Finely-Divided Metal as Nuclear Power Reactor Fuel

Abstract

We propose a new form for metallic nuclear reactor fuel, consisting of finely-divided particles mixed with sodium for thermal bond. This results in greater fuel density than with solid slugs fabricated at 75% initial smear density. 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, a simple preliminary spent-fuel processing step, and the possibility to postpone pyroelectric processing until several cycles of burnup and preliminary processing. Less frequent pyroelectric processing, simple preliminary processing, and larger surface-to-volume ratio reduce total processing cost. Preliminary processing produces separate fission products, in metallic rather than salt or mineral form, in particular producing caesium and strontium separately, 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.

1 Why metallic fuel?

Plentiful Energy [21], *Metallic fast reactor fuels* [15], *The safety of the IFR* [22], and *Treatment of Wastes in the IFR Fuel Cycle* [5] explain the benefits of metallic fuel with sodium thermal bond, for reactor safety and spent fuel processing.

2 Irradiated fuel

Hofman et al [15, p. 90] reported that EBR-II metallic fuel slugs are prepared having an initial smear density of 75%, or about 85% of the inner diameter of fuel pins. When fuel slugs are inserted into fuel pins, sodium is added to provide a thermal bond to cladding, and to allow for swelling. Irradiated fuel expands radially, and makes contact with fuel pin cladding after 1.6% burnup. Axial expansion is less than 8% [15, p. 91] in U-Fs and U-Zr fuels, and 4% for U-Pu-10Zr fuel [7, p. 162]. Expansion is driven by gases that expand into bubbles that eventually connect, resulting in sealed voids within fuel slugs. Figure 8 in [15, p. 94] and Figure 6-2 in [21, p. 125] show the size of those voids is in the range of 5–25 μm, at least in the γ phase.

The change in sodium level above irradiated fuel is not sufficient to account for radial swelling. This deficit is attributed to sodium logging. Figure 9 in [15, p. 95] shows that thermal conductivity abruptly increases at about 1.6% burnup. The associated text explains that this is also due to sodium logging.

Sodium logging occurs when pores break open at the surface of fuel, gases escape, and sodium intrudes. Expansion 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, creating larger surface area, which allows gases to escape to the plenum more efficiently.

Table 1 in [15, 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), are found in bond sodium. Other group Ia fission products – lithium, rubidium, and francium – are not mentioned, but presumably also dissolve in sodium. Whether other fission products, in particular arsenic, cadmium, selenium, and tellurium, dissolve into bond sodium, is not mentioned.

3 Finely-divided fuel

If the surface-to-volume ratio were larger, more fission-product gases would diffuse directly from the surface of fuel instead of creating voids, and more metallic fission products would diffuse into bond sodium. 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 voids would reach the surface, resulting in less swelling. The interior surface of open voids would further increase the surface area of fuel in contact with bond sodium.

For thermal bond, and to dissolve metallic fission products, sodium can be used in the same way as with solid fuel slugs.

In EBR-II, slugs were fabricated with 75% initial smear density. Axial swelling was 2-8%, depending upon alloy and burnup [15, p. 91] [7, p. 162]. Volume density was 69–73% after swelling.

Assuming spherical particles of uniform size, and no container boundary, optimum packing density is $\frac{\pi}{\sqrt{18}} \approx$ 74%. Scott and Kilgour [19] 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 into the interstices between large particles. For two sphere sizes, Yamada et al [23] found RCP density of 74.1% would occur in 4.4 mm diameter fuel pins if larger particles were 0.28 mm in diameter and smaller particles were 0.02 mm in diameter. Farr and Grote [12] report RCP densities from 64% to 97% for polydisperse spheres of log-normal distributions with $0 < \sigma < 3$. Finely-divided fuel can be fabricated with initial fuel density that is greater than with solid fuel slugs fabricated at 85% of final diameter.

If more gases and metallic fission products escape into bond sodium, fuel particle swelling would be less. Rather than being caused by trapped gases, swelling would largely be due to precipitates of rare earth and some noble metals, and metals in solution in the alloy [15, pp. 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. Fuel density should remain comparable to or greater than the density with solid fuel slugs.

