

Analysing losses: Transuranics into waste and fission products into recycled fuel

**Steven J. Piet, Nick R. Soelberg, Samuel E. Bays, Robert S. Cherry, Layne F. Pincock,
Eric L. Shaber, Melissa C. Teague, Gregory M. Teske, Kurt G. Vedros**

Idaho National Laboratory
Idaho Falls, ID, United States

Candido Pereira

Argonne National Laboratory
Argonne, IL, United States

Denia Djokić

University of California at Berkeley
Berkeley, CA, United States

Abstract

All mass streams from separations and fuel fabrication are products that must meet criteria. Those headed for disposal must meet waste acceptance criteria (WAC) for the eventual disposal sites corresponding to their waste classification. Those headed for reuse must meet fuel or target impurity limits. A “loss” is any material that ends up where it is undesired. The various types of losses are linked in the sense that as the loss of transuranic (TRU) material into waste is reduced; often the loss or carryover of waste into TRU or uranium is increased.

We have analysed four separation options and two fast reactor fuel fabrication options in a generic fuel cycle computer model. The separation options are aqueous uranium extraction plus (UREX+1), electrochemical, Atomics International reduction oxidation separation (AIROX), and melt refining. UREX+1 and electrochemical are traditional, full separation techniques. AIROX and melt refining are taken as examples of limited separations, also known as minimum fuel treatment. The fuels are oxide and metal. To define a generic fuel cycle, a fuel recycling loop is fed from used light water reactor (LWR) uranium oxide fuel (UOX) at 51 MWth-day/kg-iHM burn-up. The recycling loop uses a fast reactor with TRU conversion ratio (CR) of 0.50. Used fuel from the fast reactor is also recycled. Only waste, not used fuel, is disposed – unless the impurities in the recycled fuel accumulate to a level so that it is impossible to make new fuel for the fast reactor. Impurities accumulate in the recycled fuel as dictated by separation process removal efficiencies and fission product generation. Our model approximates adjustment to fast reactor fuel stream blending of TRU and U products to compensate for impurity accumulation. Our mass flow model ignores postulated fuel impurity limits; but we compare the calculated impurity values with those limits to identify elements of concern.

AIROX and melt refining cannot be used to separate used LWR UOX-51 because they cannot separate U from TRU. From the physics standpoint, AIROX and melt refining can serve in the recycle loop for about three recycles, at which point the accumulated impurities displace fertile uranium and the fuel can no longer be as critical as the original fast reactor fuel recipe. UREX+1 and electrochemical can serve in either capacity; key impurities appear to be lanthanides and several transition metals.

