Finely-Divided Metal as Nuclear Power Reactor Fuel
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Abstract

A new form for metallic nuclear reactor fuel is proposed, consisting of finely-divided particles (tens of micrometers) mixed with sodium for thermal bond. Fuel pins filled with this form of fuel would have greater fuel density than with solid slugs fabricated at 75% smear density. Greater fuel density reduces enrichment requirements for initial fuel loading. The form has greater thermal conductivity and expansion than solid metal, at all fuel mixing ratios and operating temperatures, with important implications for reactor stability and safety. Larger surface-to-volume ratio allows more fission product gases and metallic fission products to diffuse out of fuel particles, resulting in less swelling, greater burnup before processing, and simple preliminary thermo-mechanical spent-fuel processing steps that might be used several times before the more expensive pyroelectric process develolped for Experimental Breeder Reactor II. Less frequent pyroelectric processing, simple preliminary processing, and larger surface-to-volume ratio reduce total processing cost. Preliminary processing produces separate fission products, in particular caesium and strontium, in metallic rather than salt or mineral form, thereby simplifying and reducing storage cost. Intrinsically structurally weak fuel would not rupture fuel pin cladding by swelling. Expense and complexity of the process would be offset by reduced total system cost.

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1 Why metallic fuel?

Till and Chang [38], Hofman, Walters, and Bauer [26], Wade, Wigeland, and Hill [40], and Ackerman et al [5] explain the benefits of metallic fuel with sodium thermal bond, for reactor safety, reactor design, and spent fuel processing:

- Higher thermal conductivity means
  - lower fuel temperature,
  - excellent power transient capabilities,
  - less doppler reactivity, and
  - better passive cooling.
- Higher actinide density improves neutron economy and the internal conversion ratio.
- Spent metallic fuel is easier and less expensive to process than ceramic fuel.

2 Irradiated metallic fuel in EBR-II

Hofman et al [26, p. 90] reported that in Experimental Breeder Reactor II (EBR-II), solid metallic fuel slugs were prepared having a smear density of 75%, that is, their initial diameter was about 85% of the inner diameter of fuel pins. When fuel slugs were inserted into fuel pins, sodium was added to provide a thermal bond to cladding, and to allow for swelling. Irradiated fuel expanded about 15% radially, and made contact with fuel pin cladding after 1.6% burnup. Axial expansion was 8–10% in U-Fs[1] and U-Zr fuels, and 3–4% in U-Pu-10Zr fuel [12, p. 162] [26, p. 91]. Fuel volume density was 69–73% after swelling. Swelling in solid metallic fuel is driven primarily by fission-product gases [26, p. 85]. Figure 6-2 in [38, p. 126] and Figure 8 in [26, p. 94] show the size of those voids is in the range of 5–75 μm.

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[1] Fissium (Fs) was the name given to an artificial fission-product alloy mimicking the composition resulting from recycling by melt-refining in EBR-II before pyroelectric reprocessing was developed [38, p. 124].

The change in sodium level above irradiated fuel was not sufficient to account for swelling [26, p. 94]. The deficit was attributed to sodium logging. Figure 9 in [26, p. 95] shows that thermal conductivity abruptly increased at about 1.6% burnup. The associated text explains that this was also due to sodium logging.

Sodium logging occurs when pores break open at the surface of fuel, gases escape, and sodium intrudes. Swelling stops in part because fuel contacts the inner surface of cladding and is mechanically restrained, and in part because voids interconnect and open to the surface at about 30% swelling, at about 1.6% burnup [26, p. 88], creating larger surface area, which allows gases to escape to the plenum more efficiently.

Table 1 in [26, p. 98] remarks that at 10% burnup, 70% of alkali metal fission products (caesium and rubidium), and 20% of alkaline earth fission products (barium and strontium), were found in bond sodium. Whether other fission products, in particular low melting point metals such as arsenic, cadmium, francium, lithium, selenium, and tellurium, dissolve into bond sodium, is not mentioned.[2]

3 Finely-divided fuel

If the surface-to-volume ratio were larger, more fission-product gases would diffuse directly from the surface of fuel, and thence to the plenum, instead of creating voids, and more metallic fission products would diffuse into bond sodium. Dissolved fission products would increase the sodium level above fuel, not cause swelling. The simplest way to increase surface-to-volume ratio is to use small particles. The surface-to-volume ratio for spherical particles is $\frac{3}{r}$. With small particles, more fission gases and metallic fission products diffuse directly from the surface, and more voids reach the surface, at smaller sizes, reducing swelling. The interior surface of open voids further increases the surface area of fuel in contact with bond sodium.

With spherical particles of uniform size, and no container boundary, optimum packing density is $\frac{\pi}{\sqrt{18}} \approx 74\%$.[3] Scott and Kilgour [34] reported that when uniform-size particles were put into a container and then shaken (called *random close packing* or RCP), the density was 64%. They did not describe the shape of the container, or the relationship of the size of the container to the size of the particles. With mixed particle sizes, packing density would be greater because small particles fit between large particles. For two sphere sizes, Yamada et al [42] found RCP density up to 74.1%. Farr and Grote [20] report RCP densities from 64% to 97% for polydisperse spheres of log-normal size distributions with $0 < \sigma < 3$. Fuel pins filled with finely-divided fuel particles having a sufficiently broad polydisperse size distribution will have initial fuel density that is greater than with solid fuel slugs fabricated at 75% smear density.

When more gases and metallic fission products escape into bond sodium, fuel particle swelling is reduced. Rather than being caused by trapped gases, swelling would largely be due to fission product metals that are precipitates or in solution in the alloy [26, pp. 88, 97-98], that are less dense than fuel, and that do not migrate to bond sodium. It should be substantially less than solid fuel slug swelling. As burnup proceeds, fuel density should remain comparable to or greater than the density with solid fuel slugs. Higher fuel density improves neutron economy and makes it easier to maintain a hard neutron spectrum. Improved neutron economy allows lower fissile content in the initial fuel load, and improves the internal conversion ratio. [38, p. 312]

The primary limitation to fuel burnup is fuel pin cladding durability, and bundle-duct interaction due to radial fuel pin strain [38, p. 116] [26] [37, p. 2]. Hofman et al [26] discuss fuel-cladding mechanical interaction (FCMI) and fuel-cladding chemical interaction (FCCI). Because fuel composed of small particles and bond sodium is motile, it would not, in itself, impose significant radial stress on fuel pin cladding. Expansion would all be in the axial direction. Thermally-driven axial expansion produces negative reactivity feedback and attendant important inherent safety advantages [12, p. 167]. Radial cladding strain would be reduced or eliminated. Radial stress would be due to gases that escape from fuel particles to the plenum. A longer plenum would ameliorate radial stress, and a connection to an exhaust port would eliminate it. Reduced mechanical stress would allow fuel pin cladding materials to be chosen based more upon considerations of FCCI and irradiation damage, rather than FCMI. Thereby, it might be possible to develop more durable

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[2] Added 3 June 2022: Cadmium and sodium form alloys [31].

[3] This was conjectured by Johannes Kepler in 1611. Thomas Hales published a proof 10 August 2014 [2022-09-11].

cladding without increasing the structure fraction, allowing greater burnup before fuel processing.

In solid fuel slugs, alloy constituents redistribute along temperature gradients, creating radial zones as shown in Figure 6-4 in [38, p. 127]. This would not occur in particulate fuel. 10 wt% (25 at%) zirconium was used in EBR-II to reduce plutonium migration and prevent formation of a plutonium-cladding eutectic [38, p. 124]. It might be possible to reduce zirconium content.

To create fuel particles with polydisperse sizes, spray molten fuel through nozzles or gas-powered atomizers into a tower containing inert gas or sodium vapor, ~~and~~ or pour molten fuel onto a disk atomizer. Deposit particles into a sodium pool at the bottom. Nozzle geometry, fuel temperature, and flow rate determine the distribution of particle sizes. Cool the pool to prevent boiling, or draw off sodium vapor, condense it externally, and feed it back into the pool. Control criticality with pool geometry and batch size.