The primary limitation to fuel burnup is fuel pin cladding durability, and bundle-duct interaction due to radial fuel pin strain [21, p. 116] [15] [20, p. 2]. Hofman et al [15] discuss fuel-cladding mechanical interaction (FCMI) and fuel-cladding chemical interaction (FCCI). Fuel composed of small particles and bond sodium would not, in itself, put significant radial pressure on fuel pin cladding. Expansion driven by swelling would all be in the axial direction. Thermally-driven axial expansion produces negative reactivity feeback and attendant important safety advantages [7, 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. This can be ameliorated by a longer plenum, or by connecting the plenum to an exhaust port. 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 cladding, allowing greater burnup before fuel processing.

Small particles can be created by spraying molten fuel through a nozzle into a tower containing inert gas or sodium vapor, and a sodium pool at the bottom. This would result in polydisperse particle sizes, probably with log-normal distribution. The distribution of particle sizes can be controlled by nozzle geometry, fuel temperature, and flow rate. The pool can be cooled to prevent boiling, or sodium vapor can be drawn off, cooled, and condensed externally, then fed back into the pool. Pool Geometry and batch size can control criticality.

Another method is to 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 [14]. 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 sinter because of high surface reactivity. The result is brittle and can be easily mechanically comminuted to the desired powder size [14, p. 2]. This powder can then be mixed with sodium to make a slurry. 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 by hot compacting[1]. Sintering creates a rigid material. Notley et al [17] report preparing plutonium powder using a hydride process. Haschke et al [13] report that plutonium hydride behaves similarly to lanthanide fluorides. This might be true of other actinides as well. Zirconium hydride decomposes at 300°C [1]. Lanthanide hydrides, among the most stable hydrides [6], decompose at about 800°C. It might be possible to prepare purely-metallic powders of partially processed spent fuel using a hydride process. If prepared above liquid sodium, NaH will form, depending upon hydrogen pressure; NaH is soluble in liquid sodium. Therefore,

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

powder should be created from hydride in vacuo, and then comminuted to the desired sizes and mixed with sodium.

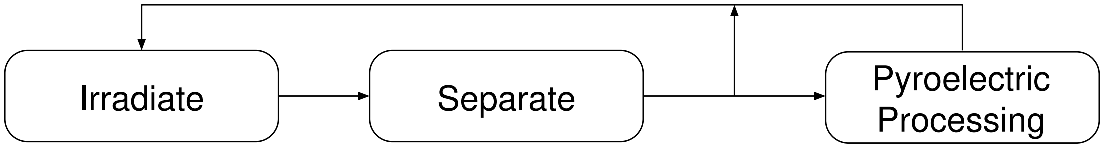
A stirred heated slurry can be drained, poured, or pumped into fuel pins, or by vacuum casting directly into fuel pins from the bottom of a settled rather than stirred sodium pool. Because it is only necessary to melt the bond sodium, not the fuel carried in the slurry, this process would use a much lower temperature than vacuum casting solid fuel slugs. It would not be necessary to use quartz molds, or to destroy molds to extract fuel slugs.

Fuel inserted into pins can be settled by heating, with vibration to remove voids. Because of the enormous difference in density between sodium and fuel, this should proceed rapidly. If fuel particles bond due to uranium's self reactivity, preventing settling, bonds can be broken by ultrasound, or by inserting a rod into the fuel pin. Excess sodium can be distilled from fuel pins, or decanted from above settled fuel. If it is decanted into the spray tower sodium pool, it does not matter if it contains fuel particles.

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

Spent fuel can be processed 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. It is likely that irradiation and the first step can be repeated several times before the second step is necessary.



