of fresh fuel; whether the material is "attractive" for direct weapon use using published criteria; the uranium utilisation and rate of consumption of natural uranium; and the long-term radiotoxicity after fuel discharge.

For context, other cases and analyses are included, primarily once-through light water reactor (LWR) uranium oxide fuel at 51 MWth-day/kg-iHM burn-up (UOX-51).

For CR < 1, the heat, gamma and neutron emission increase as material is recycled. The uranium utilisation is at or below 1%, just as it is in thermal reactors, as both types of reactors require continuing fissile support.

For CR > 1, heat, gamma and neutron emission decrease with recycling. The uranium utilisation exceeds 1%, with the highest values achieved when all the transuranic elements are recycled.

At the system equilibrium, heat and gamma vary by somewhat over an order of magnitude as a function of CR. Isotopes that dominate heat and gamma emission are scattered throughout the actinide chain, so the modest impact of CR is unsurprising. Neutron emitters are preferentially found among the higher actinide isotopes, so the neutron emission varies much stronger with CR, about three orders of magnitude.

Introduction

Steadily for the better part of this decade, ANL has examined fast reactor transmutation scenarios with respect to the fuel cycle as a whole [1-3]. In particular, a parameter scan on the fast reactor transuranic (TRU) conversion ratio (CR) has been done from 0.00 to about 1.8, providing isotopic information that we use in this paper to show the impact of TRU CR on several parameters of interest in fuel cycle studies. The TRU CR is the production rate of TRU divided by the destruction rate (fission). For fast reactors, it is numerically close to the fissile breeding ratio, which is the production rate of fissile isotopes divided by the destruction rate. (The fissile breeding destruction rate includes both fission and neutron capture that converts a fissile isotope to fertile.)

The fast reactor design concept in these parameter scans is kept constant. In particular, the neutron fluence to the fast reactor fuel cladding is kept constant. The CR is changed by altering the ratio of TRU:U and other adjustments as appropriate. For CR < 1 (burner reactors), primarily the fuel pin size is changed; there is a single core zone. For CR > 1 (breeder reactors), there is a driver core surrounded by a blanket zone; the thickness of the blanket zone increases as the CR increases.

The burner parameter scan included both oxide and metal fuel cases. The breeder parameter scan was constrained to only the metal fuel case because metal has higher heavy metal density than oxide and is therefore believed to be theoretically capable of achieving a higher TRU conversion ratio.

To study the impact of minor actinide recycle strategies, oxide-fuelled fast reactors were the focus because of the presumption that aqueous separation (for which separation of one TRU element from the others) would be used whereas the common presumption is that electrochemical separation (for which separation among TRU elements is difficult) would be used with metal fuel.

All cases start with transuranic material from separation of used light water reactor (LWR) uranium oxide fuel at 51 MWth-day/kg-iHM burn-up (LWR-UOX-51). For burner cases, this continuing supply of fresh TRU from LWR-UOX-51 continues indefinitely, each recycle. For breeder cases, this supply of TRU is only needed for the initial use. Thereafter, because CR > 1, there is sufficient recycled TRU to sustain recycling without fresh TRU.

Most of the results in this paper are at equilibrium recycle, i.e. when the recycled composition comes into equilibrium with the supply of outside material. For burner reactors, that supply is TRU from LWR-UOX-51. For breeder reactors, that supply is depleted uranium.

To capture radiological issues, particularly neutron source, reactor physics calculations taken to the end of the actinide chain, ^{252}Cf. Analysis elsewhere confirms that tracking isotopes to ^{252}Cf is sufficient to capture all isotope evolution and build-up effects as well as the corresponding influence on radiologic source terms [4].

Composition evolution

Figure 1 shows the impact of conversion ratio on fuel burn-up. In these calculations, the burn-up was allowed to float while holding the maximum fluence to the cladding constant. Since burn-up, by definition, is energy per heavy metal in fuel and because energy released and fluence scale together for a given composition, Figure 1 is really showing the impact of the composition change. Figure 2 shows that as conversion ratio increases there is more uranium for conversion into TRU. Since total energy from fission (i.e. via a constant fluence) is kept constant and ^{238}U fissions at a much lower rate, the burn-up at the same fluence necessarily decreases with higher conversion ratio and higher initial uranium mass.

Figure 1: Impact of fast reactor TRU CR on energy produced per mass of total initial HM