Alternatively, create and decompose hydrides. The formation of UH_3 is an exothermic reaction that is self-limiting at 225°C, where UH_3 formation and decomposition are in equilibrium [25]. Higher reaction rates occur with larger surface-to-volume ratio, such as with small wires. UH_3 can be decomposed in vacuo at 225°C. The resulting powder size is in the range $35 - 45\ \mu$m. Uranium particles tend to self sinter because of high surface reactivity. The result is brittle and can be easily mechanically comminuted to the desired powder size [25, p. 2]. This powder can then be mixed with sodium to make a slurry. In vacuo or argon, because of high friction, the density of the powder is low, in the range of $6 - 8$ g/cc, or $31 - 42\%$ the density of solid metal. The density can be increased to 11.5 g/cc, or 60% the density of solid metal, by cold compacting, and higher densities[4] by hot compacting, which creates a rigid material. Hausner and Zambrow [25] do not report on density, friction, or viscosity in the presence of sodium. Notley et al [32] report preparing plutonium powder using a hydride process. Haschke et al [24, p. 1] report that plutonium hydride behaves similarly to lanthanide fluorides. This might be true of other actinide hydrides as well. Zirconium hydride decomposes at 300°C [1, p. 4]. Lanthanide hydrides, among the most stable hydrides [11], decompose at about 800°C. It might be possible to prepare purely-metallic powders of partially processed spent fuel using a hydride process. Preparation above liquid sodium must proceed above 450°C, the decomposition temperature of NaH. NaH is soluble in liquid sodium. Therefore, if NaH forms in liquid sodium, and hydrogen cannot be removed by heating in vacuo, powder should be created separately, and then comminuted to the desired sizes and mixed with sodium.

Fill fuel pins by pouring or pumping a stirred heated slurry into fuel pins, or by vacuum or injection casting directly into fuel pins from the bottom of a settled rather than stirred sodium pool. Because it is only necessary to melt the bond sodium, not the fuel carried in the slurry, this process would use a much lower temperature than processes used to cast solid fuel slugs for EBR-II [38, § 5.2.1] [29, p. 3-41]. It would not be necessary to use glass molds, or to destroy molds [38, p. 123] to extract fuel slugs.

Settle hot fuel inserted into pins using vibration to remove voids. Because of the enormous density difference between sodium and fuel, this should proceed rapidly. If fuel particles bond due to uranium's self reactivity, preventing settling, break bonds by ultrasound, or by inserting a rod into the fuel pin. Distill excess sodium, or decant it from above settled fuel. If it is decanted into the spray tower sodium pool, it does not matter if it contains fuel particles. Inspect using X-ray or ultrasound to detect non-uniform fuel density.

With sufficiently small particles, fuel would initially be in a condition of essentially open porosity. During irradiation, voids would not abruptly open, and fuel would not abruptly contact cladding. Thermal conductivity would change less, and gradually rather than abruptly. Control would be simpler and safer.

4 Spent fuel processing

The ultimate goal of spent fuel processing is to increase burnup essentially to completion – to consume all actinides put into the reactor – and to produce only fission products as waste. Transuranic actinides (TRU), especially ^{239}Pu with its 24,110-year half life, are the components of spent fuel that require unrealistic custody duration [22]. Fission products require much simpler custody of much shorter duration (infra).

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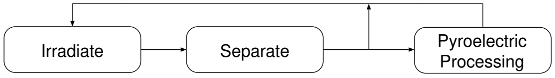
[4] The density of uranium in UO_2 is 51% the density of solid uranium.

Process spent fuel using two major steps: (1) Separate bond sodium with its dissolved metals, and low boiling point metals, from fuel particles, and (2) process fuel particles pyroelectrically in an electrolytic cell having a molten lithium-potassium chloride electrolyte. Spent fuel is the anode. When a current is applied, uranium, plutonium, and other actinides migrate through the electrolyte and deposit at cathodes. Noble metals such as molybdenum and ruthenium remain in the anode. Other fission products remain in electrolyte [38, ch. 8–11]. This is similar to widely-used metallurgical processes, such as electroplating, and the Hall-Héroult process [39] used to refine aluminum. Figure 1 illustrates that irradiation and separation can be repeated several times before more expensive pyroelectric processing is necessary.

Figure 1: Overview of fuel cycle



The Sodium Waste Treatment program [4, p. 55] determined that sodium and solid fuel could be removed from EBR-II cladding by removing the bottom end cap and heating the fuel pin. Removing the bottom end cap first allows plenum pressure to help expel fuel. To remove finely divided fuel, roll the fuel pin between three rotating cylinders in case it is necessary to break brittle [25, p. 2] self-sintered fuel, heat the fuel pin, remove the bottom end cap, and then the top end cap. Wash out residual particles using clean sodium, or a boiling fluid such as argon. Cavitation from boiling would remove particles that are not strongly bonded to cladding. Distill fuel pin cladding to remove sodium and volatile fission products, then cleanse with hydrochloric acid, and finally, if necessary, process it pyroelectrically to separate remaining spent fuel and activation products. Dry resulting chlorides and add them to electrolyte during pyroelectric processing. Pyroelectrically processing cladding separately from fuel will be substantially faster than processing fuel pins containing solid slugs that are bonded to cladding, because there will be less material to transport into and through electrolyte, and the internal surface of cladding will be accessible to electrolyte throughout processing. Cladding processed separately from fuel might have sufficiently low activity that it does not need storage; it can be immediately disposed or recycled.

4.1 Filtering

Crush brittle conglomerates in removed fuel to yield smaller particles. Then separate bond sodium and its dissolved fission products from fuel particles by filtering.

To reduce the amount of fission products adhered to fuel particles, wash crushed fuel using aliquots of clean sodium, then purge remaining sodium by flowing argon through the fuel in the filter.

After mechanically removing particulate residue, cleanse a sintered-glass filter by back-flowing argon, and then hydrochloric acid. Dry resulting chlorides and add them to electrolyte during pyroelectric processing. Back-flowing would reduce accumulation of insoluble AgCl and $Sn(OH)_x$. If a sintered-fuel filter is used, simply crush it and process it as particulate residue.

Wash and filter at high temperature to reduce sodium viscosity and increase sodium halide solubility [33]. The advantage of washing and filtering is that barium and strontium dissolve in sodium [26]. If fuel and bond sodium are not mechanically separated, all of the innocuous barium remains with fuel particles, increasing the frequency of pyroelectric processing, and it is eventually (but pointlessly) stored in the mineral waste form produced after pyroelectric processing [38, §8.5].

Hofman et al [26] do not discuss whether metals such as cadmium and chalcogenides dissolve into bond sodium. As shown in [18], small amounts of antimony, beryllium, cadmium, gallium, germanium, indium, selenium, silver, tellurium, and tin are soluble in sodium at operating temperature. ~~If so, or if they can be washed from the surface of fuel particles, they can be separated from fuel particles by filtering.~~

4.2 Distillation

Filtering produces two products: (1) filtrate composed of sodium, dissolved fission products, and particles small enough to slip through the filter, and (2) fuel particle residue. Process both, separately, by distillation.

Distilling filtrate separates

- low boiling point alkali metals – caesium and rubidium with picogram quantities of francium,
- sodium,
- sodium iodide,
- strontium and sodium bromide, and
- a residue consisting mostly of barium, with small amounts of gallium, germanium, and indium. Depending upon fuel particle sizes and filter porosity, the residue might contain small amounts of fuel. Separate barium ~~and fuel~~ using sulfuric acid – barium sulfate is not soluble in water. Convert dissolved sulfates – fuel and fission products other than barium – ~~to chlorides using barium chloride, which will be converted to insoluble barium sulfate.~~ to carbonates using $(NH_4)_2CO_3$. Separate carbonate precipitates from $(NH_4)_2SO_4$. Decompose $(NH_4)_2SO_4$ to NH_3 and H_2SO_4 [23]. Convert carbonates to chlorides using HCl. Save the evolved CO_2. Dry resulting chlorides and heat them sufficiently to drive off NH_3 and HCl. Save the evolved HCl. Combine NH_3 with CO_2 to re-form $(NH_4)_2CO_3$. ~~and add them~~ Add chlorides to electrolyte during pyroelectric processing. If strontium is removed by distillation, most of the sulfate residue will be innocuous barium. Activity of the residue should be low enough that it can be processed in a glove box, not a hot cell.

Distilling fuel particle residue separates

- cadmium and selenium,
- tellurium,
- residual sodium,
- caesium, rubidium, and strontium, from adhered residual sodium, or not diffused into bond sodium and recovered in filtrate, and
- fuel residue with fission products not removed by distillation – barium, lanthanides, and noble metals.

As alloys, boiling points change. There might be azeotropes that make high separation factors difficult, but the experience of the Sodium Waste Treatment project [4, pp. 68-69] suggests otherwise.