The Sodium Waste Treatment program [4, p. 55] determined that sodium and solid fuel could be removed from EBR-II cladding by cutting the bottom end cap and heating the fuel pin. Finely divided fuel that has sintered is brittle [14, p. 2] and can be broken by rolling the fuel pin between three rotating cylinders. If the bottom end cap is removed first, plenum pressure helps to expel fuel. Residual particles can be washed out by sodium, or a boiling fluid such as argon. Cavitation from boiling would remove particles that are not strongly bonded to cladding. Fuel pin cladding can then be cleansed by distillation to remove sodium and volatile fission products, and then processed pyroelectrically, if necessary, to separate remaining spent fuel and activation products. Pyroelectrically processing cladding separately 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. Separately-processed cladding might have sufficiently low activity that it does not need storage; it can be immediately disposed or recycled.

4.1 Filtering

If conglomerates remain in removed fuel, they can be crushed to yield smaller particles. Bond sodium and its dissolved fission products can then be separated from fuel particles by filtering.

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

A sintered-glass filter can be cleansed by back-flowing hydrochloric acid after residue is removed. Resulting chlorides can then be dried and added to electrolyte during pyroelectric processing. Back-flowing would reduce accumulation of insoluble AgCl. A sintered-fuel filter can be crushed and processed with fuel particles.

Washing and filtering should proceed at high temperature to reduce sodium viscosity and increase sodium halide solubility [18]. The advantage of washing and filtering is that barium and strontium dissolve in sodium [15]. If filtering is not done, 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. Sodium also remains with the fuel, and must be removed by subequent distillation, or it will eventually be incorporated into the mineral waste form, needlessly increasing its volume.

Hofman et al [15] do not discuss whether other metals, such as cadmium and chalcogenides, dissolve into bond sodium. 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: Filtrate composed of sodium and dissolved fission products, and fuel particle residue. Both are processed 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. Depending upon fuel particle sizes and filter porosity, the residue might contain small amounts of fuel. Barium and fuel can be separated as sulfates. Actinide and other fission product sulfates are much more soluble in water than barium sulfate – 400,000 times more for uranium. Dissolved sulfates – fuel and fission products other than barium – can be converted to chlorides using barium chloride, which will be converted to insoluble barium sulfate. The chlorides can then be dried and added 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.

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

Cont.

Low boiling point fission products from 50.689 GWth-day LWR burnup

Element	Mass Grams	Moles	Boiling Point °C	Element	Mass Grams	Moles	Boiling Point °C
Total	9611 grams, 78.17 moles, excluding sodium metal and intermetallic compounds						

The amounts of low boiling point fission products given here are per tonne (1000 kg) of spent LWR fuel, according to output from the ORIGEN 2 computer program [2] [9]. 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 IFR fuel, the amounts would be greater, but the proportions roughly the same [8].

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.

Arsenic, cadmium, selenium, and tellurium might be present as intermetallic compounds, not separate metals. Cadmium forms alloys with plutonium [11, p. 25]. Till and Chang report that cadmium can be distilled from fuel [21, pp. 171, 176] [20, p. 3]. 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 [15] is about 990°C [11, p. 184]. With constant zirconium content, increasing plutonium content lowers 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 above metals 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.

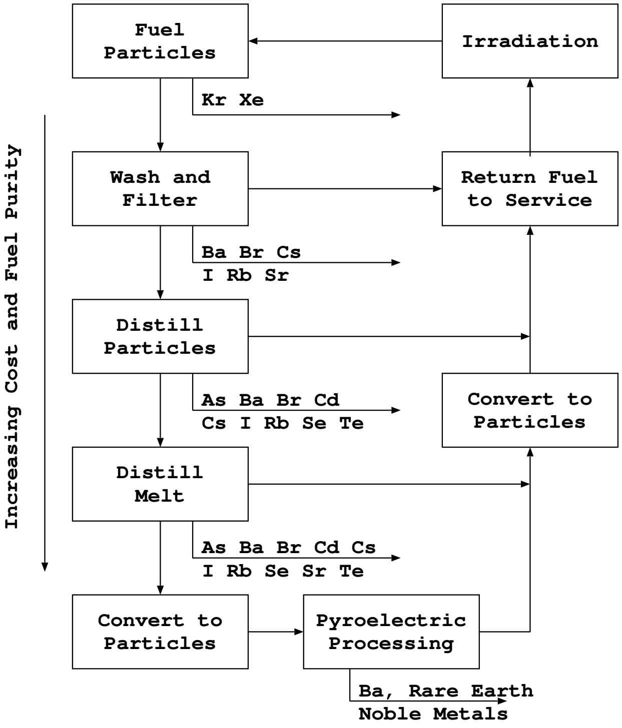
This suggests two regimes for treating particulate filtering residue:

1. Distill arsenic, caesium, rubidium, selenium, and As_2Se_3, and cadmium to the extent it is not bound in high boiling point chalcogenide intermetallic compounds that do not decompose, from fuel particles. It might be possible to distill significant amounts of elemental tellurium without melting fuel particles because its amount is much larger than the amounts of metals with which it might form compounds.
2. Distill higher boiling point metals from molten spent fuel. If strontium is 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. Low density metals and intermetallic compounds that do not dissolve in fuel, or have a higher melting point, might float atop molten fuel. This is similar to the pyrometallurgical process used at EBR-II, and abandoned when pyroelectric processing was developed [21, pp. 25, 168] [20, p. 6].

   If strontium is not effectively removed by diffusion into bond sodium, it should be distilled from molten fuel, to separate it from innocuous barium

Figure 1: Closed Fuel Cycle



before pyroelectric processing, so that strontium is not included in the mineral waste form, even if this stage does not produce immediately re-usable fuel.

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.

Figure 1 shows that fuel processed by any of these steps could be immediately diverted back to service without costly pyroelectric processing. An advantage of reusing particulate fuel, without melting it, is that particles' surface areas are increased by pores opening. The selection of fuel pins or fuel assemblies from which fuel can be diverted at each stage can be determined from models, analyses, and processing records. Figure 2 (page 16) shows the process in more detail.

Selenides and tellurides of strontium and barium have melting points above 1600°C. Boiling and melting point data for arsenides, selenides, and tellurides of sodium, caesium, and rubidium, are not available.

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

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

Ideally, caesium should be separated from rubidium because 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. They can be separated using potassium aluminum sulfate ($KAl(SO_4)_2$). If not separated, the volume of arsenic, caesium, francium, selenium, and rubidium together would be about 19% greater than the volume of caesium alone, and the specific heat production 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.

4.3 Crystallization

The relationships between the melting points of the fission products described above are different from the relationships of their boiling points. Elements and compounds that do not dissolve in sodium can be removed by cooling and filtering filtrate, or using a cold plate.

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

Cont.

Melting points of some fission products

	Mass Grams	Boiling Point °C		Mass Grams	Boiling Point °C
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				

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.

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

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 distillate tray, provided they do not form an alloy. If As_2Se_3 does not cause a problem in fuel, sodium with small amounts of As_2Se_3 can be used as thermal bond.

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. This monograph does not concern disposing excess sodium, so calcining is not considered.

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, 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 [21, 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.

Fission product gases and the above fission products constitute 34.3% by weight of fission products. With those removed, it might be possible to re-insert fuel particles and sodium into cladding, several times, before pyroelectric processing is necessary. This would reduce processing expense. If increased burnup is made possible by changes in cladding material durability resulting from reduced FCMI, this benefit might not accrue, but increased burnup in itself reduces processing expense.

With the above metallic fission products removed, pyroelectric processing would proceed at a faster rate. If fuel is not melted to distill 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 concentration of remaining fission products will be greater. This increases 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 the frequency of cleansing electrolyte. 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. Fuel pin fabrication cost and durability, reprocessing cost, storage cost, and the value of recovered fuel 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 a different component cost. Reducing component costs can be done by improving component processes, or by performing component processes less frequently. The latter is a system effect that does not directly depend upon individual process costs.

The only fission products that require long-term 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.

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" [21, p. 241].

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

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 innocuous.

Till and Chang report that high-nickel containers ought to be impervious for at least twenty thousand years [21, 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. 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. 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.