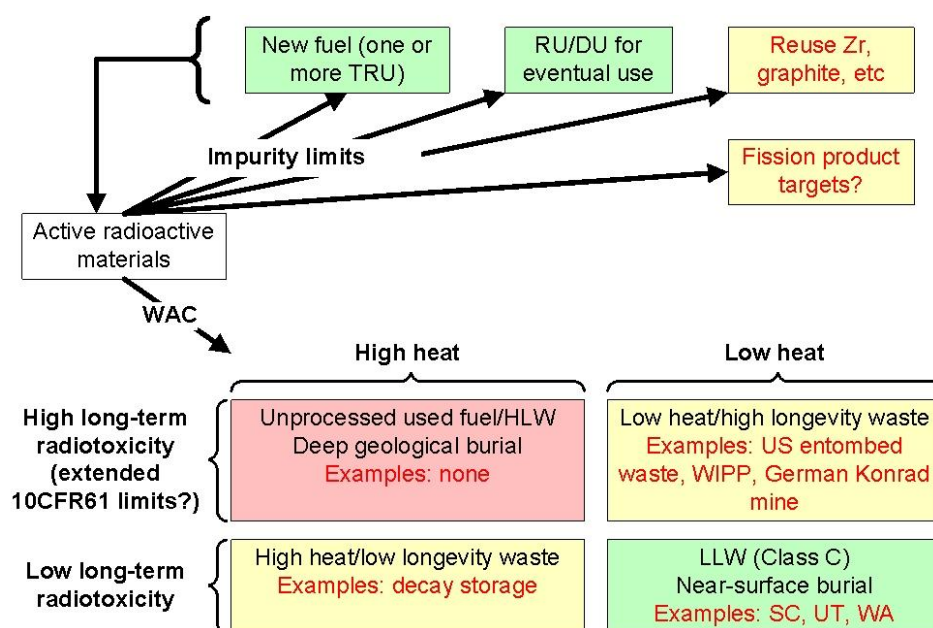
Introduction

The team aimed to understand the broad implications of changes of operating performance and parameters of a fuel cycle component on the entire fuel cycle. At this time, we have focused on a fuel cycle that is based on recycling used light water reactor (LWR) uranium oxide fuel. The recycling loop uses a fast reactor with TRU conversion ratio (CR) of 0.50. We documented the study of the impact of changing the loss of fission products into recycled fuel and the loss of actinides into waste streams [1].

When this effort started in spring 2009, the objective was “the number of nines” – how would the cost of separation, fuel fabrication, and waste management change as the number of nines of separation efficiency changed? The intent was to determine the optimum “losses” of TRU into waste for the single system that had been the focus of the Global Nuclear Energy Partnership (GNEP), namely sustained recycle in burner fast reactors, fed by transuranic (TRU) material recovered from used uranium oxide fuel (UOX at 51 MWth-day/kg-iHM burn-up) from LWR. That objective proved to be neither possible (insufficient details for the former GNEP options, change in national waste management strategy from a focus on Yucca Mountain) nor appropriate given the change to a science-based programme considering a wider range of options.

Figure 1 illustrates the central challenge of the discharge side of nuclear fuel cycles – how can radioactive materials “in service” be dispositioned? All mass must eventually leave active status, to either be readied for recycling or permanently disposed. These questions arise: What options exist for individual technologies, how should such options integrate to make a system, and when and for how long can materials be recycled? In short, what are the sagacious disposition options?

Figure 1: Central challenge of the downstream side of nuclear fuel cycles [2]



Our analyses are limited to commercial nuclear energy fuel cycles, uranium-transuranic element fuel cycles (as opposed to thorium-uranium fuel cycles), light water reactors and sodium-cooled fast reactors, and fast reactors that consume more TRU than they make, i.e. burner fast reactors as opposed to breeder fast reactors. AIROX and melt refining are examples of limited separation techniques that could support modified open cycles (limited number of recycles); UREX+1 and electrochemical separation could support either modified open or full, sustained recycle.

The rest of this paper pertains to five topics. Each pulls together information scattered throughout the full report [1] with quantitative analyses performed with our Fuel-cycle Integration and Trade-off (FIT) model [3]. These five topics are cross-cuts that pervade our results.

Topic 1: The R&D programme must improve chemical completeness and consistency

The first issue in this area can be seen via the periodic table. Often, the sources of chemical elements are organised horizontally in the periodic table. For example, the first row of transition metals contains most of the constituents of steels, a likely source of impurities. The second full row of the periodic table (with the second row of transition metals) contains most of the fission products, a known source of impurities. Furthermore, when fission products decay into each other, they move left or right in the periodic table.

The behaviours and constraints of chemical elements are often organised vertically in the periodic table. For example, the halogens' column (Group 17) have similar chemical properties and indeed the 2009 working Fuels Campaign limits on metal fuels [3] constrain the TRU feedstock's content of total halogens (F+Cl+Br+I) to 300-ppm total, without regard to the mix among those elements, because of fuel cladding corrosion. The corresponding halogen limits on oxide fuels constrain the TRU feedstock content of F+Cl to 60 ppm; no limit is given for Br or I.

In general, the Fuels Campaign working limits are often for groups of elements that have similar impact on fuel performance. But, many of these limits include elements with different origins (fission products and non-fission products). Therefore, one must eventually analyse all impurities together, regardless of source. For example, it would be non-conservative to allocate or "use up" all of an impurity limit on fission product-origin impurities if there are other potential impurities in that group. For metal fuels, this means one would presumably not want to allocate the entire 300 ppm-halogen limit to only the fission products Br and I, leaving no margin for F or Cl.