The amounts of low boiling point fission products given in Table 1 are per tonne (1000 kg) of spent Light Water Reactor (LWR) fuel, according to output [13] from the ORIGEN 2 computer program [2] [16]. Fuel had undergone 50.689 GWth-day burnup at 36.542 MWth, or 1387 days of operation, with 3.14×10^{14} n cm^{-2} s^{-1} neutron flux, resulting in 52.18 kilograms of fission products, and was then stored for ten years. In higher-burnup metallic Integral Fast Reactor (IFR) fuel, the amounts would be greater, but the proportions roughly the same [13].

Table 1: **Low boiling point fission products from 1000 kg of 50.689 GWth-day LWR burnup**

	Mass	Moles	Boiling Point °C		Mass	Moles	Boiling Point °C
At	< 1.0 pg	< 1.0 pmol	336.8	Po	10.43 pg	< 1.0 pmol	962
As	0.2982 g	3.981 mmol	613	Te	749.1 g	5.797 mol	967.8
Cs	3.680 kg	27.3 mol	670.8	Es	< 1.0 pg	< 1.0 pmol	996
Fr	< 1.0 pg	< 1.0 pmol	676.8	As_2Te_3			1027
Se	84.68 g	1.048 mol	684.8	CdTe			1050
Rb	524.5 g	6.079 mol	688	Yb	42.96 μg	252.7 nmol	1196
Cd	198 g	1.776 mol	767	NaI	421.6 g	2.784 mol	1304
NaH		Decompose	800	Li	25.74 μg	3.708 μmol	1330
As_2Se_3			861	Sr	1.135 kg	12.76 mol	1382
Na			882	NaBr	41.04 g	0.395 mol	1390
Zn	9.716 mg	9.716 mmol	905	Ba	2.776 kg	20.23 mol	1897
Total	9611 grams, 78.17 moles, excluding sodium metal and intermetallic compounds						

Arsenic, cadmium, selenium, and tellurium might be present as intermetallic compounds, not separate metals. Cadmium forms alloys with plutonium [19, p. 25]. Till and Chang report that cadmium can be distilled from fuel [38, pp. 171, 176] [37, p. 3] [8, p. 9]. It might not be possible to distill chalcogenides individually. Distilled sodium might be contaminated with millimole quantities of As_2Se_3 because of their essentially identical boiling points. It might be possible to separate As_2Se_3 from sodium by crystallization.

The melting point of U-19Pu-10Zr [26] is about 990°C [19, p. 184]. With constant zirconium content, increasing plutonium content reduces the melting point. Data concerning the melting point of spent fuel, as a function of burnup, are not available. If the melting point is not significantly increased by fission products, it is not possible to distill all the metals in Table 1 from spent fuel particles without melting them. It is possible to separate arsenic, cadmium, caesium, rubidium, selenium, and As_2Se_3, and possibly tellurium, depending upon the extent to which they diffuse to the surface of fuel particles.

This suggests two regimes for treating particulate filtering residue:

1. Distill fuel particles without melting to remove arsenic, caesium, rubidium, selenium, residual sodium, and As_2Se_3, and cadmium to the extent it is not bound in high boiling point chalcogenide intermetallic compounds that do not decompose. It might be possible to distill significant amounts of elemental tellurium without melting because its amount is much larger than the amounts of metals with which it might form high boiling-point compounds.
2. Melt fuel particles. Immiscible fission products have low density and float atop fuel. The ones with high melting points can be separated from fuel by crystallization (Table 2); ones with lower melting points than fuel might be carried away with molten residue after fuel solidifies. The primary contaminants remaining in fuel are noble metals such as molybdenum and ruthenium [29, p. 1-7]. This is similar to the melt refining process used at EBR-II before pyroelectric processing was developed [38, pp. 25, 168] [37, p. 6].

   Distill metals separated from fuel, primarily to separate strontium from barium, and to keep them out of the mineral waste form resulting from pyroelectric processing. If barium and strontium are effectively removed by diffusion into bond sodium, this step might not be advantageous, depending upon whether tellurium can be removed without melting fuel particles, and the reduction in fuel worth caused by the presence of tellurium.

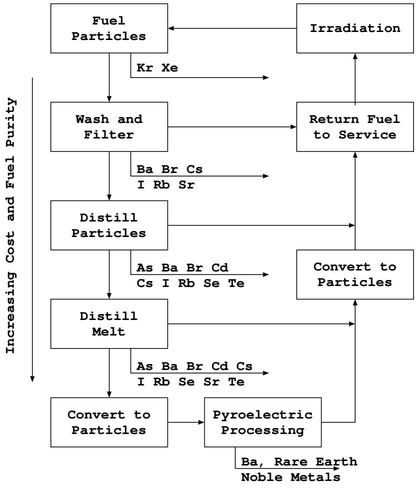
   As_2Te_3, CdTe, and probably CdSe, would be collected with tellurium.

An alternative or precursor to distilling particles is to hydride, dehydride, crush, mix with sodium, and separate again. This re-distributes elements that are soluble and not soluble in sodium into different particles, and different distances from the surfaces of particles, so that more alkali and alkaline-earth metals, and perhaps cadmium and chalcogenides, can be washed from particles and separated by filtering. Differential solubilities of hydrides in liquid sodium might be exploited before dehydriding. This would certainly remove all fission-product gases. Sufficient repetition might eliminate the need to distill from particles.

The reason for filtering instead of simply distilling mixed fuel particles and bond sodium is that strontium and barium dissolve in bond sodium, but strontium cannot be distilled without melting fuel, and barium can be distilled only at very high temperature.

Selenides and tellurides of strontium and barium have melting points above 1,600°C. Boiling ~~and melting~~ point data for arsenides, selenides, and tellurides of sodium, caesium, and rubidium, are not available. Melting points of caesium, rubidium, and sodium arsenides are assumed to be similar to potassium arsenides.

Figure 2: Closed Fuel Cycle



If distillation uses a constant temperature sufficiently high to boil all volatile products, collect different distillates in trays at different levels in a tower, maintained at appropriate temperatures. If different metals are collected in a retort by changing the temperature, different containers should be used to collect them, to reduce cross contamination and to avoid collecting sodium with fission products.

Because of the larger surface-to-volume ratio, more of the fission products that have not diffused into bond sodium can be distilled from finely-divided fuel particles than would be possible with solid fuel slugs. More bromine, iodine, and low boiling point metals would be driven out of small fuel particles than could be driven out of solid fuel slugs.

Ideally, caesium should be separated from rubidium because fission-product rubidium is not significantly radiotoxic, 3.352 μSv/gm, 60.4 nCi/gm, about twice the activity of rubidium in nature. Because their boiling points are different by only 17.2°C, it might be difficult or expensive to separate them by distillation. In mining operations, they are separated using potassium aluminum sulfate ($KAl(SO_4)_2$). If not separated, the volume of arsenic, caesium, francium, rubidium, and selenium together would be about 19% greater than the volume of caesium alone, and the specific heat production (W/liter) would be reduced by about 16%.

Whether to distill further metals by using higher temperatures can be decided in due course. The next metals to evaporate would be microgram and picogram quantities of californium and thallium, at 1472°C, significant amounts of europium at 1529°C, and small amounts of antimony at 1587°C. Barium boils at 1897°C. Removing europium is attractive because, after removing caesium and strontium, it dominates heat production and radiotoxicity in electrolyte, and in the mineral waste form.

Figure 2 shows that fuel processed by any of these steps could be immediately diverted back to service without costly pyroelectric processing [38, ch. 8–11]. An advantage of reusing particulate fuel, without melting it and making new particles, is that particles' surface areas are increased by pores having opened. Determine the selection of fuel pins or fuel assemblies to divert at each stage using models, analyses, and processing records. Figure 3 (page 20) shows the process in more detail.

4.3 Crystallization

The relationships between the melting points of the fission products described in Table 2 are different from the relationships of their boiling points described in Table 1. Several elements and compounds can be removed by cooling and filtering filtrate~~, or~~. There are no data whether alloys with sodium can be separated using a cold plate, Czochralski process, or zone melt melting.

~~Most fission products cannot be separated by starting at the highest melting point and gradually cooling because the melting points of many are higher than the boiling points of others.~~

The binary alloys with small concentrations of sodium-soluble products all have melting points near pure sodium. Data for *n*-ary alloys are not available. Only small amounts of cadmium can be extracted because there is a 95.4°C eutectic at 97.2 wt.% Na [18, p. 689].

When distilling the filtrate or fuel particle residue, it is likely that caesium, picogram quantities of francium, and rubidium will be collected together. ~~There are no data whether caesium and rubidium can be separated by crystallization, but it is unlikely because their melting points differ by only 10.86°C.~~ Starting from an alloy of Cs-13 wt.% Rb, significant amounts of caesium can be separated from rubidium, but they cannot be completely separated because there is a 9.7°C eutectic at 47 wt.% caesium [18, p. 900].

Borho et al proposed a method to separate eutectic mixtures [9].

When distilling the fuel particle filtering residue, it is likely that arsenic, astatine, and selenium are collected together. The melting points of arsenic and selenium are very different from the melting points of the others. It might be easy to separate them by crystallization, provided they do not form an alloy. As_2Se_3 might be collected with sodium by distillation. As_2Se_3 and sodium have very different melting points and could be separated by crystallization, perhaps as sodium flows from a distillation tray, provided they do not form an alloy. Sodium with small amounts of As_2Se_3 can be used as thermal bond.[5]

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[5] 83% of selenium in spent fuel is ^{80}Se and ^{82}Se, which have approximately the same thermal neutron absorption cross section

Table 2: **Melting points of some fission products**

	Mass Grams	Melting Point °C		Mass Grams	Melting Point °C
Fr	< 1.0 pg	27	As_2Te_3		621
Cs	3.680 kg	28.44	NaI	421.6 g	661
Rb	524.5 g	39.3	Ba	2776 g	727
Na		97.79	NaBr	41.04 g	747
Li	25.74μ g	180.5	Sr	1.135 kg	777
Se	84.68 g	220	As	0.2982 g	816.8
Po	10.43 pg	254	Yb	42.96μ g	824
At	< 1.0 pg	302	Es	< 1.0 pg	860
Cd	198 g	321.1	Na_2Se		875
As_2Se_3		360	Na_2Te		953
Zn	9.716 mg	419.5	CdTe		1041
Te	749.1 g	449.5	CdSe		1268
Total	9611 grams, 78.17 moles, excluding sodium metal and intermetallic compounds				

4.4 Prior art

Starting in 1978, Argonne National Laboratory–West (now Idaho National Laboratory) developed a process to dispose of elemental sodium waste [4]. The original reason for the program was that DoE would not (and still will not) accept materials for burial if they contain elemental sodium, because of the risk of corrosion, fire, or explosion, if moisture intrudes.

The objective was to remove sodium from spent fuel, and from scrap such as fuel assemblies.

Several methods were considered. The one ultimately recommended was called MEDEC, for **ME**lt, **D**rain, **E**vaporate, **C**alcine. Calcining converts sodium to sodium carbonate, Na_2CO_3, for disposal, and is not further discussed here.

In addition to processing sodium for disposal, methods to purify sodium for re-use were developed in 1980. Distillation was the method that was ultimately chosen [4, pp. 68-69]. The conclusion was that caesium would be separated from sodium, and sodium would be separated from barium, strontium, other fission products, and fuel. They did not remark whether they attempted to separate strontium from the others.

Because the purpose of the test was to determine whether distillation was a potential method to purify sodium for re-use, not to perfect it, distribution factors were not measured, and the project was discontinued.

Till and Chang mention distilling bond sodium, but only in the context of reducing mineral waste form volume by keeping sodium out of electrolyte [38, p. 187].

4.5 After separation

Fission product gases will be essentially absent. Due to increased surface-to-volume ratio, and fewer closed voids, krypton and xenon will have largely escaped into the plenum during operation. Bromine and iodine react with sodium, forming NaBr and NaI, which dissolve in sodium and can be separated from filtrate or fuel particle residue by distillation or crystallization.

Fission product gases, and fission product metals in Table 1 or 2, constitute 34.3% by weight of fission products. With those removed, it might be possible to re-insert fuel particles and clean sodium into cladding,

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as sodium. The cross section for the others is roughly ten times larger. Heat capacity and thermal conductivity are much less than for sodium. Small amounts of As_2Se_3 would not cause problems, but it would eventually need to be removed.

several times, before pyroelectric processing is necessary.[6] This would reduce processing expense. If increased burnup before each fuel processing cycle is made possible, not least by improved cladding durability, this benefit might not accrue, but increased burnup before each fuel processing cycle in itself reduces processing expense.

With the metallic fission products in Table 1 or 2 removed, pyroelectric processing would proceed at a faster rate. If particles are not melted to remove strontium, or if the residue after melt distillation is converted to particles, the increased surface-to-volume ratio of fuel particles would allow pyroelectric processing to proceed at a faster rate than with solid cladding-bonded fuel slugs.

If fuel is cycled several times, using separation without pyroelectric processing, when it is pyroelectrically processed the concentrations of remaining fission products will be different, and perhaps greater. This might increase the average cost of individual pyroelectric processing runs, because it will be necessary to cleanse electrolyte more frequently. Offsetting the higher concentration, decay heat production within electrolyte will be reduced because caesium and strontium will be absent or much reduced, thereby reducing electrolyte cleansing frequency. The combination of these effects should reduce total pyroelectric processing expense.

5 Storage

The cost to store a fission product depends upon its thermal output, the degree and duration of its radiotoxicity, the density of the product in the storage form, and the volume of the storage form. Segregating fission products does not eliminate the need for a repository – but it does allow different repositories, with different technical requirements, and different costs, for different fission products [21]. Fuel pin fabrication cost and durability, reprocessing cost, storage cost, the value of recovered fuel, and the value of some separated fission products, are part of a system. The objective is to minimize total system cost, not to minimize individual component costs independently. Sometimes, reducing total system cost is achieved by increasing one component cost, resulting in greater decrease in different component costs, or reducing their frequency. Improving component processes, or performing component processes less frequently, reduces total component costs. The latter is a system effect that does not directly depend upon individual process costs.

The fission products that require the longest duration of storage (300-400 years) are caesium (30.04 year half life), strontium (28.79 year half life), and products collected with them. Of all fission products, they produce 99.3% of the radiotoxicity and 88% of thermal power (after ten years' storage), but constitute only 9.26% of the mass. A significant fraction of these is separated by filtering and distillation.

Till and Chang wrote [38, p. 241] that removing caesium and strontium would "increase repository space utilization by the huge factor of 225. . . . In the absence of these two activities. . . there would simply be no need for a highly engineered repository."

Among distilled metals, after caesium and strontium are removed, ${}^{125m}\text{Te} \xrightarrow[57.40\text{d}]{\gamma} {}^{125}\text{Te}$ and ${}^{113m}\text{Cd} \xrightarrow[14.10\text{y}]{\beta} {}^{113}\text{In}$ (Table 3) dominate radiotoxicity (99.6%) and heat production (99.9%). Cadmium constitutes only 2.1% by weight of the metals collected by distillation, and ^{113m}Cd is only 0.014% by weight of total cadmium.

Table 3: **Radiotoxicity and heat production of distilled metals, per tonne of spent fuel, after ten years**

Element	Mass	Radiotoxicity	Heat	Element	Mass	Radiotoxicity	Heat
Cadmium	198.0 g	50.99 kSv	100.8 mW	Iodine	357.6 g	196.5 Sv	233.3 μW
Tellurium	749.1 g	13.07 kSv	341.1 mW	Selenium	84.68 g	66.32 Sv	153.9 μW
Total	947.1 g	64.06 kSv	441.9 mW	Total	442.28	262.85 Sv	387.2 μW
Per gram		67.63 Sv/g	466.6 μW/g	Per gram		594.3 mSv/g	875.4 nW/g

Barium and rubidium are radiotoxically innocuous.

---

[6] The need for pyroelectric processing would likely be determined by buildup of samarium, a powerful neutron poison, which does not dissolve into bond sodium.

Till and Chang report that high-nickel containers ought to be impervious for at least twenty thousand years [38, p. 239]. Therefore, metals collected as such (not as chlorides) by distillation can be safely stored without converting them to minerals, if they do not damage their containers, and if DoE and NRC rules allow storing them as elements. Density of stored metals should approach 100%, depending upon container size and design. Small amounts and high density make these metals attractive candidates for deep borehole storage.

Density of fission products in the final mineral waste form described in [5, p. 141] is 4% by weight. Ackerman and Johnson [6] report that significant amounts of electrolyte are included as occluded and surface salt. Specific heat production from the mineral waste form will be much less than from pure metals. Metallic and mineral waste forms can be stored in different repositories, for different durations, at different costs [21]. If DoE or NRC rules require converting caesium and strontium to mineral form for storage, those minerals can and should be stored separately from mineral forms of other fission products, in different repositories, at different costs.