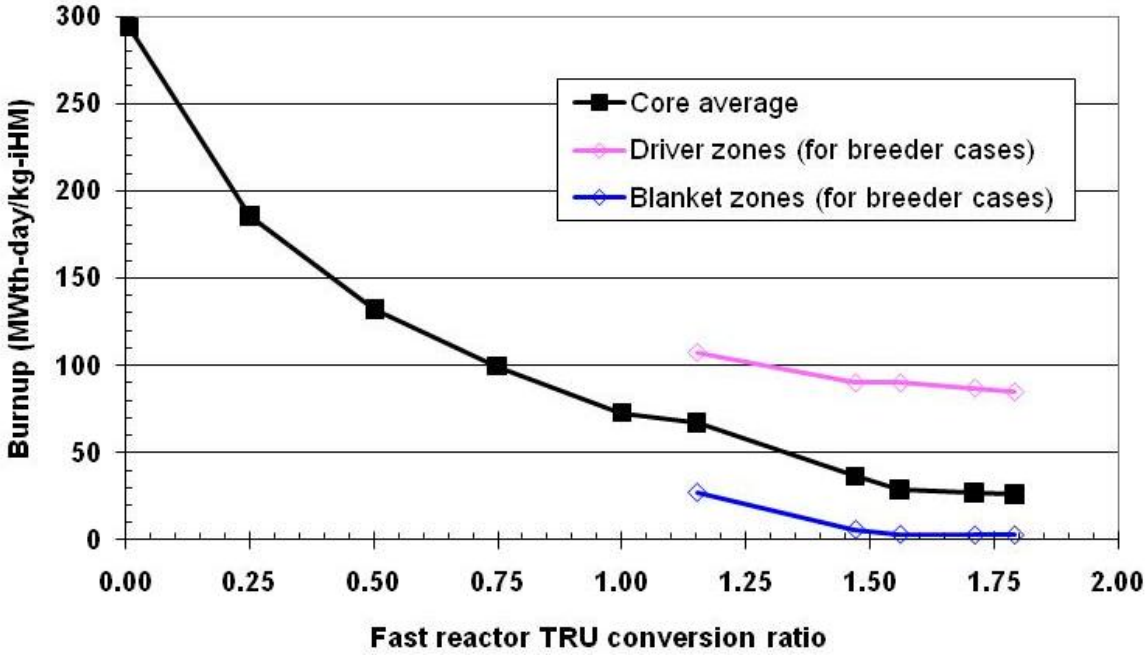


Figure 2: Impact of fast reactor TRU conversion ratio on initial HM composition of fresh fuel

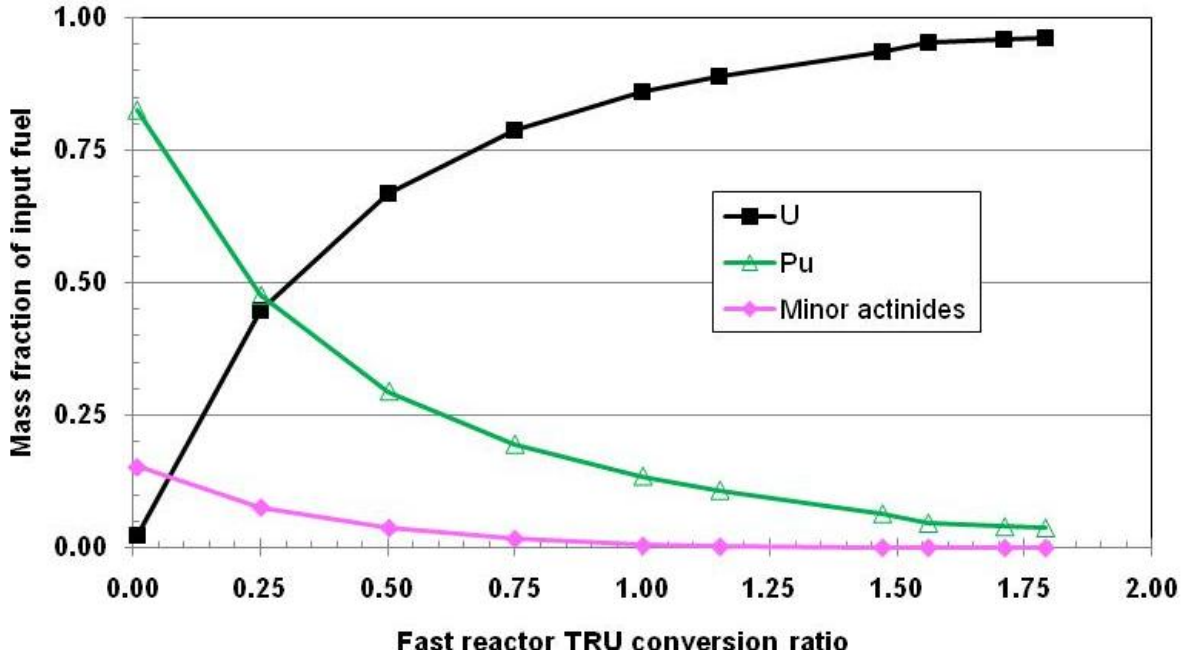
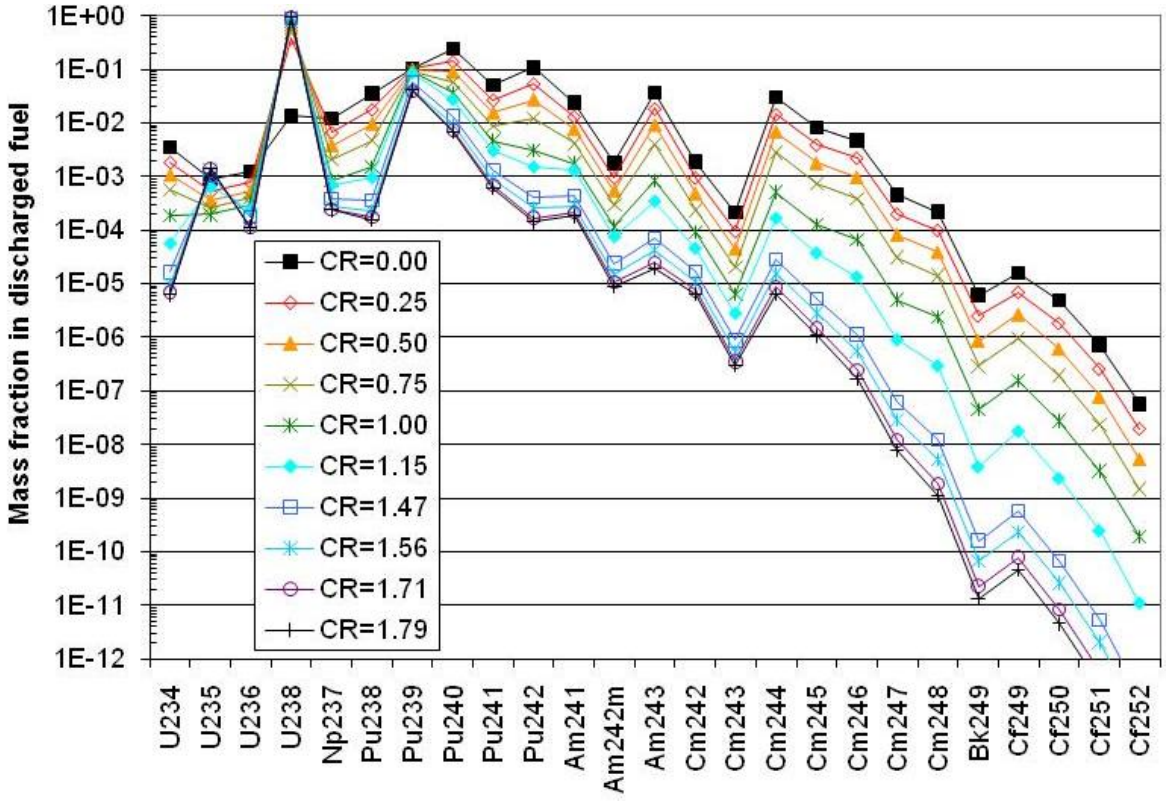