The boiling points of NaI and strontium are sufficiently different that they can be separated by distillation. Iodine has much lower activity than strontium, and ought to be stored in the mineral form derived from electrolyte cleansing. If NaBr does not damage high-nickel containers, there is no point to separate it from strontium. Storing NaBr with strontium would increase the storage volume by 2.7%. If necessary, it might be possible to separate NaBr and strontium by crystallization. Otherwise, they can be separated as phosphate or fluoride because $Sr_3(PO_4)_2$ and SrF_2 are essentially insoluble in water. These result in salts that might need to be converted to a less dense mineral storage form, unless they can be stored in high-nickel containers without damaging them. Phosphates form glass well. If NaBr is a problem, and only appears 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.

If NaI is separated from the above products, it can be adsorbed into Zeolyte A and combined with Zeolyte-adsorbed fission products cleansed from electrolyte, to store iodine in a mineral form. Including iodine would increase thermal production by 41 ppb and mass by 3%. Nonradioactive NaBr can be disposed.

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/T, and thermal power is reduced 92.6% to 101.9 W/T. 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 separated.

If not separated by distillation, cadmium would remain with noble metals in the anode basket of the pyroelectric refiner. Distilling and storing it separately from those metals reduces the radiotoxicity of that alloy by 20%, to 201.4 kSv/T, the mass by 1.1%, to 18.08 kg/T, and heat production by 1.3%, to 7.5 W/T. Heat production from cadmium is dominated by ${}^{113m}\mathrm{Cd} \xrightarrow[14.10\mathrm{y}]{\beta} {}^{113}\mathrm{In}$. With cadmium removed from the alloy, ${}^{106}\mathrm{Ru} \xrightarrow[1.020\mathrm{y}]{\beta} {}^{106}\mathrm{Rh} \xrightarrow[30\mathrm{s}]{\beta} {}^{106}\mathrm{Pd}$ produces most of the heat (2.66 μW/g) and radiotoxicity (10.9 Sv/g). After another ten years, ${}^{99}\mathrm{Tc} \xrightarrow[214\mathrm{ky}]{\beta} {}^{99}\mathrm{Ru}$ 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, trivalent lanthanides are deposited at a third cathode [3, p. 6]. Assuming actinides are present in electrolyte only below 100 ppm, 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 contributes 624.1 kSv/T, 90% of radiotoxicity, and produces 73.35 W/T, 93.6% of thermal power. Its presence requires custody of the mineral waste form from electrolyte cleansing for about 100 years. Without it, custody would be less than thirty years. Its sulfate is the least soluble of remaining elements, by a factor of 6×10^{-5}. It is almost pointless to remove samarium, because it has a 90 year half life, and therefore low radiotoxicity and heat production, and it is 70% of the remaining fission products, after europium is removed. Antimony half life is 2.759 years, and thermal power is 5.2 watts.

Waste forms that produce less heat can be stored more densely, which would be less expensive than the cost to store a mineral waste form that includes caesium, europium, strontium, and tellurium. Excluding those metals, radiotoxicity and thermal power decline on human, not geologic, time scales. Waste forms can be periodically removed from high activity, low density, high cost storage to low activity, high density, low cost storage, and ultimately disposed, thereby reducing the volume and 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 no fixed boundary. Smaller wires 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. Swelling should be less than for solid metal. 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.

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.

In either case, bond sodium and its dissolved fission products could not be removed simply by filtering. 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 fuel slugs, with sodium rather than inert gas in the interstices, provided this allows gases to escape to the plenum, and they do not swell significantly. It is possible to comminute sintered spent fuel to fine powder because it is brittle [14, p. 2], allowing the benefits for processing as particles to accrue.

7 Experiments

Several facets must be examined by experiment under normal operating conditions 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 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 sintering has on the ability to remove spent fuel from fuel pins.
  - Determine whether a thin coating over particles, such as zinc, has an effect on 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. Too-large particles with narrow size distribution result in swelling, and low density due to container boundary effects. The smallest possible 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 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 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.
- Determine whether gases migrate longitudinally within wire bundles, or remain in place and cause the bundle to swell.
- Determine the wire size that maximizes density within the fuel pin after irradiation.