Strontium, NaI, and NaBr all have sufficiently high boiling points that they would only be separated from fuel by melt distillation. Some stainless steel alloys are susceptible to damage by chlorides, and might be susceptible to iodides and bromides. If NaI and BaBr would not damage high-nickel containers, there is no point to separate them: Storing NaBr with strontium would increase the storage volume by 91 ppm. Storing NaI with strontium would increase the storage volume by 0.08%. Otherwise, they must be removed, if strontium is to be stored as a metal. The boiling points of NaI and strontium are sufficiently different that they can be separated by distillation. It might be possible to separate NaBr and strontium by crystallization. If they cannot be separated by distillation or crystallization, separate NaI and NaBr as phosphate, sulfate, or fluoride: $Sr_3(PO_4)_2$, $SrSO_4$, and SrF_2 are ~~is~~ insoluble in water (SrF_2 is the least soluble), while Na_3PO_4, Na_2SO_4, NaF, NaI, and NaBr are soluble. $Sr_3(PO_4)_2$, $SrSO_4$, or SrF_2 might need to be converted to a less dense mineral storage form, unless ~~it~~ they can be stored in high-nickel containers without damaging them. Phosphates form glass well. If NaI and NaBr are problems, and only appear in strontium collected from filtrate or from particle residue, but not both, strontium collected in those ways should be stored separately, one as metal and the other within a mineral form.

Iodine has much lower activity than strontium. If NaI is separated from strontium, it ought to be stored in the mineral form derived from electrolyte cleansing. Adsorb it into Zeolyte A and combine it with Zeolyte-adsorbed fission products cleansed from electrolyte, to store iodine in mineral form. Including iodine in the mineral form would increase thermal production by 41 ppb and mass by 3% (assuming barium has been removed). Dispose nonradioactive NaBr.

Total radiotoxicity of fission products, without removing cadmium, caesium, strontium, and tellurium, is 155.5 MSv per tonne of ten-year old 5.2%-burnup LWR fuel. Thermal power is 1,375 watts. After removing those metals, and waiting about a month for ^{90}Y to decay, radiotoxitity is reduced 99.4%, to 885 kSv, and thermal power is reduced 92.6% to 101.9 W. This would allow to process more fuel batches before cleansing electrolyte of fission products. This benefit does not accrue if caesium and strontium are not removed.

If not separated by distillation, cadmium would remain with noble metals in the anode basket of the pyroelectric refiner [38, ch. 8]. Distilling and storing it separately from those metals reduces the radiotoxicity of that alloy by 20%, to 201.4 kSv, the mass by 1.1%, to 18.08 kg, and heat production by 1.3%, to 7.5 W. Heat production from cadmium is dominated by $^{113m}\text{Cd} \xrightarrow[14.10\text{y}]{\beta} {}^{113}\text{In}$. With cadmium removed from the alloy, $^{106}\text{Ru} \xrightarrow[1.020\text{y}]{\beta} {}^{106}\text{Rh} \xrightarrow[30\text{s}]{\beta} {}^{106}\text{Pd}$ produces most of the heat (2.66 μW/g) and radiotoxicity (10.9 Sv/g). After another ten years, $^{99}\text{Tc} \xrightarrow[214\text{ky}]{\beta} {}^{99}\text{Ru}$ produces most of the heat (457 nW/g) and radiotoxicity (25 mSv/g).

After uranium and TRU are deposited at two cathodes in the pyroelectric refiner [38, ch. 8], trivalent lanthanides are deposited at a third cathode, leaving divalent europium and samarium in electrolyte [3, p. 6]. Assuming actinides are present in electrolyte only below 100 ppm, and caesium and strontium have been removed, radiotoxicity, heat, and custody duration of remaining fission products in electrolyte are dominated by antimony, europium and samarium.

Europium is only 9.4% of fission products remaining in electrolyte, but it contributes 90% of radiotoxicity (624.1 kSv) and produces 93% of thermal power (73.35 W). Its presence requires custody of the mineral waste form from electrolyte cleansing for about 100 years (see Figure 4 on page 21). Without it, custody would be less than thirty years. It could be removed as sulfate because its sulfate is the least soluble of remaining elements, by a factor of 6×10^{-5}, assuming barium and strontium have been removed. Simply adding water to start a sulfate process to remove europium would remove silver and tin because AgCl is insoluble and tin chlorides hydrolyze to insoluble hydroxides. After silver, tin, and europium are removed, samarium constitutes 54% of remaining fission products and contributes 2.9% of remaining radiotoxicity. It could be removed by further titration using sulfate. After silver, tin, europium, and samarium are removed, only nonradioactive yttrium (95.4%), indium (0.41%), germanium (0.13%), and microgram quantities of others remain, along with antimony (4.1%). Reagent-grade YCl_3 has a value of about $5,000 per tonne of 5.2%-burnup fuel. ^{125}Sb half life is 2.759 years, and thermal power is 5.2 watts. Their trichlorides can be separated by distillation or crystallization.

If barium and strontium are not effectively removed by filtering and distillation, they will appear in electrolyte. Strontium should be separated, for reasons above described. There is no point to include innocuous barium in a repository. Barium can be separated from other fission-product chlorides as nitrate: $Ba(NO_3)_2$ solubility in water is about 1/8 the solubility of $Sr(NO_3)_2$, and about 1/15 the solubility of $Eu(NO_3)_2$. Other fission-product nitrates are more soluble. This difference is not dramatic, so several steps might be necessary. As remarked above, europium can be removed as $EuSO_4$, but care must be taken not to remove strontium at the same time; their solubilities are different by a factor of 73. Finally, strontium can be removed as $SrSO_4$. Its solubility is less than sulfates of remaining fission products by a factor of 2,500. Barium should be removed as nitrate before europium and strontium because the solubility of $BaSO_4$ is a factor of 46 less than that of $SrSO_4$. It might be possible to remove those three, and samarium, separately as sulfates, by careful sequential titration.

Waste forms that produce less heat can be stored more densely, which would be less expensive than the cost to store a much larger mineral waste form that includes caesium, europium, strontium, and tellurium [21]. Excluding those metals, radiotoxicity and thermal power decline on human, not geologic, time scales. Remove waste forms periodically from high activity, low density, high cost storage to low activity, high density, low cost storage, and ultimately dispose them, thereby reducing the volume and total system cost of storage.

6 Alternatives to finely-divided particles

For a cylinder of radius r and height h, the surface-to-volume ratio is $\frac{2}{r} + \frac{2}{h}$. For fixed h, it increases for small r, i.e., fine wires; for fixed r, it increases for small h, i.e., thin sheets.

One alternative is to fabricate fuel slugs as bundles of fine wire, with sodium in the interstices. The maximum density is $\frac{\pi}{\sqrt{12}} \approx 91\%$, assuming uniform size and no fixed boundary. Smaller wires or a hexagonal container reduce container boundary effects. As is the case for particles, increased surface-to-volume ratio allows gases to escape rather than forming sealed pores and causing swelling. Gases would migrate longitudinally rather than remaining in the bundle. This might be subverted by fuel ductility allowing cylindrical wires to deform to hexagons, filling the interstices, sintering to each other, and trapping gases. Textured rather than smooth wires would reduce this, and increase surface-to-volume ratio, but would reduce fuel density. Swelling should be less than for solid metal.

A second alternative is to fabricate sheets with quasi-random texture. Punch disks having notches in their edges, and stack with sodium between them to form a slug. Choose texture to produce desired fuel density, and to leave channels even after disks weld or sinter where they contact, allowing gases to escape, and fission products to diffuse into sodium. Gases would migrate horizontally between disks, and then vertically through edge notches. Swelling should be less than for solid metal, and essentially all in the axial direction.

In either case, bond sodium and its dissolved fission products could not be removed simply by filtering as effectively as would be the case for particles. It should be possible to remove significant amounts by rolling extracted fuel to thin sheets, expelling bond sodium and its dissolved fission products, followed by distillation as above. Pyroelectric processing of thin sheets would proceed faster than processing solid slugs.

A third alternative is to fabricate rigid compacted sintered-powder fuel slugs, provided this allows gases to escape to the plenum, and they do not swell significantly. Sintering creates a rigid but not solid material – there are interstitial voids. Sodium rather than inert gas should fill the voids. It is possible to comminute sintered spent fuel to fine powder because it is brittle [25, p. 2], allowing the benefits for processing as particles to accrue.

7 Experiments

Several facets must be examined by experiment under normal operating conditions, or in a facility such as the proposed Versatile Test Reactor, to verify that the method works and is beneficial, and to optimize it if so.