Figure 3 shows the discharged fuel composition as a function of conversion ratio at equilibrium recycle. All cases except CR~0.00 have high ^{238}U content and all cases have ~10% ^{239}Pu. Close inspection of Figure 2 and Figure 3 shows the ^{238}U concentration increases with CR. This increase is offset by the decrease in virtually all the other isotopes as CR increases. As CR increases, there is more ^{238}U, less ^{239}Pu, and less of the high-neutron emitting isotopes high on the actinide chain. Later subsections will illustrate how this changing composition impacts various parameters of interest.

Figure 3: Impact of fast reactor TRU CR on FR-metal fuel discharge equilibrium composition



Impact of actinide partitioning scenario

Figure 4 shows the impact of the actinide partitioning strategy on equilibrium composition – recycle Pu, NpPu, NpPuAm or all-TRU. For this parameter scan, we used an oxide fuel fast reactor with CR = 0.50. Changing from Pu recycle to NpPu recycle has little impact except for increasing the fraction of ^{237}Np and ^{238}Pu, which results from ^{237}Np from neutron capture by ^{237}Np. Changing from NpPu recycle to NpPuAm recycle changes the composition for isotopes ^{241}Am and above. The change from NpPu to NpPuAm is almost an order of magnitude from ^{244}Cm to ^{252}Cf.

Changing from NpPuAm recycle to all-TRU recycle means that isotopes of Cm, Bk, and Cf are also recycled. The two lines in the figure diverge at ^{244}Cm. ^{242}Cm and ^{243}Cm are fairly short-lived, and therefore they do not increase significantly if Cm itself is recycled. There is significant accumulation of the higher TRU isotopes with CmBkCf are recycled, increasing to 4-5 orders of magnitude for the Cf isotopes. These higher TRU isotopes are typically copious neutron emitters and therefore there is a marked increase in neutron emission, even in a fast reactor, when CmBkCf are recycled.

Heat, gamma and neutrons

Figures 5, 6 and 7 show the heat, gamma and neutron emission as a function of conversion ratio. The first recycle of TRU in the fast reactor is always shown with a dashed line. For the first recycle, the output (heat, gamma, neutron emission) is fairly constant as a function of TRU CR. The TRU:HM ratio and burn-up (fission energy/HM) vary similarly with TRU CR so that the ratio of TRU per energy is almost constant. Since the TRU composition for first recycle is constant (the TRU is always from LWR UOX 51), then the heat, gamma and neutron per fission energy is roughly constant for the first recycle.

Figure 4: Equilibrium fuel cycle isotopic concentration of FR-oxide (CR = 0.5) for four minor actinide partitioning strategies