8 Metallic fuel for LWR

As described in [21], [15], and [22], 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 an explosion. 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 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 instead of ceramic fuel might compensate for this. Beyond the safety advantages, this provides the additional benefits that metallic fuel is denser, and much easier and less expensive to process than oxide fuel. The distillation steps before pyroelectric refining also work if the thermal bond material is zinc. If group Ia and IIa metals do not dissolve into zinc thermal bond, separating them by filtering is not possible, and barium cannot be separated. Like sodium, zinc has no long-lived activation products, the longest-lived being ${}^{65}\mathrm{Zn} \xrightarrow[244.1\mathrm{d}]{\epsilon} {}^{65}\mathrm{Cu}$.

9 Cumulative fission-product production

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

$$\begin{aligned} \frac{\mathrm{d}x}{\mathrm{d}t} &= k_x - a_x x \\ \frac{\mathrm{d}y}{\mathrm{d}t} &= k_y + \rho_{xy} a_x x - a_y y \,, \end{aligned} \tag{1}$$

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.

With $x(0) = 0$ and $y(0) = 0$, the solutions for Equations (1) are

$$y(t) = \frac{k_y}{a_y}\left(1 - e^{-a_y t}\right) + \frac{k_x \rho_{xy}}{a_y}\left(1 + \frac{a_y e^{-a_x t} - a_x e^{-a_y t}}{a_x - a_y}\right), \tag{2}$$

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 $t = 0$):

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

After fission ceases at time t, thereafter $k = 0$ and only decay continues. The solutions for Equations (1), at time $t + \tau$, become

$$y(t+\tau) = y(t)\, e^{-a_y \tau} - a_x \rho_{xy}\, x(t)\, \frac{e^{-a_x \tau} - e^{-a_y \tau}}{a_x - a_y} . \tag{4}$$

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

$$\frac{e^{-a_x \tau} - e^{-a_y \tau}}{a_x - a_y} = \tau e^{\sigma\tau} \frac{\sinh \delta\tau}{\delta\tau} \text{ and } \frac{a_x e^{-a_y t} - a_y e^{-a_x t}}{a_x - a_y} = e^{\sigma\tau} \left(\cosh \delta\tau + \sigma\tau \frac{\sinh \delta\tau}{\delta\tau} \right) . \tag{5}$$

A Maclaurin series

$$\frac{\sinh \delta\tau}{\delta\tau} = 1 + \frac{\delta^2 \tau^2}{6} + O(\delta^4 \tau^4) \tag{6}$$

shows that the relative error in computing these expressions is approximately one third 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 from all parent-daughter isotope decay systems $x_1 \to y_1 \to z_1$, $x_2 \to y_2 \to z_2$, … for a particular element,

$$P(\tau) = p_{x_1} x_1(\tau) + p_{y_1} y_1(\tau) + p_{x_2} x_2(\tau) + p_{y_2} y_2(\tau) \ldots , \tag{7}$$

the maximum decay thermal power production, and the time of maximum decay thermal power production, where p is specific power production (watts per mole) from each isotope's decay.

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

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

For 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 $\tau = 0$. Otherwise, the daughter accumulates fast enough, and produces enough heat, to increase total heat production initially. Then it falls into equilibrium with the parent. For a multi-step process with $\frac{\mathrm{d}P}{\mathrm{d}\tau}\big|_{\tau=0} > 0$, maximum decay thermal power production occurs for $\tau > 0$. To calculate τ, 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 (4) into Equations (1), we have

$$\frac{\mathrm{d}y}{\mathrm{d}\tau} = -a_y y(t)\, e^{-a_y \tau} + a_x \rho_{xy}\, x(t)\, \frac{a_x e^{-a_x \tau} - a_y e^{-a_y \tau}}{a_x - a_y} . \tag{8}$$

If there are only two radioactive isotopes, related by a two-step decay, it is possible to solve $\frac{\mathrm{d}P}{\mathrm{d}\tau} = 0$ for τ (the solution is complicated) and substitute that into Equation (4) (the result is complicated). If there is more than one radioactive parent isotope with at least one decaying in a two-step process, a numerical solution is necessary.