The second issue is the mismatch between those elements for which Fuels Campaign working limits exist versus those for which there is separation data. Typically, there are separation data for major fission products, but there are often not fuel impurity limits for those elements. Therefore, it is impossible to know if the separation efficiency is too tight or too loose for those elements. It would be incorrect to assume that a non-limited element is truly without limit. Br and I in oxide fuel TRU feedstock is an example; although they are not currently limited, we believe it is likely they should be included with the F and Cl limit. For example, with uncertainties of at least a factor of 2 (up and down), we estimate Pd at ~1 000 ppm in both types of fuels. Should there be a limit on Pd impurity? Also, Te appears to be an impurity in either oxide or metal fuel at hundreds of ppm, is this acceptable?

There are Fuels Campaign working limits for many elements for which there are little or no separation efficiency data. In some cases, there are limited data that have not yet been included in mass balances. For example, we find little or no separation data for first row transition metals in uranium-extraction (UREX+1), electrochemical, air reduction/oxidation (AIROX), or melt-refining, even though that row contains the major constituents of steel and nickel-based alloys (Cr, Mn, Fe, Co, Ni). Here, "UREX+1" refers to any of the process schemes that produce uranium and mixed-TRU streams for recycle.

Because fuel fabrication and performance are based on different chemical characteristics for oxide versus metal fuels, one cannot assume that elements of the same chemical group face comparable limitations in two different fuel types. In short, there are little or no separation data for several elements the Fuels Campaign limits; and no limits for several elements for which there are separation data.

The third issue is that chemical alteration to fuel and waste streams is not limited to separations. Most fuel fabrication methods include a feed conditioning step that involves dissolution or melting of the feedstock. This has to be considered in the proliferation risk assessments and the definition of "minimum fuel treatment." It also motivates integration of separation and fuel fabrication.

The definition and assessment of “minimum fuel treatment” must reflect both separation and subsequent fuel fabrication. For example, current TRISO fuel fabrication involves a dissolution step, apart from whether or not separation of used fuel is the source of the feed material. All fuel treatment methods (even minimum fuel treatment) have some chemistry associated with them, i.e. oxidation reactions to enable separation due to the variable volatilities among different chemical species in AIROX. “Chemistry” includes physical chemistry such as volatility as well as redox and solution chemistry.

Uranium enrichment alters chemistry. We define four types of uranium: depleted uranium (DU) left over from enrichment, natural uranium (NU), enriched uranium (EU) and recovered uranium (RU) from separations. DU and EU are considered extremely pure, as they are the result of uranium enrichment, in which the uranium was fluoridated into UF_6 gas. NU or RU will not be as pure. The differences in uranium isotopes among DU, NU, EU, and RU are naturally important. Of course, NU or RU could be purified to the same degree as DU or EU with corresponding increase in cost.

Finally, the reasons for working limits on impurities in oxide and metal fuel feedstock include fabrication difficulties, formation of oxides/intermetallics, attack on fuel cladding, displacement of fissile content, and neutron absorption. If the programme is to consider minimum fuel treatment options such as AIROX or melt refining, then higher levels of impurities will exist in the recycled fuel material (compared to impurity levels remaining after full separations), and solutions will have to be developed to resolve the impacts of those impurities on the recycled fuels.

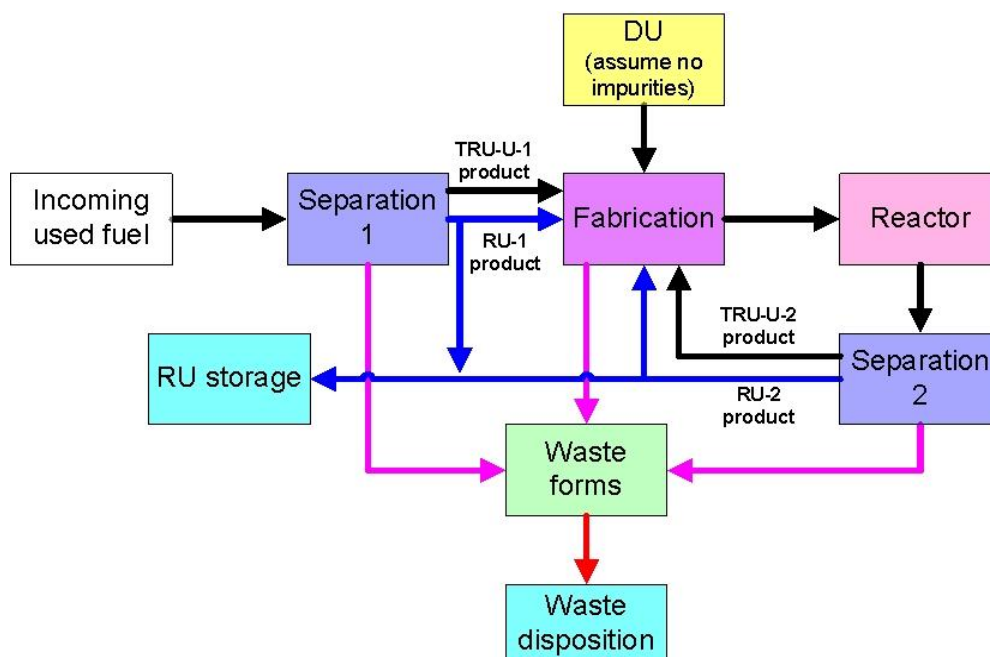
Topic 2: Do not forget the uranium

Figure 2 illustrates the basic mass flows. It is the simplest framework on which we could base our analysis. Several user-defined inputs were fixed for the initial analysis – on technologies and parameter ranges for which we had the most knowledge and for which individual parts of the calculation could best be validated. The incoming used fuel is always LWR UOX-51 in this study. Separation of used UOX-51 starts a recycle loop with a fast reactor, which is always a fast reactor with a transuranic conversion ratio of 0.50 in this report.

Separation-1 and/or Separation-2 may have both a TRU-U product and a recovered uranium (RU) product. Fundamental reactor physics indicates that when recycling, there is excess uranium if the transuranic conversion ratio is less than 1 (i.e. burner reactors). If instead the reactor in the recycle loop is a breeder, the incoming feed does not have to be used fuel, it can be any type of uranium; we did not consider breeder reactors in our quantitative analyses.

Said another way, for burner reactors, there is always excess uranium. Therefore, in general, either RU-1 or RU-2 (from Separation-1 and Separation-2, respectively) must be non-zero so that excess uranium can be extracted from the system, i.e. either Separation-1 or Separation-2 must provide some degree of separation of U from TRU. To use a separation technique that does not separate U from TRU (AIROX, melt refining, etc.), one of the following must be true.

1. The fuel is used in a reactor that requires less “octane” than where the TRU-U came from, e.g. used LWR is taken to the more neutron-efficient heavy water reactor (HWR).
2. A separate source of high “octane” enriched uranium or TRU is added to the TRU-U stream, requiring either high enriched uranium or some new source of TRU. This would be physically easy to do with melt refining; it is not so obvious how to do with AIROX.
3. It is a breeder reactor, in which case the incoming mass can be any uranium once in the “breeder” recycling loop. We mention this option for completeness; neither FIT nor our analyses to date have considered breeder reactors.

Figure 2: Basic mass flows

At the first iteration, there is no RU-2 because material has not yet come around the recycle loop. Thus, AIROX and melt refining cannot be used on used LWR UOX-51 fuel (and so cannot be used in Separation-1) if the product is intended for any reactor in which the TRU (fissile) content needs to be higher than the 1.3% TRU in used UOX-51, which includes all MOX or inert fuel matrix options in an LWR or HTGR, or any fast reactor previously studied. The only option would appear to be an HWR.

AIROX and melt refining can be used in the recycle loop until impurities accumulate to a level such that adding the incoming fuel (with low impurities) will not keep the recycling mass critical. Our rough estimate is that three recycles may be possible (the first from used UOX-51 via UREX+1 plus two more with either AIROX or melt refining) if impurity-tolerant fuels are used.

Uranium must also be considered as a potential source of impurities. The 2009 Fuel Campaign working limits on TRU feedstock into fast reactor recycling was based on a particular fast reactor conversion ratio (hence TRU:U ratio of fresh fuel) and on assuming pure uranium feedstock such as depleted uranium [4]. However, there are three good reasons why recovered uranium must be considered an option. First, if recovered uranium is not used, it must be stored (for eventual use in breeder reactors) or disposed; both have costs. Second, there are proliferation resistance motivations to keep as much uranium with TRU as possible, per assessment in some methodologies. That means that some of the uranium in the fuel is RU, not DU. Third, in some technological options, separating U from TRU can imply increased cost.

So, our analysis considered both “DU feed” and “RU feed” cases. In the current separation flow sheets for UREX+1 and electrochemical separation, not only do the RU streams have impurities, but for some elements, they are higher than the TRU-U product stream.

Topic 3: The sustained recycle strategy requires

Losses of waste elements into fuel must be low enough that the impurity limits are met for whatever fuel technology and performance levels are established. The new tool FIT allows estimation of the impurity concentrations, constrained only by mass balance and the requirement that the reactor in the recycling loop has fuel with estimated criticality (by one-group cross-section approximations) to match that of the original pure fuel specification.

Figure 3 shows an example – the estimated lanthanide impurity in oxide fuel with DU feed, with RU feed, or AIROX treatment of used FR fuel. In all cases, UREX+1a is assumed for used UOX-51 fuel separation. FIT cannot provide a direct estimate of individual lanthanide elements to compare with the limits of 660 ppm for the lowest-atomic-number lanthanides [lanthanum, cerium, praseodymium, neodymium, and samarium (the first lanthanides minus promethium, which does not occur in nature)] and 132 ppm for the later lanthanides [samarium, europium, gadolinium and dysprosium (some but not all of the rest of the lanthanides)]. Instead, FIT estimates the total lanthanide impurity level, a reasonable approximation given the relative abundances from fission. The figure indicates that the lanthanide impurity limits are met for the working UREX+1a separation factors. The “AIROX” case uses UREX+1 separation for used UOX-51, thus, recycle-1 is the same as the “RU feed” case. Thereafter, the inability of AIROX to separate lanthanides from TRU causes the lanthanide content to increase rapidly. The total impurities go off-scale (from Figure 3) after the sole UREX+1 separation (recycle-1) and reach ~13% of fuel after two AIROX separations (recycle-3). Thereafter, our approximations indicate the fuel cannot be blended and remain adequately critical and so no recycles are possible beyond three.

Figure 3: Estimated lanthanide impurity in oxide fuels for fast reactor TRU CR = 0.5

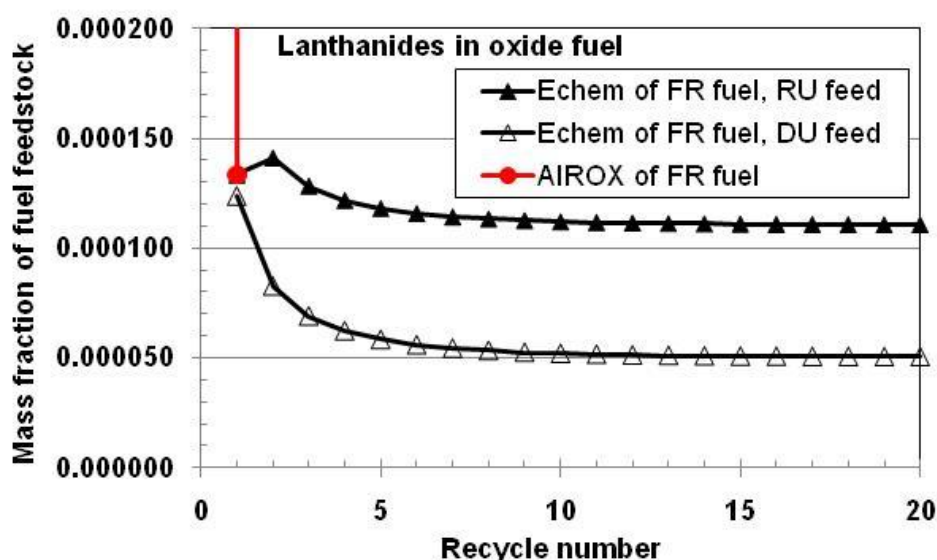
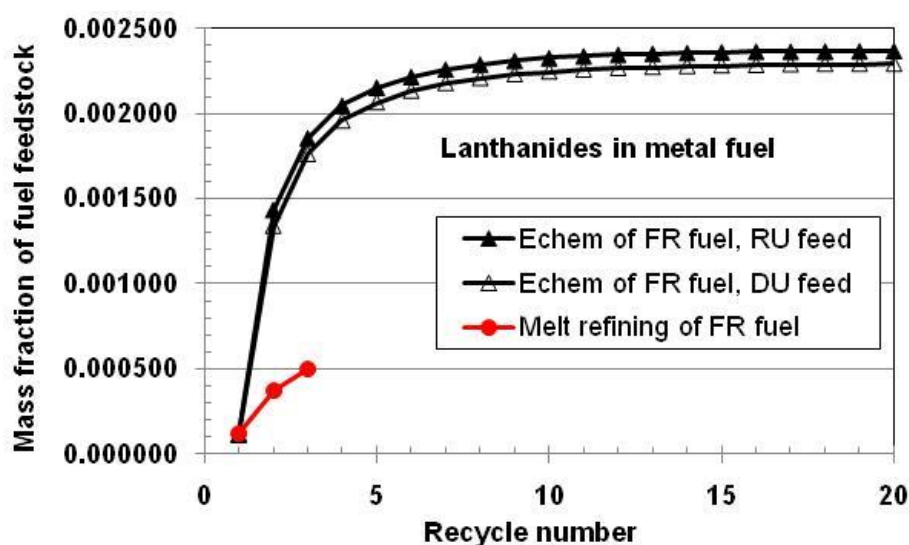


Figure 4 shows the same type of graph for metal fuel. Used fast reactor fuels are assumed separated by either electrochemical (in the “DU feed” and “RU feed” cases defined in the previous example) or melt refining. The carryover of lanthanides into recovered uranium in electrochemical separation is at or below detection limits, therefore we have assumed that only 0.1% of the lanthanides are in the RU stream. The low lanthanide impurity in the RU stream results in the “RU feed” case being only slightly higher than the “DU feed” case, unlike the previous example.

Because the electrochemical separation factors for the TRU-U product are still being determined, a conservative 5% of the lanthanides are assumed to be going into that product. This conservatism leads to exceeding the current 775-ppm working limit on lanthanides assumed for metal fuel. The little information available on melt refining indicates ~99% removal of lanthanides. If true, the separation of lanthanides from TRU would be better than that in electrochemical; this causes the low lanthanide impurity for the melt refining case in the figure. The inability of melt refining to remove other impurities leads to the total impurity level to reach ~8% after the third recycle (one from UREX+1 feed and two with a mix of UREX+1 and melt refining feed). At this point, the FIT model indicates that the reactor core cannot be made critical again without abandoning the limited separation strategy. The available feed streams cannot be blended to make a fuel with reactivity equal to that of the original fuel recipe.

Figure 4: Estimated lanthanide impurity in metal fuels for fast reactor TRU CR = 0.5

For sustained recycle, the following conclusions and observations can be drawn for the cases we analysed – fast reactors at transuranic conversion ratio of 0.5 fed by TRU from separated used UOX-51 fuel. The results will of course change for other sets of parameters. Nonetheless, they do suggest increased attention to transition metals and lanthanides.

- Lanthanides. These are a potential problem in metal fuel with the assumed conservative working electrochemical separation values; the lanthanide impurity limit is met for oxide fuel with UREX+1 separation.
- Fuel limits for alkali metals (Group 1), which are only limited for oxide fuels, are apparently met with working UREX+1a separation values.
- Fuel limits for alkaline earth metals (Group 2), which are only limited for oxide fuels, are met when the UOX and FR separations process is UREX+1a.
- Molybdenum (metal)/molybdenum plus silver (oxide). In both metal and oxide fuels, these elements are part of a group transition metal limit that contain potential impurities that are not fission products and are therefore not addressed in this study. Thus, it is particularly worrisome if the fission product impurities alone exceed the working limits. This appears in the oxide/UREX+1a system for the set of working separation factors. However, as explained in more detail in the full report, all calculations involving transition metals are particularly uncertain.
- Ruthenium (only limited for oxide fuel). Our first rough estimate for this transition metal shows its working limit for the oxide fuel is exceeded. However, this limit is apparently based solely on displacement of fissile content, so it may be too restrictive.
- Cadmium (metal)/cadmium plus tin (oxide). In both metal and oxide fuels, these elements are part of a group transition metal limit, with both fission product and non-fission product elements. The estimated values in the oxide fuel exceed the working limit, but these estimates must be considered very uncertain because of the current approximations in FIT involving transition metals.
- Halogens. Impurity limits appear easily met for both oxide and metal fuels.

Topic 4: The single recycle strategy requires...

One possible single recycle approach is use of “minimum fuel treatment” options such as AIROX or melt refining. With this approach, the fundamental need to make single recycle options attractive are impurity-tolerant fuels and either: i) the use of reactors that require lower TRU enrichment than used UOX-51 (1.3% TRU); ii) a way to increase the reactivity of fuels made with minimum fuel treatment via addition of enriched uranium.

High impurities are analogous to extra long-life (high burn-up) in that more unfissionable material accumulates in in-service fuel. Therefore, research and development (R&D) thrusts like impurity-tolerant fuel matrices and clad coatings that protect against impurities may enable minimum fuel treatment and/or high burn-up. The Fuels Campaign seeks to determine how impurities generated during irradiation behave versus how impurities that remain in recycled fuel residual after separations behave.

Topic 5: More complete disassembly of used fuel would have advantages, and costs

The working value for the fraction of TRU in undissolved fuel solids (hence waste) in UREX+1 is 1%. This is a factor of 10 higher than the 0.1% assumption in the 2008 DSARR report [5]. Furthermore, the 0.1% value was to represent all losses of TRU into waste, separation, undissolved solids, and fuel fabrication.

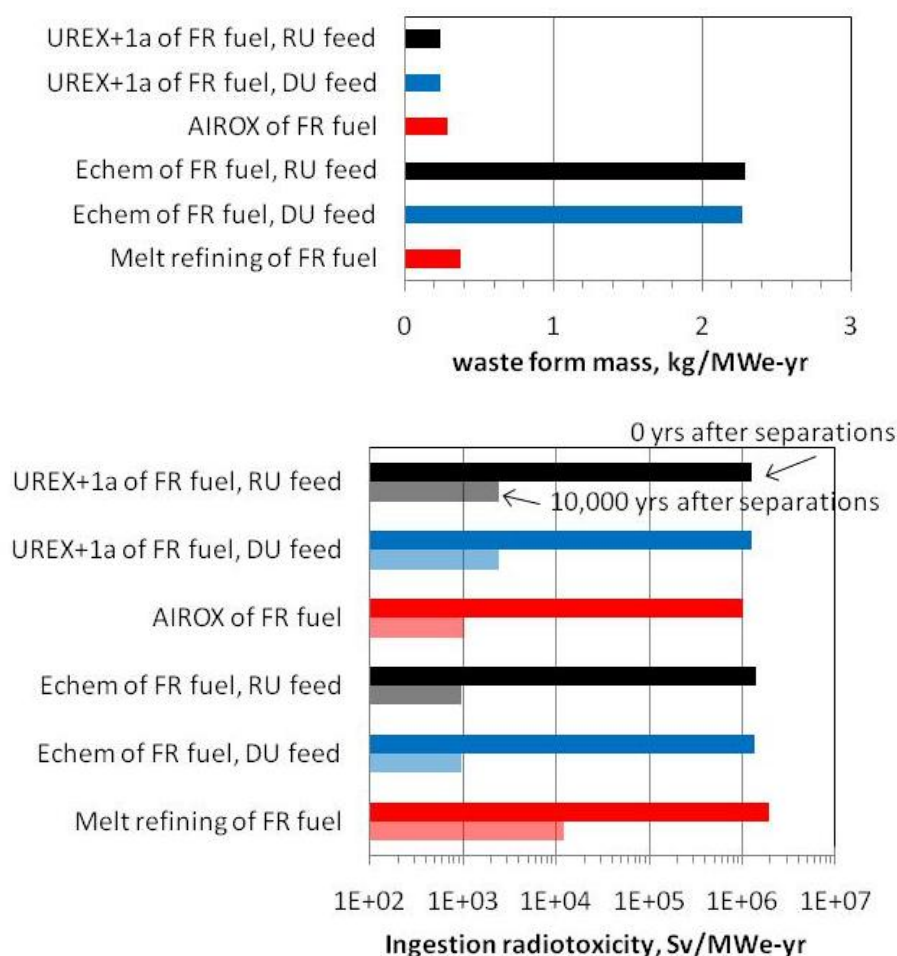
The working value for the fraction of TRU in cladding or disposed salt in electrochemical separation is 0.7% to cladding and 0.1% to salt; again, an order of magnitude higher than the 2008 assumption of 0.1% [5].

The 2008 assumption of 0.1% is, of course, neither a requirement nor even a guideline. But, the generally attractive waste management results of that study do depend on the fraction of TRU in waste. The working values of 1% and 0.8% (0.7%+0.1%) could be lowered by new advances or changes in technology, e.g. dissolution of undissolved solids by hydrofluoric acid, with corresponding costs and complexity. The details of such changes are beyond the scope of this report.

Other “solids” include cladding or fuel coatings. Several of the working impurity limits in oxide or metal fuel pertain to the impact on cladding, not the fuel matrix itself. This must be remembered as alternative cladding/coatings are considered. For example, use of a more resistant coating/cladding can loosen working impurity limits. Consideration of a new coating/cladding should include its potential compatibility issues with the fuel matrix plus the fuel’s likely impurities. For example, palladium is an issue in HTGR fuels due to attack on SiC. That would seem to be an issue if SiC is considered as a cladding option on other fuels at similarly high operating temperatures. This example is more relevant for TRU-based fuels than U-based fuels because the fission product yield for Pd is higher with ^{239}Pu fission than ^{235}U fission.

Figure 5 shows how different separations processes can affect the amounts and radiotoxicity of the waste streams that contain fission products, as normalised to the electrical energy produced. To include data for the AIROX and melt refining cases, the values in this figure are from the third recycle and so do not represent a sustained recycle. The mass of the waste forms (borosilicate glass and glass bonded zeolite) that contains the fission products (FP) from the cases that use UREX+1 and echem for the separations processes is almost a factor of 10 larger than for the cases that do not use echem, because of the expectedly low waste loading in the echem glass-bonded zeolite.

The radiotoxicity of the waste forms that contain the fission products is similar for all of the analysed cases at the time of when the waste forms are generated; but after 10 000 years decay, the radiotoxicity of the waste forms from melt refining ranges about a factor of 10 above the ranges of radiotoxicity for the other cases. This is because melt refining is expected to incompletely separate TRU isotopes from the waste, leaving an estimated 8% of the TRU elements in the fission product waste streams. The other separations processes leave between about 0.1% and 1% of the TRU in the separated waste streams.

Figure 5: Fission product waste stream mass and radiotoxicity for selected FIT cases

Similar trends are true for decay heat. But the decay heat of the separated waste streams for different separations processes can be further impacted. The melt refining literature indicates that melt refining can separate Group 1 elements such as Cs from Group 2 elements such as Sr. If these streams are not recombined to produce a single high-heat, high-radiotoxicity waste stream, then it could create multiple high-heat and high-radiotoxicity fission product waste streams, which could complicate waste disposition and increase costs.

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David Shropshire led this activity until late February 2010, when he started a two-year assignment in the Netherlands, where he is thriving. We thank him and wish him continued good times and success.

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