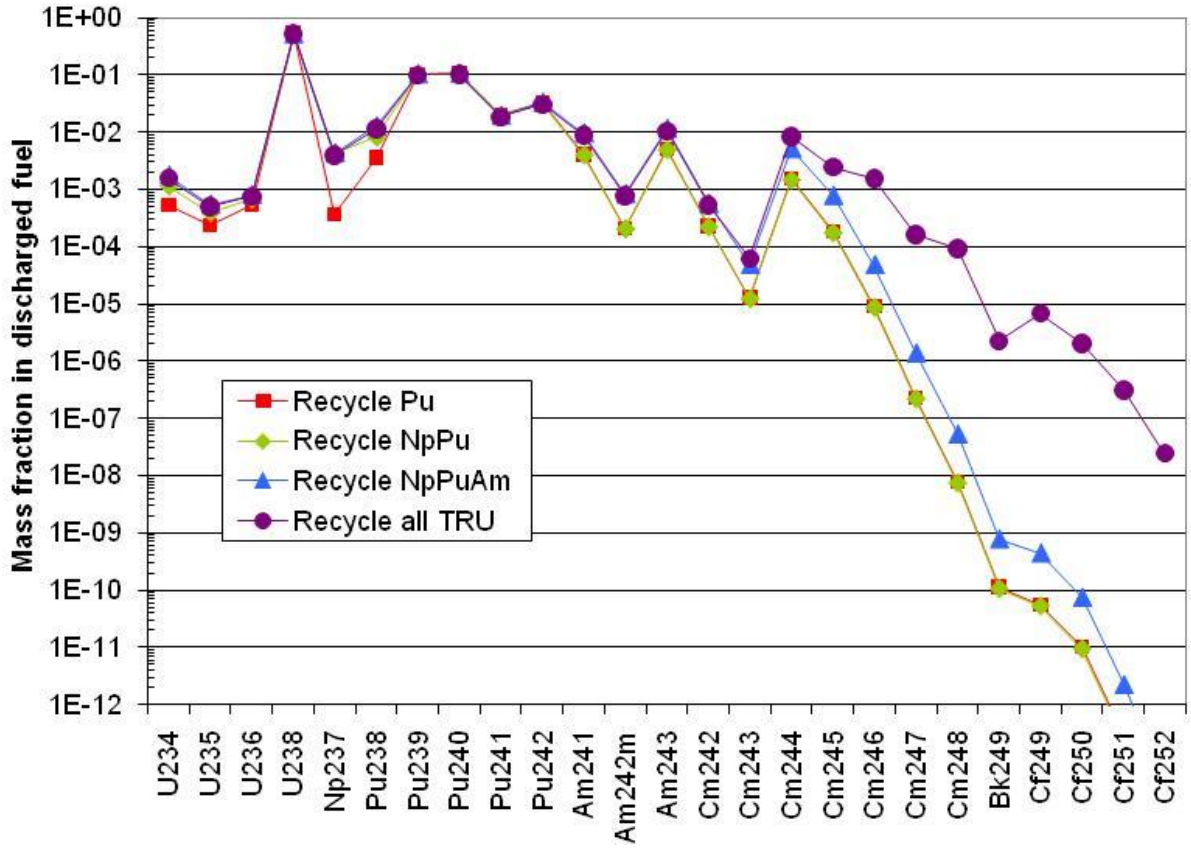


Figure 5: Heat from fresh fast reactor fuel as function of fast reactor TRU CR

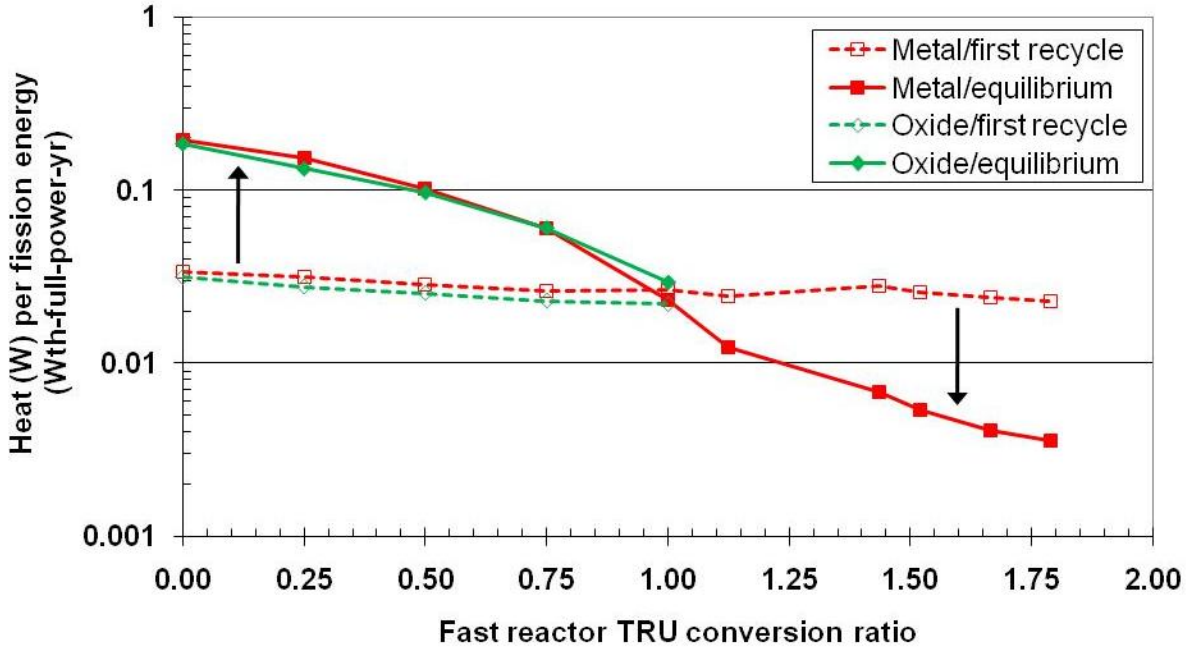


Figure 6: Gamma emission from fresh fast reactor fuel as function of fast reactor TRU CR

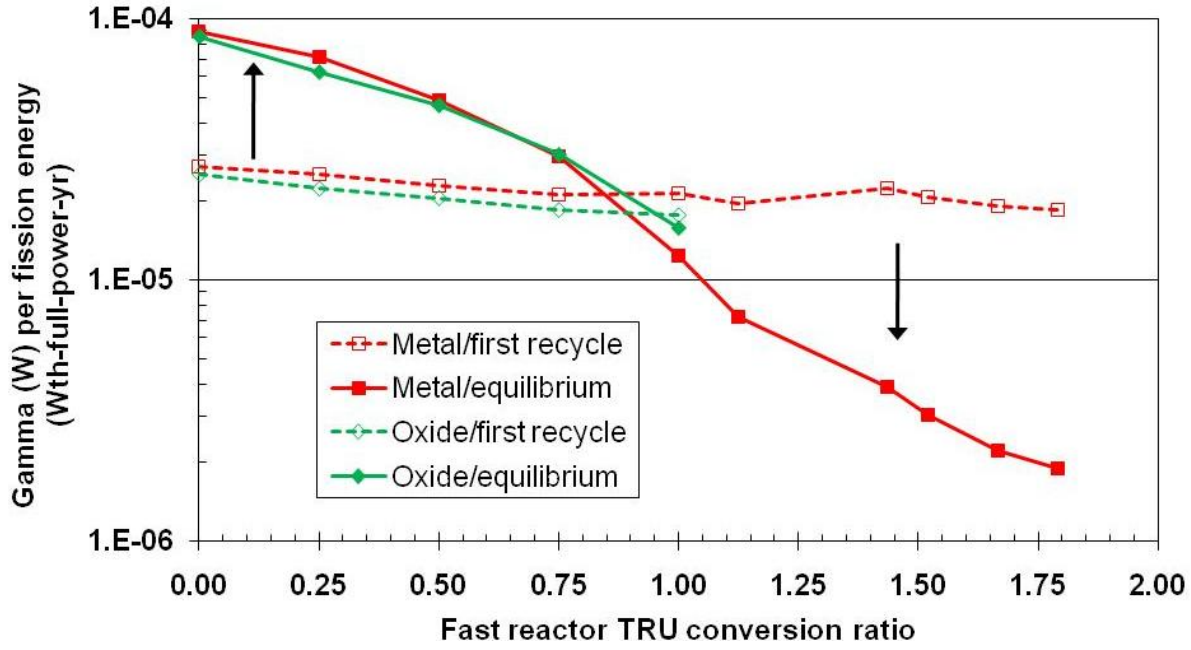
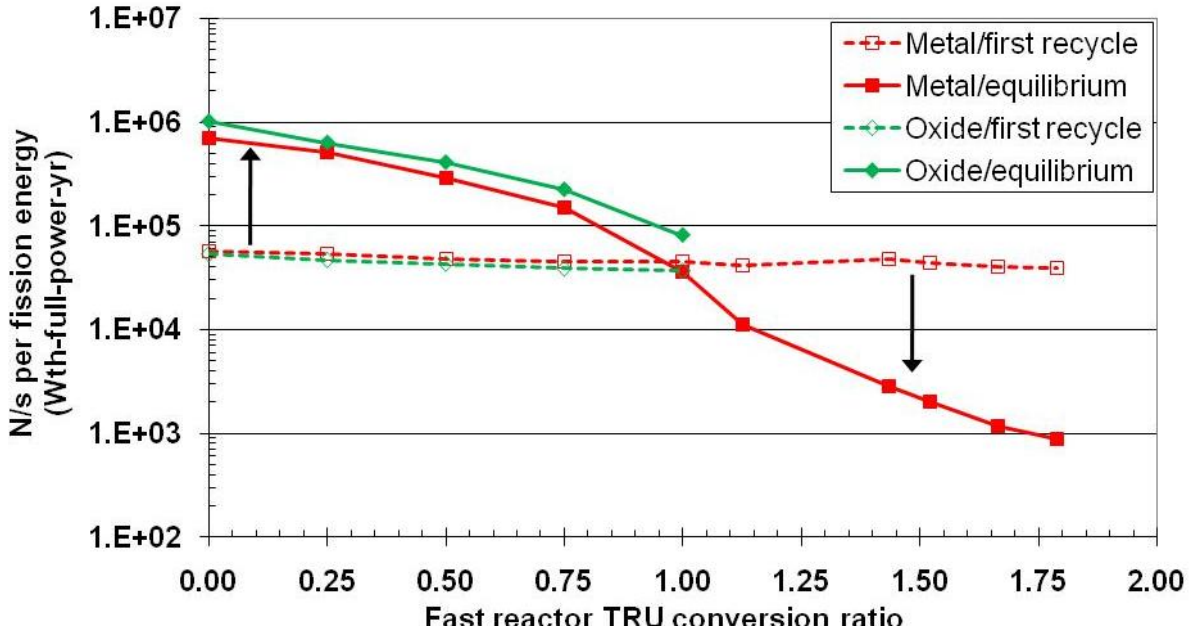


Figure 7: Neutron emission from fresh fast reactor fuel as function of fast reactor TRU CR



The equilibrium recycle composition is always shown with solid lines. The isotopic composition of recycled material changes during repeated recycles. For burner cases, the heat, gamma and neutron emission increase and eventually attain equilibrium, with the highest increase for the lowest CR. For burner cases, the heat, gamma and neutron emission decrease with the largest reduction for the highest CR. The higher the CR, the more the composition of relatively high heat, gamma and neutron emitting isotopes is diluted with ^{239}Pu bred from ^{238}U.