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

$$\tau_{n+1} = \tau_n - \left.\frac{P'}{P''}\right|_{\tau_n} \tag{9}$$

for which we need

$$\frac{\mathrm{d}^2 y}{\mathrm{d}\tau^2} = a_y^2\, y(t)\, e^{-a_y \tau} - a_x \rho_{xy}\, x(t)\, \frac{a_x^2\, e^{-a_x\tau} - a_y^2\, e^{-a_y\tau}}{a_x - a_y}\,. \tag{10}$$

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

$$\frac{a_x^2\, e^{-a_x\tau} - a_y^2\, e^{-a_y\tau}}{a_x - a_y} = e^{\sigma\tau}\left(2\sigma \cosh \delta\tau + \delta \sinh \delta\tau + \sigma^2 \tau \frac{\sinh \delta\tau}{\delta\tau}\right)\,. \tag{11}$$

Using the same data from the ORIGEN-2 program as above, production constants k were calculated for cadmium, caesium, strontium, and tellurium. Activity constants a and branching ratios ρ were taken from JEFF 3.11 data [16]. 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. Assuming the four elements below 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. Equations (4) and (7) show that decay thermal power declines exponentially after the maximum.

Radiotoxicities were calculated using adult ingestion dose factors from ICRP publication 119 [10].

Figures per liter after storage assume fission products are not separated from daughters, and do not account for volume changes due to different densities of daughter products. Caesium 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.

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

		Volume	Maximum Power			Per liter after specified storage		
	Mass	Liters	Time τ	Watts	Per liter	Storage	Watts	Radiotoxicity
Cs	1374 kg	711.7	37.34 mins	334.0 kW	469.3 W	300 years	66.36 mW	25.67 kSv
Sr	371.7 kg	140.8	14.66 days	232.2 kW	1649 W	300 years	180.5 mW	138.6 kSv
Cd	63.95 kg	7.359	0 days	8.326 kW	1131 W	30 years	801.4 mW	401.9 kSv
Te	1.458 kg	0.2337	0 days	102.7 W	439.6 W	5 years	154.7 μW	5.510 Sv

11 Conclusions

Using finely divided fuel particles results in

- higher fuel density, both initially and after irradiation,

- more fission gas diffusing from particles to plenum, and more fission products diffusing to bond sodium, resulting in less fuel swelling,
- less cladding stress and essentially eliminated cladding strain, allowing greater burnup,
- more fission products diffusing to bond sodium, allowing simple separation from fuel,
- a sequence of simple mechanical and thermal processing steps from which fuel can be diverted directly back to service, at any stage, without costly chemical or pyroelectric processing,
- fewer fission products, with lower specific heat production and less radiotoxicity, sequestered in electrolyte, and then in mineral waste forms, and
- caesium and strontium, the two most radiotoxic fission products, with the highest long-term specific heat production, and the longest duration of custody, having been separated from other fission products as high-density relatively pure metals, resulting in lower storage cost for other fission products, and lower total storage costs.