- Determine whether, or to what extent, fuel particles self sinter.
  - Determine whether immediately depositing particles created by spraying, or powder created from hydride decomposition, into sodium, reduces self sintering.
  - Determine the effect self sintering has on fission gas migration to the plenum.
  - Determine the extent to which rigid sintered fuel slugs, with sodium rather than inert gas in the interstices, allow gases to escape, and prevent swelling.
  - Determine the effect self sintering has on the ability to remove spent fuel from fuel pins.
  - Determine whether a thin particle coating, such as graphite or polysiloxane, reduces self sintering. If it reduces sintering, determine whether it inhibits diffusion of gases and metallic fission products into bond sodium, and whether it interacts detrimentally with cladding or bond sodium.
- Determine the density that can be attained as a function of particle size distribution. Too-small particles might self sinter, even at low temperature, and create a high-friction high-viscosity low-density slurry, and eventually rigid rather than motile fuel within the pin. Too-large particles with narrow size distribution result in swelling, and low density due to container boundary effects. The smallest possible average particle size should be used, to maximize fission product release and minimize container boundary effects.
- Determine the melting point of spent fuel particles and whether tellurium can be distilled from them without melting, as a function of burnup.
- Determine the distribution of sizes of sealed fission-gas pores, the distribution of pores that break open at the surface, and fuel-particle swelling, as a function of particle size distribution and burnup.
- Determine the extent to which fuel, from which gases and low boiling point metallic fission products have been separated, can be reused without complete pyroelectric processing, as a function of burnup.
- If metallic powder is produced by decomposition of hydrides of spent fuel that has not been pyroelectrically processed, determine whether lanthanide hydrides remain, and whether their presence in fuel as hydrides rather than elemental metal is detrimental.
- Determine whether the reduction of mechanical stress allows to improve durability of cladding using a coating, such as polysiloxane, graphite, or electroplated zinc.
- ~~Determine the bulk thermal conductivity of metallic fuel particles mixed with bond sodium, and assess the effect of any significant difference from bulk thermal conductivity of solid metal fuel, for purposes of passive cooling and inherent safety.~~

  The thermal conductivity can be estimated using Equation (6.4) from [28]:

  $$\frac{\kappa_p}{\kappa_{Na}} = 1 + 3\beta\phi + 3\beta^2\phi^2\left(1 + \frac{9\beta}{16(\beta+5)} + \frac{\beta}{4} + \frac{\beta^2}{2^6} + \dots\right) + O(\phi^3)\,, \tag{1}$$

  where where $\alpha = \kappa_f/\kappa_{Na} \simeq 0.556$ and $\beta = (\alpha - 1)/(\alpha + 2) \simeq -0.314$. With ϕ, the volume fraction occupied by metallic fuel, ranging from 0.6 to 0.95, κ_p declines nearly linearly from about 40 to about 36 W/m/K. The thermal conductivity of uranium is $\kappa_f \simeq 35$ W/m/K.
- Determine whether gases migrate longitudinally within wire bundles, or remain in place and cause the bundle to swell.

- Determine whether this process, or some subset of the steps in this process, reduces the total system cost, from fuel production, through operation and spent fuel processing, to fission product storage.

8 Metallic fuel for LWR

As described in [38], [26], and [40], metallic fuel has significant advantages. It is not possible to use metallic fuel with sodium thermal bond in a water-cooled reactor. Even a pinhole leak would cause a fire in the coolant and a large rupture would cause an explosion. Although a fire can be easily controlled in a storage pond, by removing the leaking object, it is not feasible to control it within an operating reactor. A possibility that ought to be investigated in this context is to use zinc for thermal bond in water-cooled reactors. Zinc molar density (0.1027 mol/cc) is 2.43 times sodium molar density (0.0422 mol/cc) and zinc thermal neutron absorption cross section (1.1 b) is 2.07 times sodium cross section (0.53 b). Neutron absorption in zinc is 5.04 times greater than in the same volume of sodium. Greater fuel density allowed by using metallic rather than ceramic fuel might compensate for this.

The filtering and distillation steps before pyroelectric refining also work if the thermal bond material is zinc. Like sodium, zinc has no long-lived activation products, the longest-lived being $^{65}\mathrm{Zn} \xrightarrow[244.1\mathrm{d}]{\epsilon} {}^{65}\mathrm{Cu}$. ~~If g~~Group Ia ~~and IIa~~ metals do not dissolve into zinc thermal bond, so separating them by filtering is not possible~~, and barium cannot be separated easily~~. Barium, gallium, germanium, indium, silver, strontium, and tin form alloys with zinc [18] [27]. After separating zinc, a hydride-dehydride process would allow to dissolve group Ia metals into sodium.

Beyond the safety advantages, metallic fuel is denser, and easier and less expensive to process than oxide fuel, as summarized in Table 4.

Table 4: Costs for reprocessing [10]

Plant	Area Hectares	Maximum Capacity T/yr	Capital Cost $US	Operating Cost ¢/kWh
Rokkasho	380	800	21 billion	0.52
THORP	285	900	6.3 billion	0.16
La Hague	300	1,700	18 billion	0.15
Barnwell	113	1,500	1.2 billion	0.081
Pyroelectric [3] [14]	17	400	900 million	0.05

Rokkasho, La Hague, THORP, and Barnwell all use solvent refining processes, either PUREX or derivatives of it. THORP was closed because of a leak that could not be repaired. Barnwell never operated. Rokkasho is projected to open in 2022.

Operating costs do not include capital amortization, decommissioning, or waste storage.

American utilities contributed 0.1 cents per kWh to the Nuclear Waste Fund [15] from 1983 until 2014, when courts held that utilities need not continue to pay into the fund because DoE had not accepted its legal responsibility to take custody of spent fuel. The fund now stands at $43.4 billion.

9 Cumulative fission-product production

The cost to store fission products depends upon their amounts, the amount of heat they generate, and the required custody duration. This section explains how to calculate total amounts of fission products. The next section analyzes thermal production.

For a two-step decay process $x \to y \to z$ with z stable, the Bateman equations [7] for x and y are

$$
\begin{aligned}
\frac{\mathrm{d}x}{\mathrm{d}t} &= k_x - a_x x \\
\frac{\mathrm{d}y}{\mathrm{d}t} &= k_y + \rho_{xy} a_x x - a_y y\,,
\end{aligned}
\tag{2}
$$

where k is production rate, $a = \frac{\ln 2}{\lambda}$ is activity, λ is half life, and ρ_{xy} is the branching ratio from decay of x to production of y (usually 1.0). Equations for additional steps in longer decay chains are similar to the equation for y. Software for numerical solution of the general case was described in [35].

With $x(0) = 0$ and $y(0) = 0$, the solutions for Equations (2) are

$$
y(t) = \frac{k_y}{a_y}\left(1 - e^{-a_y t}\right) + \frac{k_x \rho_{xy}}{a_y}\left(1 + \frac{a_y e^{-a_x t} - a_x e^{-a_y t}}{a_x - a_y}\right), \tag{3}
$$

where results for x are the same as for y, mutatis mutandis, and with $\rho = 0$.

One would expect that modeling fission and reactor operation in detail is necessary to calculate k. If one assumes, however, that k is roughly constant if fission conditions are roughly constant, Equations (2) can be solved for k if we have amounts at a specific time (other than $t = 0$):

$$
k_y = \frac{1}{1 - e^{-a_y\, t}}\left[a_y\, y(t) - k_x \rho_{xy}\left(1 - \frac{a_x e^{-a_y t} - a_y e^{-a_x t}}{a_x - a_y}\right)\right]. \tag{4}
$$

After fission ceases at time t, thereafter $k = 0$ and only decay continues. The solutions for Equations (2), at time $t + \tau$, become

$$
y(t+\tau) = y(t)\, e^{-a_y \tau} - a_x \rho_{xy}\, x(t)\, \frac{e^{-a_x \tau} - e^{-a_y \tau}}{a_x - a_y}\,. \tag{5}
$$

If $a_x \approx a_y$, evaluating the expressions in which $a_x - a_y$ appears in the denominator in the obvious way produces results with large relative error. This can be reduced by replacing $a_x = \sigma + \delta$ and $a_y = \sigma - \delta$, where $\delta = \frac{1}{2}(a_x - a_y)$ and $\sigma = \frac{1}{2}(a_x + a_y)$. Then

$$
\frac{e^{-a_x \tau} - e^{-a_y \tau}}{a_x - a_y} = \tau e^{\sigma\tau} \frac{\sinh\,\delta\tau}{\delta\tau} \text{ and } \frac{a_x e^{-a_y t} - a_y e^{-a_x t}}{a_x - a_y} = e^{\sigma\tau}\left(\cosh\,\delta\tau + \sigma\tau \frac{\sinh\,\delta\tau}{\delta\tau}\right). \tag{6}
$$

Maclaurin series expansions

$$
\frac{\sinh\,\delta\tau}{\delta\tau} = 1 + \frac{\delta^2\tau^2}{6} + O(\delta^4\tau^4) \text{ and } \cosh\,\delta\tau = 1 + \frac{\delta^2\tau^2}{2} + O(\delta^4\tau^4) \tag{7}
$$

show that the relative error in computing these expressions for small $\delta\tau$ is on the order of the square of the absolute error in computing $\delta\tau$.

10 Thermal calculation

Having amounts at specific times after fission ceases, we will be concerned about thermal power production, the maximum decay thermal power production, and the time of maximum decay thermal power production.

The power production from all parent-daughter isotope decay chains $x_1 \rightarrow y_1 \rightarrow z_1$, $x_2 \rightarrow y_2 \rightarrow z_2$, … for all radioactive isotopes of a particular element x is

$$P_x(\tau) = p_{x_1} x_1(\tau) + p_{y_1} y_1(\tau) + p_{x_2} x_2(\tau) + p_{y_2} y_2(\tau) \ldots , \tag{8}$$

where p is specific power production (watts per mole) from each isotope's decay.

For strontium, there is only one thermally significant decay chain: ${}^{90}\mathrm{Sr} \xrightarrow[28.79\mathrm{y}]{\beta} {}^{90}\mathrm{Y} \xrightarrow[2.671\mathrm{d}]{\beta} {}^{90}\mathrm{Zr}$.

For caesium, there are two: ${}^{134}\mathrm{Cs} \xrightarrow[2.065\mathrm{y}]{\beta} {}^{134}\mathrm{Ba}$, and ${}^{137}\mathrm{Cs} \xrightarrow[30.04\mathrm{y}]{\beta} {}^{137m}\mathrm{Ba} \xrightarrow[2.552\mathrm{m}]{\gamma} {}^{137}\mathrm{Ba}$.

For europium there are five: ${}^{150}\mathrm{Eu} \xrightarrow[36.36\mathrm{y}]{\epsilon} {}^{150}\mathrm{Sm}$, ${}^{152}\mathrm{Eu} \xrightarrow[13.52\mathrm{y}]{\epsilon} {}^{152}\mathrm{Sm}$ ($\rho = 0.721$), ${}^{152}\mathrm{Eu} \xrightarrow[13.52\mathrm{y}]{\beta} {}^{152}\mathrm{Gd}$ ($\rho = 0.279$), ${}^{154}\mathrm{Eu} \xrightarrow[8.593\mathrm{y}]{\beta} {}^{154}\mathrm{Gd}$, and ${}^{155}\mathrm{Eu} \xrightarrow[4.753\mathrm{y}]{\beta} {}^{155}\mathrm{Gd}$.

For a single-step decay process, such as for ${}^{125m}\mathrm{Te} \xrightarrow[57.40\mathrm{d}]{\gamma} {}^{125}\mathrm{Te}$, or if $\frac{\mathrm{d}P}{\mathrm{d}\tau}\big|_{\tau=0} < 0$, maximum power production occurs at $\tau = 0$. For a multi-step process with $\frac{\mathrm{d}P}{\mathrm{d}\tau}\big|_{\tau=0} > 0$, maximum decay thermal power production occurs for $\tau > 0$ because the daughter accumulates fast enough, and produces enough heat, to increase total heat production initially. Then the daughter falls into equilibrium with the parent. To calculate the instant τ of maximum heat production, derivatives are needed. Fission has ceased; $k = 0$, and $x(t)$ and $y(t)$ are amounts at the time t when fission ceased. Substituting Equations (5) into Equations (2), we have

$$\frac{\mathrm{d}y}{\mathrm{d}\tau} = -a_y y(t)\, e^{-a_y \tau} + a_x \rho_{xy}\, x(t)\, \frac{a_x e^{-a_x \tau} - a_y e^{-a_y \tau}}{a_x - a_y} . \tag{9}$$

A good numerical method to solve for τ such that $\frac{\mathrm{d}P}{\mathrm{d}\tau} = 0$ is a Newton iteration:

$$\tau_{n+1} = \tau_n - \left.\frac{P'_x}{P''_x}\right|_{\tau_n} \tag{10}$$

for which we need

$$\frac{\mathrm{d}^2 y}{\mathrm{d}\tau^2} = a_y^2\, y(t)\, e^{-a_y \tau} - a_x \rho_{xy}\, x(t)\, \frac{a_x^2\, e^{-a_x \tau} - a_y^2\, e^{-a_y \tau}}{a_x - a_y} . \tag{11}$$

As above, if $a_x \approx a_y$, this can be evaluated without significant relative error using

$$\frac{a_x^2\, e^{-a_x \tau} - a_y^2\, e^{-a_y \tau}}{a_x - a_y} = e^{\sigma\tau} \left(2\sigma \cosh \delta\tau + \delta \sinh \delta\tau + \sigma^2 \tau \frac{\sinh \delta\tau}{\delta\tau} \right) . \tag{12}$$

Using the amounts from Table 1, and activity constants a and branching ratios ρ taken from JEFF 3.11 data [30], production constants k were calculated for cadmium, caesium, europium, strontium, and tellurium. Having k, a, and ρ, the total amounts of those elements were calculated for sixty years of operation at 1.0 GWth, assuming the same fuel replacement schedule (50.689 GWth-days per tonne), followed by ten years' storage before final spent-fuel processing. Specific power production (watts per mole) for each significant isotope was calculated from ORIGEN-2 output [2]. Assuming the elements in Table 5 are separated from daughters and metals collected with them, after final processing, and then stored without further processing, the time τ of maximum power output, and that maximum, were calculated.[7] Equations (5) and (8) show that decay thermal power declines exponentially after the maximum.

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[7] Software and data are available from the author.

Radiotoxicities were calculated using adult ingestion dose factors from ICRP publication 119 [17].

Table 5: **Fission products from 60 years' operation at 1 GWth in fuel processed after ten years' storage**

	Mass	Volume	Maximum Power			Per liter after specified storage		
	kg	Liters	Time τ	Watts	Per liter	Duration	Watts	Radiotoxicity
Cs	1355	702.1	37.34 mins	334.0 kW	391.7 W	300 years	66.53 mW	25.73 kSv
Sr	365.2	138.3	14.66 days	232.2 kW	1649 W	300 years	180.5 mW	138.6 kSv
Eu	85.88	16.36	0 mins	7.467 kW	455.3 W	100 years	142.8 mW	1.178 kSv
Cd	63.95	7.359	0 mins	8.326 kW	1131 W	30 years	801.4 mW	401.9 kSv
Te	1.458	0.2337	0 mins	102.7 W	439.6 W	5 years	154.7 μW	5.510 Sv
Europium would probably be stored as 139.8 kg EuSO$_4$, 27.14 liters, 83.53 mW/l after 100 years.								

Required custody durations can be estimated from decay curves calculated by the author (and many others) as shown in Figure 4 (page 21). ^{125m}Te is not shown because its half life is only 57.40 days, and its decay product is stable. It is a decay product of ^{125}Sb, which has a half life of 2.759 years. One tonne of ten-year-old fuel contains only 1.610 grams of ^{125}Sb. The relationship to uranium in nature was taken from [41].

Figures per liter in Table 5 assume fission products are not separated from daughters, and do not account for volume changes due to different densities of daughter products. Caesium, europium, and strontium decay to denser daughters, and can therefore be stored in containers without a void space. Cadmium and tellurium decay to less dense daughters, and therefore would require a void space for expansion within their containers.

11 Conclusions

Using finely divided metallic fuel particles produces the following benefits:

- Higher fuel density, both initially and after irradiation, improves neutron economy and the internal conversion ratio, makes it easier to maintain a hard neutron spectrum, and allows lower fissile content in the initial fuel load.
- More fission gases diffuse from particles to plenum, and cause less fuel swelling.
- More fission product metals diffuse to bond sodium and cause less swelling, which allows simple separation from fuel.
- Less cladding stress and essentially eliminated cladding strain allows greater burnup.
- A sequence of simple thermo-mechanical processing steps reduces fuel-cycle costs because fuel can be diverted directly back to service, at any stage, several times, without costly chemical or pyroelectric processing.
- Fewer fission products, with lower specific heat production and less radiotoxicity, in electrolyte, and then sequestered in mineral waste forms, reduces long-term storage cost.
- Innocuous nonradioactive barium, which would otherwise be 52 wt.% of fission products in electrolyte after pyroelectric processing, is removed from the storage stream, which reduces long-term storage cost.
- Caesium and strontium, the two most radiotoxic fission products, with the highest long-term specific heat production, the longest duration of custody, and presumably the highest storage cost per liter, are separated from other fission products as high-density relatively pure metals. This results in lower storage cost for other fission products, and lower total storage costs
- Expense and complexity of the processes ought to be offset by lower total system costs.

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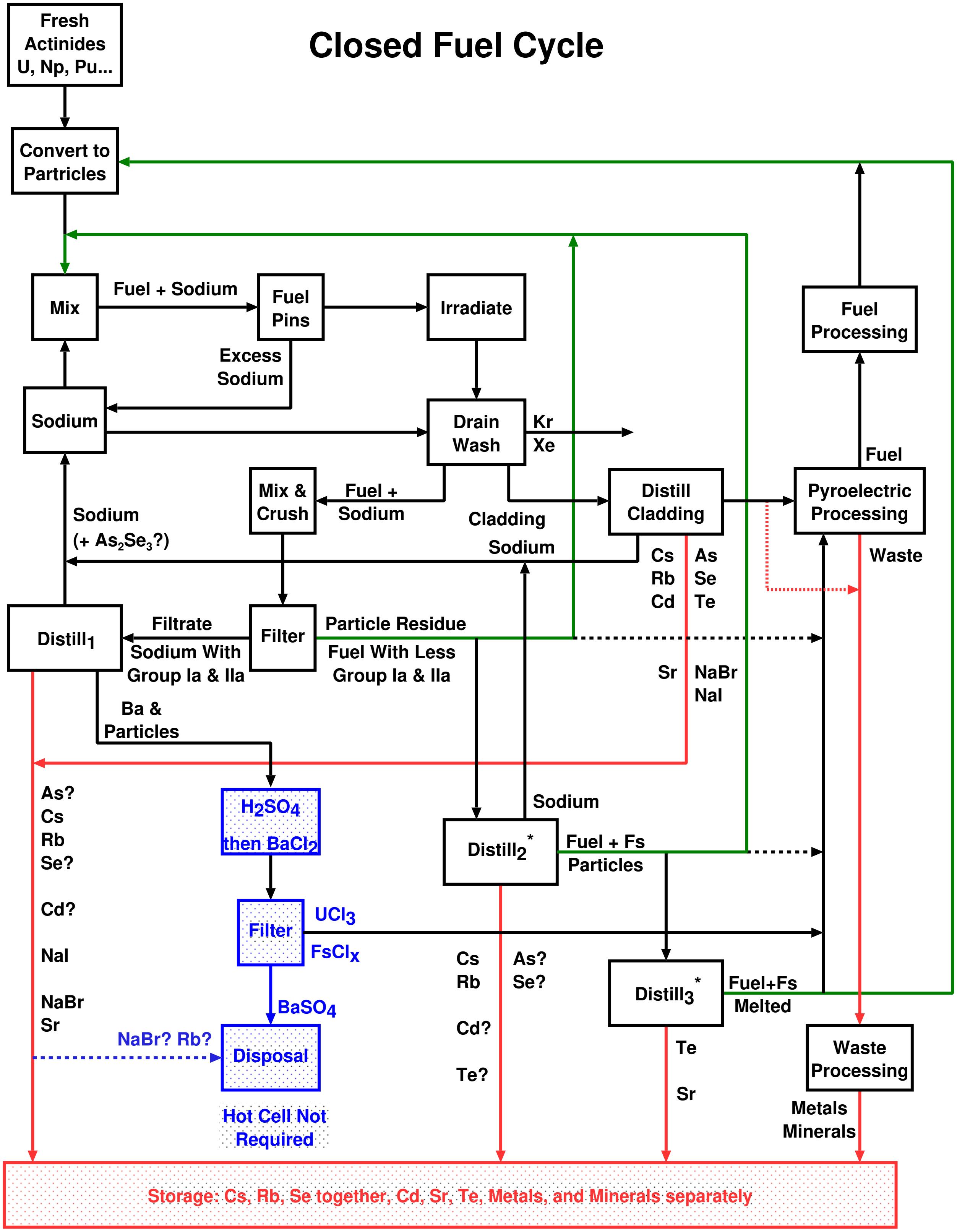

*Distill$_2$ and Distill$_3$ do not separate barium

Figure 3: Closed Fuel Cycle

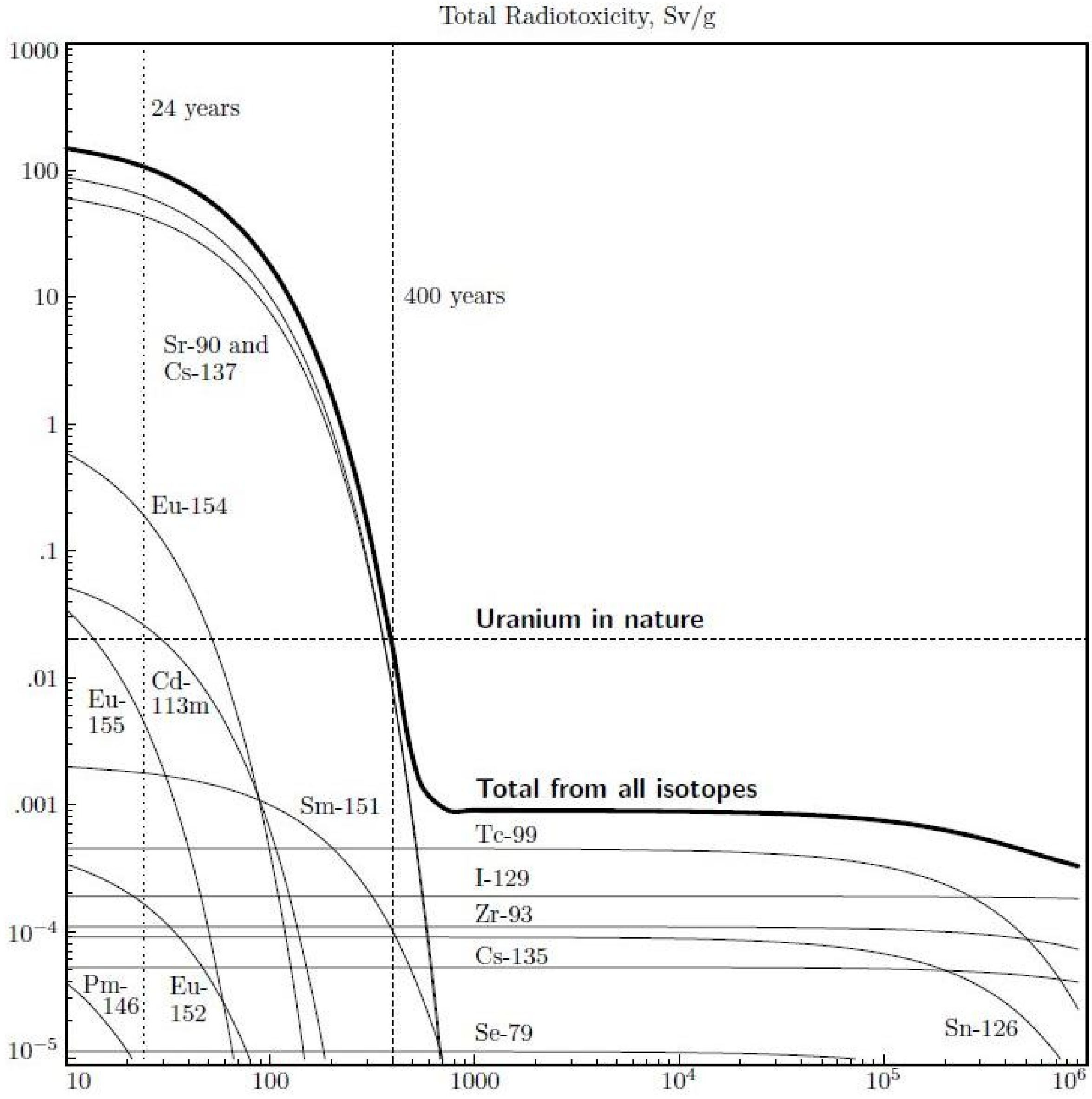

Figure 4: Radiotoxicity of Fission Products Compared to Uranium in Nature