The figures demonstrate that assessment of fast reactors can change dramatically as a function of fast reactor CR. The most sensitive parameter is neutron emission, as it is the one most impacted by the isotopes at the top part of the actinide chain. That is, the highest heat and gamma emitting isotopes are scattered throughout the actinide chain. However, the highest neutron emitting isotopes are preferentially at the top of the chain, e.g. ^{252}Cf, and therefore neutron emission is the parameter most impacted as the fuel composition changes from that of LWR UOX-51.

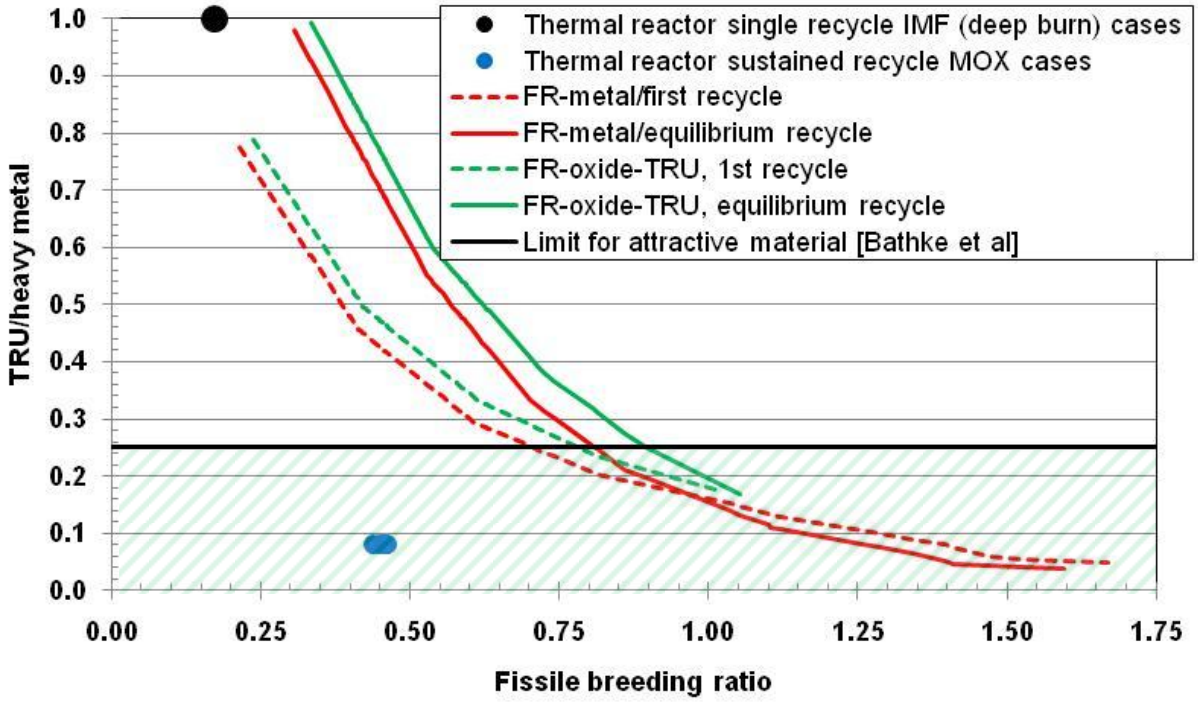
Attractive material

Assessment methodologies of proliferation resistance and physical protection include many parameters. One of these is whether fresh fuel is directly usable in a weapon. It is well known that a fresh uranium fuel composition with ^{235}U enrichment below 20% is not considered directly weapons-usable. For ^{233}U, this threshold is 12%. The two uranium isotopes can be combined with the expression of (1.67 ^{233}U + ^{235}U) < 20%.

There is less agreement on the thresholds for Pu and TRU-based fuels. The most recent study that directly addressed this question defined two figures of merit (FOM), one for national groups and one for subnational groups [5]. They find that the TRU fraction of heavy metal (TRU/HM) must be below 25% to keep material from being attractive for direct weapon use.

Figure 8 shows the composition of various cases thermal reactor cases (LWR and HTGR) with a single recycle of inert matrix fuel (IMF), called "deep burn" in the HTGR community, use a fuel that is initially all TRU. LWR with mixed-oxide (MOX) of TRU and uranium use fuel that is below 10% TRU. (We are unaware of studies of HTGR analogous to LWR-MOX.) From the reactor physics standpoint, these LWR-MOX cases can be sustainably recycled [4,6,7] provided that there is continuing fresh TRU support, just as in the case of burner fast reactors.

Figure 8: Enrichment required, TRU/HM; this is a topic addressed by the IAEA and all methodologies but the criteria for Pu and TRU vary

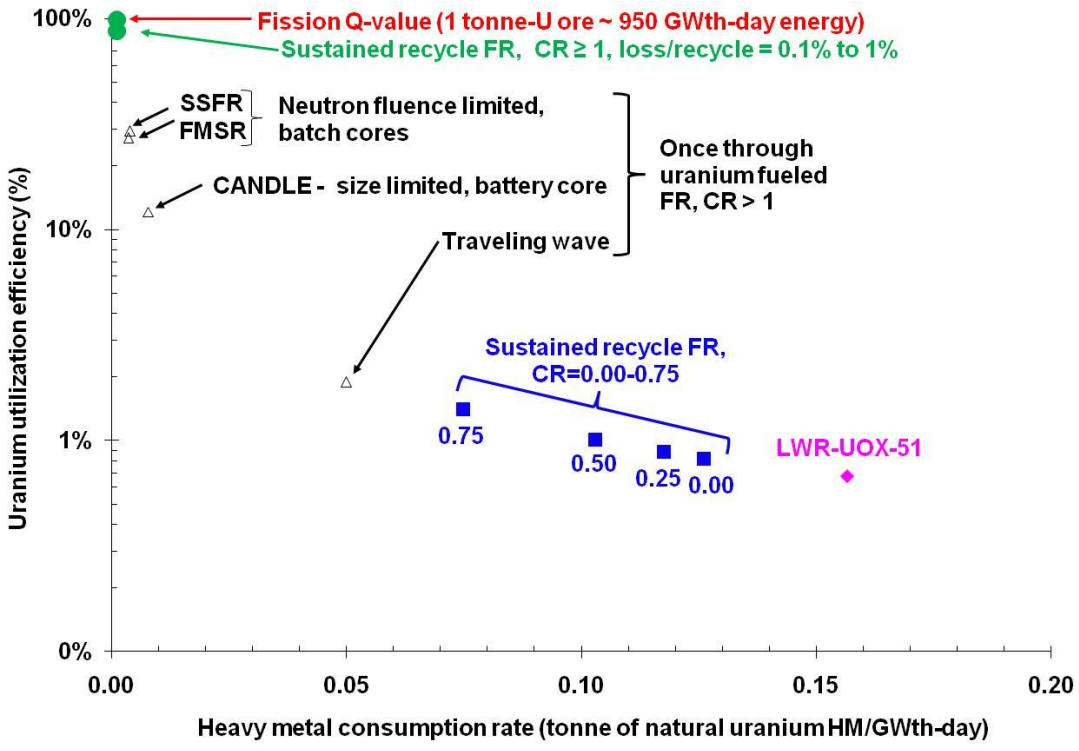


Because thermal reactors are included, the X-axis is the fissile breeding ratio, rather than the TRU conversion ratio as used in other graphs in this paper. Although the fissile breeding ratio and TRU CR are numerically similar for fast reactors, they diverge significantly for thermal reactors. In fact, for LWR UOX-51, although the fissile breeding ratio is under 1.0, its TRU CR averages over 2. For both thermal and fast reactors, the TRU/HM ratio decreases as the fissile breeding ratio increases. In thermal reactors, the TRU/HM is well under 0.25 for sustainable MOX recycle. In fast reactors, to attain TRU:HM below 0.25, the fissile breeding ratio (and TRU CR) must be at least 0.75-1.00. Using the criterion of Bathke, *et al.* [5] we find that fast breeder reactors do not use fresh fuel that is attractive for direct weapon use; the fertile uranium content is too high. Of course, if the TRU:U mixture is separated after diversion or theft, the separated TRU would be attractive.

Uranium utilisation

Figure 9 shows the uranium utilisation and consumption rate of natural uranium for various cases. The figure includes LWR UOX 51 for comparison. It and all other thermal reactors (even with sustained recycle) have uranium utilisation under 1% [8]; all such reactors have fissile breeding ratios under 1.0 and therefore remain dependent on a continuing supply of either fissile ^{235}U or fresh TRU from separation of used enriched-uranium fuel (such as used LWR-UOX-51). The same behaviour is seen in the figure for fast burner reactors (even with sustained recycle). For higher CR, they only slightly exceed 1%; they also require a continuing supply of ^{235}U or fresh TRU.

Figure 9: Uranium utilisation and consumption as function of TRU CR



To achieve uranium utilisation significantly well above 1% requires a fast breeder reactor. Recent studies of once-through uranium-fuelled fast breeder reactors show uranium utilisation as high as 30% [8,9]; these have been called "breed and burn" because they start with two zones, enriched ^{235}U and ^{238}U, the enriched ^{235}U is critical and provides neutrons to breed ^{239}Pu in the ^{238}U, which eventually achieves criticality. For this to happen without recycling, the fuel and its cladding must tolerate high neutron fluence and accumulation of fission products.

Fast reactors in which fuel is repeatedly recycled can approach 100% uranium utilisation, limited only by the processing losses of TRU material each recycle. Loss rates of 0.1% and 1.0%/recycle lead to uranium utilisation of 0.99 and 0.87 respectively. At CR = 1, the uranium utilisation can be estimated by (fractional burn-up during irradiation – processing loss)/(fractional burn-up during irradiation). If only Pu is recycled, rather than all TRU, the disposed TRU is effectively a processing loss and the uranium utilisation cannot exceed 0.9.

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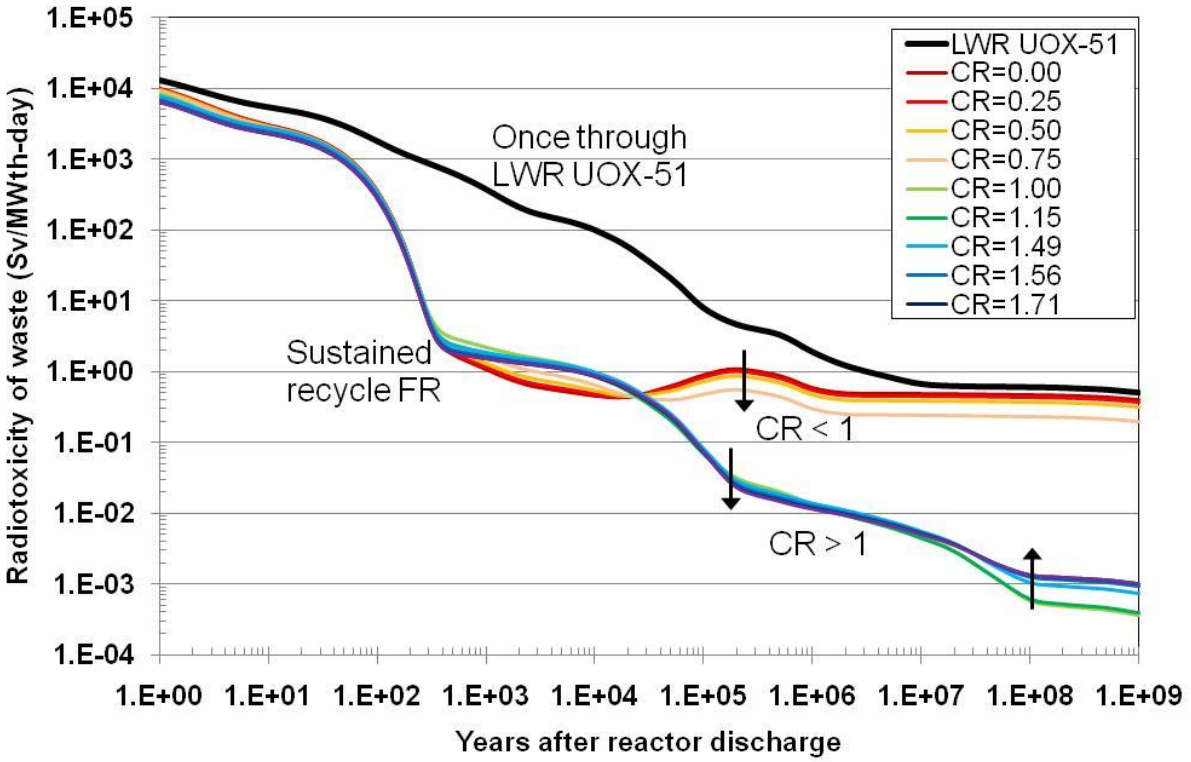
Long-term radiotoxicity

Figure 10 shows the radiotoxicity of waste material as a function of TRU CR. Used LWR UOX-51 is included for comparison. The FR cases are at equilibrium and include unused uranium recovered from separation of used LWR (if any), all fission products, and 0.1%/recycle of transuranic isotopes. The inhalation dose conversion factors from ICRP72 were used in the calculations. The burner cases and breeder cases diverge at longer times because the burner cases have excess uranium recovered from used LWR fuel and the breeder cases do not. This excess uranium slowly becomes more radiotoxic as its decay progeny isotopes build to equilibrium. (Depleted uranium is not included; if it were included, the breeder cases would not change. The LWR UOX-51 and burner cases would increase slightly at times over ~100 000 years. This would increase the observed differential for burners vs. breeders.)

Figure 10: Radiotoxicity of waste from sustained recycle as function of TRU CR



If processing losses are higher or if minor actinides are disposed, the radiotoxicity of the sustained recycle cases would increase.

Radiotoxicity of wastes is sometimes compared to the radiotoxicity of natural uranium ore that is mined to start the process. The radiotoxicity of natural uranium per mass is, of course, a constant. However, the amount of natural uranium per fission energy release (MWth-day) is not. Therefore, the radiotoxicity of the appropriate amount of natural ore would not be a single line in Figure 10. The lines would be several Sv/MWth-day/kg. Thus, the radiotoxicity of used LWR UOX-51 exceeds natural uranium until several hundreds of thousands of years. The radiotoxicity of waste from sustained FR recycle exceeds natural uranium only until approximately a thousand years.

Conclusions

For CR < 1, the heat, gamma and neutron emission increase as material is recycled. The uranium utilisation is at or below 1%, just as it is in thermal reactors as both types of reactors require continuing fissile support.

For CR > 1, heat, gamma and neutron emission decrease with recycling. The uranium utilisation exceeds 1%, especially as all the transuranic elements are recycled.

At the system equilibrium, heat and gamma vary by somewhat over an order of magnitude as a function of CR. Isotopes that dominate heat and gamma emission are scattered throughout the actinide chain, so the modest impact of CR is unsurprising. Neutron emitters are preferentially found among the higher actinides, so the neutron emission varies much stronger with CR, about three orders of magnitude.

The uranium utilisation is of course a strong function of CR.

It is generally imprudent to generalise the performance of breeders and burners. In several ways, recycling in burner fast reactors is more similar to recycling in thermal reactors, which also require continuing support of fresh fissile material, than it is to recycling in breeder fast reactors.

Acknowledgements

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