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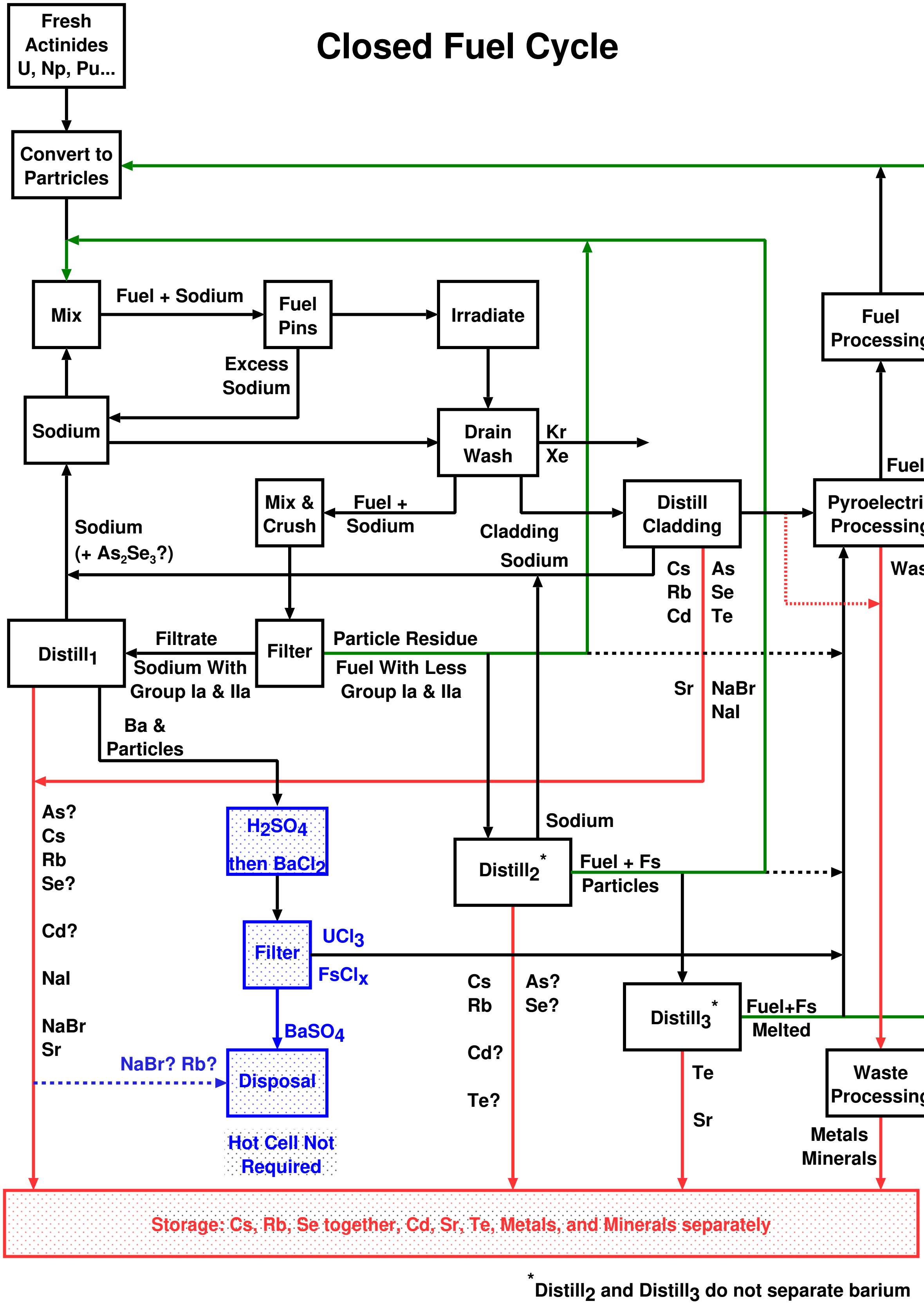
Closed Fuel Cycle
Fresh Actinides U, Np, Pu...
Convert to Partricles
Mix
Fuel + Sodium
Fuel Pins
Irradiate
Fuel Processing
Excess Sodium
Sodium
Drain Wash
Kr
Xe
Mix & Crush
Fuel + Sodium
Cladding
Distill Cladding
Pyroelectric Processing
Fuel
Waste
Sodium (+ As_2Se_3?)
Sodium
Cs Rb Cd
As Se Te
Distill$_1$
Filtrate
Sodium With Group Ia & IIa
Filter
Particle Residue
Fuel With Less Group Ia & IIa
Sr
NaBr NaI
Ba & Particles
As? Cs Rb Se?
Cd?
NaI
NaBr Sr
H_2SO_4 then $BaCl_2$
Filter
UCl_3
$FsCl_x$
$BaSO_4$
NaBr? Rb?
Disposal
Hot Cell Not Required
Sodium
Distill$_2$*
Fuel + Fs Particles
Cs Rb
As? Se?
Cd?
Te?
Distill$_3$*
Fuel+Fs Melted
Te
Sr
Waste Processing
Metals Minerals
Storage: Cs, Rb, Se together, Cd, Sr, Te, Metals, and Minerals separately
*Distill$_2$ and Distill$_3$ do not separate barium

Figure 2: