Process to Separate Strontium and Caesium
from Pyroelectric Refiner Electrolyte
Without Using Toxic Solvents

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Introduction

One tonne (1000 kg) of ten year old spent light-water reactor (LWR) fuel contains 52.18 kg of fission products.

Processing that fuel using a pyroelectric refiner leaves 11.8 kg (96.6 moles) of fission products dissolved in an electrolyte that is a mixture of lithium and potassium chlorides (LiCl and KCl).

Element	Amount	Radiotoxicity	Element	Amount	Radiotoxicity
Antimony	29.06 g	67.74 kSv	Lithium	25.74 μg	0
Arsenic	298.2 mg	0	Rubidium	524.5 g	1.758 mSv
Barium	2776 g	0	Samarium	1284 g	2.017 kSv
Beryllium	228.7 μg	180.9 μSv	Selenium	84.68 g	66.32 Sv
Bromine	31.96 g	0	Silver	115.4 g	30.36 Sv
Carbon	40.17 μg	3.844 mSv	Strontium	1135 g	86.97 MSv
Caesium	3680 g	67.54 MSv	Tellurium	749.1 g	13.07 kSv
Europium	233.4 g	624.1 kSv	Thulium	95.35 μg	217.3 μSv
Gallium	79.28 μg	0	Tin	139.9 g	213.8 Sv
Germanium	995.1 mg	0	Ytterbium	42.96 μg	0
Indium	2.892 g	19.88 nSv	Yttrium	676.2 g	0
Iodine	357.6 g	196.6 Sv	Zinc	9.716 mg	0

Assuming that substances that require longer durations of custody have greater costs for storage, it is desirable to separate caesium and strontium from other fission products because they, together, in ten-year-old spent fuel, constitute only 9.23% of the mass of all fission products (including those not dissolved in the electrolyte), but produce 99.4% of the total radiotoxicity of all fission products. The radioactive isotopes caesium-137 and strontium-90 have half lives of 30.04 and 28.79 years, respectively. They require custody for $300 - 400$ years. Graphs of fission-product decay appear in the appendix.

It is not necessary to produce pure caesium and strontium separations, provided the amounts of the other fission products mixed with them does not significantly increase the storage volume.

It is undesirable, however, to have significant amounts of caesium or strontium mixed with the other fission products. If they were mixed in significant quantity with other fission products, the entire amount of fission products would need long-term ($300 - 400$ year) custody.

Of the remaining fission products other than cadmium and europium, half are less radiotoxic than uranium in nature before ten years, and the other half are not radioactive.

Numerous processes have been described to separate caesium[1,2,3] and strontium[4] from other fission products. These are designed to produce essentially pure and complete separations. Most processes to separate them have an aqueous phase containing nitrate salts, and an immiscible organic solvent phase. The organic solvent is usually toxic and expensive. Additional toxic agents are used to adjust pH, enhance phase separation, and prevent formation of a third phase. Most of these processes were developed to work with nitrate salts, and would not work with chloride salts.

Their advantages are that they

- are continuous processes that can be operated using sequences of simple devices, such as centrifugal contactors, that alternately mix and separate the phases, arranged with aqueous and organic phases flowing in opposite directions,

- can have high separation ratios, simply by using more stages, and

- do not increase the total amount of material processed.

Two effective processes to separate strontium use monosodium titanate or monosodium pertitanate. These materials are very expensive ($200/kg), and it is not possible to separate strontium from them to reuse them. Another expensive process, developed by Eichrom, uses crown ethers bound to a resin substrate.

The entirely aqueous processes described here use common technical grade (not reagent grade) salts and acids, and zeolyte. They produce solid crystals of salts of silver, strontium, nonradioactive barium, mixed salts of antimony, europium, samarium, and tin, and chloride solutions of caesium and rubidium that can be captured in zeolyte, separating them from electrolyte. All other salts remain in solution and can be captured in zeolyte.

The process is intended to operate at room temperature, but at some stages, adjusting the temperature might increase the separation ratio.

The mixture of strontium isotopes in spent fuel generates 85.53 mW heat per gram of metal. If allowing the temperature to increase reduces separation effectiveness, it might be necessary to cool the apparatus at the stage of separating strontium precipitate from the solution of the other salts.

One disadvantage of this process is that it requires adding material.

Another disadvantage is that it is a batch process if centrifugation or dead-end filtration is used to separate precipitates from solutions. This is more difficult to automate than continuous solvent separation. Cross-flow filtration would allow a continuous process.

Subsequent discussion assumes processing one tonne of spent fuel, that has been stored for ten years. Scale specified amounts to the actual quantity of spent fuel processed, and the concentrations of fission products, depending upon the age of the spent fuel.

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[1]Bruce A. Moyer et al, **Next Generation Solvent Development for Caustic-Side Solvent Extraction of Cesium** ORNL/TM-2014/22, Oak Ridge National Laboratory (2014)

[2]J.-F.Dozol et al, **Extraction of Cesium by Calix[4]arene-crown-6: From Synthesis to Process** in **Calixarenes for Separations** (Gregg. Lumetta et al eds.) ACS Publications (2000)

[3]Harry Harmon et al, **Savannah River Site Salt Processing Project: FY 2002 Research and Development Program Plan**, Pacific Northwest National Laboratory (December 2001)

[4]Eight methods are reviewed in **Review and Assessment of Technologies for the Separation of Strontium from Alkaline and Acidic Media** by R. J. Orth and D. E. Kurath for Westinghouse Hanford Company.

1 Process

1.1 Dissolve loaded electrolyte in water

The first step is to dissolve electrolyte, loaded with fission products that do not remain in the anode basket or deposit at cathodes, or that escape as gases, by adding water.

Silver appears at the rate of 1.06 moles (115.4 grams). Silver chloride (AgCl) is much less soluble than the other fission product chlorides (192 μg per 100 grams H_2O at 20° C). Silver chloride will precipitate. Bromine (0.395 moles) and iodine (2.784 moles) have very low boiling points. They might evaporate during pyroelectric refining. If they remain, silver bromide (13.28 μg/100 grams H_2O) and silver iodide (0.3 μg/100 grams H_2O) will precipitate. There is insufficient silver (1.06 moles) to capture all the bromine and iodine.

Radioactive silver fission product isotopes other than ^{108m}Ag and ^{110m}Ag have very short half lives. Even though ^{108m}Ag has a half life of 418 years, only a very small amount (1.817 μg) is present. ^{110m}Ag (61.68 μg) has a half life of 249.8 days, and needs custody for seven years.

Separate AgCl precipitates from solution using filtration or centrifugation.

Tin appears at the rate of 1.152 moles (139.9 grams). Tin chlorides ($SnCl_2$ and $SnCl_4$) are very soluble in cold water (89 g / 100 g H_2O at 0° C). $SnCl_2$ tends to hydrolyze to Sn(OH)Cl in hot water or basic solution. Sn(OH)Cl is less soluble than other fission product chlorides. Prevent production of Sn(OH)Cl by controlling the temperature and (if necessary) reducing the pH of the initial solution using hydrochloric acid.

Tin (stannic) chloride ($SnCl_4$) is very soluble in cold water but it is a strong acid. It also has a tendency to hydrolyze. In hot water it decomposes to form hydrochloric acid.

Tin (1.152 moles) forms SnI_2 (0.99 g/100 grams H_2O), and will capture iodine that is not precipitated as AgI. Tin bromide is very soluble

If small amounts of bromine and iodine remain, they will be captured with the minor elements in zeolyte in the final stage (*viz infra*).

1.2 Carbonate separation

Among fission products and electrolyte, carbonates other than those of lithium, potassium, rubidium, and caesium are essentially insoluble, and lithium carbonate solubility is very small.

Carbonate solubilities, grams per 100 grams H_2O at 20° C

$SrCO_3$	1.1 mg
$BaCO_3$	2.40 mg
Li_2CO_3	1.33 g
K_2CO_3	111 g
Rb_2CO_3	98 g
Cs_2CO_3	260.5 g
others	ins

Add 103.55 moles (14.3 kg) of potassium carbonate (K_2CO_3) to the solution to precipitate carbonates of all remaining elements other than rubidium and caesium, and some additional potassium

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carbonate because lithium carbonate also has very low solubility; lithium from the electrolyte will compete with other cations in the formation of carbonate precipitates. Excess potassium carbonate does not cause a problem because caesium carbonate is more soluble than caesium chloride.

$$2\,LiCl + K_2CO_3 \rightarrow 2\,KCl\ (aq) + Li_2CO_3\ (ppt)$$
$$BaCl_2 + K_2CO_3 \rightarrow 2\,KCl\ (aq) + BaCO_3\ (ppt)$$

etc. for other insoluble carbonates.

Remove water using vacuum evaporation to increase concentrations significantly above saturation levels for the carbonates other than those of potassium, rubidium, and caesium. Carbonate precipitates of fission products other than rubidium and caesium will form. Some lithium chloride from the original electrolyte will also precipitate as lithium carbonate. If concentrations of caesium and rubidium salts are not near saturation (i.e., enough water was used to dissolve the loaded electrolyte, or not too much water was therafter removed), those salts will not precipitate. Small amounts of other elements' carbonates or chlorides might remain in solution, but that is not a problem. Remember that the goal is not to obtain pure caesium, but rather to obtain essentially caesium-free carbonate precipitates.
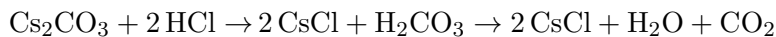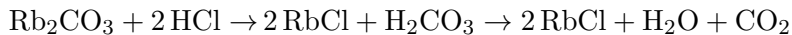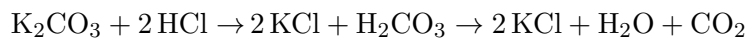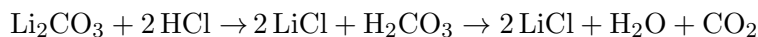
The supernatant will contain 27.3 moles of caesium as chloride (and some as carbonate), 6.079 moles of rubidium as chloride (and some as carbonate), lithium chloride from the original electrolyte to the extent lithium has not precipitated as carbonate, and significant potassium chloride, both from the original electrolyte and resulting from the formation of carbonates.

Separate carbonate precipitates from solution. Wash the precipitates to remove caesium and rubidium chlorides and carbonates.

1.3 Carbonate separation supernatant

The supernatant resulting from carbonate separation will consist of lithium chloride (LiCl) potassium chloride (KCl), rubidium chloride (RbCl), caesium chloride (CsCl), carbonates of potassium (K_2CO_3), rubidium (Rb_2CO_3), and caesium (Cs_2CO_3) to the extent that excess potassium carbonate was added, very small amounts of other chlorides that were not converted to carbonates, and very small amounts of other carbonates in solution.

Convert carbonates in solution to chlorides by adding hydrochloric acid:

$$Li_2CO_3 + 2\,HCl \rightarrow 2\,LiCl + H_2CO_3 \rightarrow 2\,LiCl + H_2O + CO_2$$
$$K_2CO_3 + 2\,HCl \rightarrow 2\,KCl + H_2CO_3 \rightarrow 2\,KCl + H_2O + CO_2$$
$$Rb_2CO_3 + 2\,HCl \rightarrow 2\,RbCl + H_2CO_3 \rightarrow 2\,RbCl + H_2O + CO_2$$
$$Cs_2CO_3 + 2\,HCl \rightarrow 2\,CsCl + H_2CO_3 \rightarrow 2\,CsCl + H_2O + CO_2$$

Numerous aqueous processes have been developed to separate these chlorides, The simplest one uses potassium aluminum sulfate $(KAl(SO_4)_2)$, commonly called alum. Lithium apparently does not form an alum.

Alum solubilities, grams per 100 grams H_2O

Molecule	0° C	10° C	20° C	50° C	80° C	100° C
$KAl(SO_4)_2$	3.90 g	9.52 g	16.1 g	44.11 g	134.47 g	357.48 g
$RbAl(SO_4)_2$	0.71 g	1.09 g	1.63 g	4.98 g	21.60 g	
$CsAl(SO_4)_2$	0.19 g	0.29 g	0.42 g	1.235 g	5.29 g	

Rather than separate them using an aqueous process, caesium and rubidium chlorides can be separated from lithium and potassium chlorides using zeolyte. Lithium and potassium chlorides, which are not captured in the zeolyte, should be sufficiently pure that they can be re-used as electrolyte.
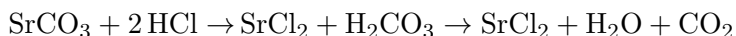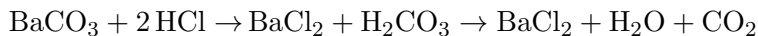
^{87}Rb has a very long half life (48.10 Gy), and is therefore only slightly radioactive. Rubidium in nature contains about 28% ^{87}Rb, while rubidium fission products are 69% ^{87}Rb. The other rubidium isotope (^{85}Rb) is not radioactive; rubidium does not need special custody. It would be desirable (but difficult) to separate rubidium from caesium; if they remain together, the amount of material requiring long-term (300 – 400 year) custody is increased by about 12%.

By separating these four alkali metals from the other fission products, and then separating lithium and potassium from rubidium and caesium, the amount of metal requiring further processing is reduced by 4.2 kg, or 31%, to about 7.6 kg.

1.4   Carbonate precipitates

1.4.1   Dissolve carbonate precipitates

Add 207.1 moles (7.55 kg) hydrochloric acid to the carbonate precipitates to convert them to chlorides in solution. The following reactions will result:

$$BaCO_3 + 2\,HCl \rightarrow BaCl_2 + H_2CO_3 \rightarrow BaCl_2 + H_2O + CO_2$$
$$SrCO_3 + 2\,HCl \rightarrow SrCl_2 + H_2CO_3 \rightarrow SrCl_2 + H_2O + CO_2$$

and similarly for other carbonate precipitates.

1.4.2   Barium nitrate separation

The only radioactive barium isotope (^{137m}Ba) is the decay product of ^{137}Cs, and has a very short half life (2.552 minutes). After separating caesium, it no longer accumulates among the elements precipitated as carbonates. Barium does not need custody.

The amount of barium in fission products (2.776 kg, 20.23 moles) is more than twice the amount of strontium (1.135 kg); removing a significant fraction of barium reduces the amount of remaining metal requiring processing by 36%, to about 4.8 kg.

Nitrate Solubilities, grams per 100 grams H_2O at 20° C

Barium	9.02
Europium	140.9
Samarium	137.5
Strontium	70.8

Add at least 40.46 moles (2.79 kg) of lithium nitrate to the solution that results from dissolving carbonates using hydrochloric acid. The following reaction will result:

$$2Li(NO_3) + BaCl_2 \rightarrow 2LiCl\ (aq) + Ba(NO_3)_2\ (\ ppt)\,.$$

Remove water using vacuum evaporation to increase concentration significantly above saturation for barium nitrate, but not above saturation for other nitrates; barium nitrate will precipitate.

Separate barium nitrate precipitate from solution. Wash precipitate with small amounts of concentrated lithium nitrate solution to dissolve other nitrates and retain barium nitrate as a precipitate.

1.4.3   Europium and samarium separation

Europium isotopes constitute 0.44% of the mass of fission products and produce 0.4% of the total radiotoxicity of all fission products. The amount of samarium is 5.5 times the amount of europium. It is far less radiotoxic, but difficult to separate from europium. It is desirable to remove europium and samarium so as not to combine them with strontium.

Significantly radioactive Europium isotopes per tonne of spent fuel

Isotope	Amount	Half life	Radiotoxicity
^{150}Eu	375.0 ng	36.36 y	1.195 mSv
^{152}Eu	37.85 mg	13.52 y	339.2 Sv
^{154}Eu	29.45 gm	8.593 y	588.6 kSv
^{155}Eu	6.383 gm	4.753 y	35.17 kSv

These are all small amounts, especially ^{150}Eu, the longest-lived one. They can probably be safely stored with fission products that need only short-term ($\sim$ 30 year) custody, but if separated it would be desirable to store them for $\sim$ 50 years.

Europium ferrocyanide and potassium samarium ferrocyanide are known to be insoluble. Presumably, potassium europium ferrocyanide and samarium ferrocyanide are also insoluble. In addition, antimony and tin ferrocyanides are insoluble. Barium ferrocyanide is essentially insoluble (0.009732 grams per 100 grams H_2O). Strontium ferrocyanide is soluble (50 grams per 100 grams H_2O). Lithium ferrocyanide is very soluble.

Add 5.75 moles (1.38 kg) of lithium ferrocyanide to the supernatant that results from barium nitrate separation. Ferrocyanides of antimony, europium, samarium, and tin, and remaining barium, will precipitate.

Antimony (26.06 g) and tin (139.3 g) do not require long-term custody, but precipitating them with europium (233.4 g) and samarium (1.284 kg) increases the amount needing long-term custody ($\sim$ 100 years) by only 11%.

Significantly radioactive Tin isotopes per tonne of spent fuel

Isotope	Amount	Half life	Radiotoxicity
^{119m}Sn	2.193 μg	293.0 d	123.7 mSv
^{121m}Sn	4.782 mg	55.00 y	3.979 Sv
^{123}Sn	1.506 ng	129.2 d	962.2 μSv
^{126}Sn	23.49 g	230.0 ky	209.8 Sv

The very long half life of ^{126}Sn means it has very low radioactivity. ^{121m}Sn, is present in especially small amounts. It decays to ^{121}Sn, which decays with a 1.12 day half life to stable ^{121}Sb.

Alkali metal ferrocyanides produce bulky precipitates. Calcium ferrocyanide produces a more condensed crystalline precipitate. Calcium sulfate has very low solubility. Calcium chloride that results from ferrocyanide extraction of europium, samarium, antimony, and tin will precipitate with strontium sulfate (*viz infra*). Calcium can be separated from strontium as permanganate.

Separate precipitates from solution. This reduces the amount of material requiring further processing by 1.683 kg, to about 2.6 kg.

1.4.4 Strontium separation

Strontium appears at the rate of 12.76 moles (1.135 kg). Add at least 12.76 moles (1.4 kg) of lithium sulfate, to precipitate strontium sulfate:

$$\text{Li}_2\text{SO}_4 + \text{SrCl}_2 \rightarrow 2\,\text{LiCl (aq)} + \text{SrSO}_4\text{ (ppt)}.$$

Strontium sulfate solubility is very low (13.2 mg/100 g H_2O at 20° C). Remaining barium will also precipitate as barium sulfate because of its low solubility (0.285 mg/100 g H_2O at 20° C).

Sulfate solubilities, grams per 100 g H_2O at 20° C

Element	Mass	Moles	Solubility	Radiotoxicity	$T_{1/2}$
As	0.2982 g	3.98 mM	s	0	
Ba	2.776 kg	20.23 M	0.000285	0	
Be	22.9 mg	23.2 μM	41.4	181 μSv	1.600 My
Eu	233.4 g	1.525	2.56	624 kSv	8.593 y
Ga	1.73 μg	114 nM	m.s.	0	
In	2.892 g	25.2 mM	53.92	19.9 nSv	441.0 Ty
Sb	29.06 g	0.2379 M	m.s.	67.74 kSv	2.759 y
Sm	1.284 kg	8.6 M	2.7	2.02 kSv	90.00 y
Sn	139.9 g	1.152 M	33	213.8 Sv	230.0 ky
Sr	1.135 kg	12.76 M	0.0132	87.0 MSv	28.79 y
Te	749.1 g	5.797 M	m.s.	13.1 kSv	57.40 d
Tm	95.4 μg	564 nM	m.s.	217 μSv	1.917 y
Yb	43.0 μg	243 nM	38.4	0	
Y	676.2	7.605	34.8	8.39 MSv	2.671 d
Zn	9.72 mg	1.40 μM	7.3	0	

Although strontium sulfate has the lowest solubility of remaining fission products, other fission product sulfates also have low solubility, and some might precipitate with strontium. In particular, europium and samarium sulfates have low solubility, and therefore barium, europium, and samarium should be removed before separating strontium.

The reason to separate barium, europium, and samarium before strontium is to minimize the storage requirement for strontium. Perfect separation of barium, europium, and samarium from strontium is not necessary. The amount of barium fission products (2.776 kg) is more than twice the amount of strontium (1.135 kg). The amounts of europium (233.4 g) and samarium (1.135 kg) are together greater than the amount of strontium. If strontium, barium, europium, and samarium are directly precipitated together as sulfate, without first precipitating barium nitrate, europium ferrocyanide, and samarium ferrocyanide, the amount of material needing long-term (300 – 400 year) custody

(5.378 kg) is 4.74 times the amount of strontium alone (1.135 kg).

1.4.5 Remaining solution

After caesium, rubidium, barium, antimony, europium, samarium, silver, strontium, and tin have been removed, the only fission products remaining in significant quantity are selenium (84.68 g), tellurium (749.1 g) and yttrium (676.2 g). The isotope of tellurium that produces significant radiotoxicity has a half life of 57.40 days. Within two years tellurium is innocuous. ^{90}Y is the decay product of ^{90}Sr, and has a half life of 2.671 days. Within 90 days of separating strontium, yttrium is essentially nonradioactive; in the first table above, its radiotoxicity before separation is included in the radiotoxicity of strontium. The radioactive isotope of selenium has a half life of 377,000 years, therefore has very low activity, and only 8.869 grams appear. The boiling points of selenium chloride (127° C) and tellurium chloride (380° C) are significantly lower than yttrium chloride (1507° C); they could (if necessary) be separated from yttrium chloride by distillation. They might evaporate during pyroelectric processing.

If the remaining solution is significantly radioactive after two years, it might be contaminated with caesium or strontium. Repeat the carbonate precipitation step, convert the resulting solution to chlorides, and process it using zeolyte. If the resulting carbonate precipitate is significantly radioactive it is probably still contaminated with strontium. Convert it to chloride using hydrochloric acid, and precipitate strontium using lithium sulfate.

Repeat if necessary, then process the remaining solution using zeolyte. The solution purified using zeolyte might contain significant lithium sulfate or lithium ferrocyanide, unless those salts are captured in zeolyte, so it cannot be directly re-used as electrolyte without further processing.

2 Summary of separations

	Grams per tonne	Fraction	Custody	Elements	Process	Final Form
	389.6	3.3%	none	Br, I	Evaporate	Gas
1	115.4	0.98%	∼ 7 years	Ag	H_2O	Precipitate
2	4204	35.6%	300–400 years	Cs, Rb	CO_3	Zeolyte
3	2776	23.5%	none	Ba	NO_3	Precipitate
4	1686	14.3%	∼ 60 years	Eu, Sb, Sm, Sn	$Fe(CN)_6$	Precipitate
5	1135	9.6%	300–400 years	Sr	SO_4	Precipitate
6	1510	12.8%	< 2 years	Se, Te, Y	Remaining	Zeolyte
6	4.195	0.035%	none	As, Be, C, Ga, Ge, In, Tm, Yb, Zn	Remaining	Zeolyte
	11821			Total		

The amounts and radiotoxocities of Arsenic, beryllium, carbon, gallium, germanium, indium, thulium, ytterbium, and zinc are extremely small. They do not need special custody. They are not separated from selenium, tellurium, and yttrium.

Caesium (with or without rubidium), europium, antimony, samarium, strontium, and tin should be converted to impervious insoluble ceramics for long-term storage. Silver, selenium, tellurium,

yttrium, and elements remaining in solution can be stored in simple containers and then disposed. Barium, bromine, and iodine can be disposed immediately.

One might observe that 45% of materials require 300 –400 years' custody, and 14.3% require 100 years' custody. Separating all fission products from electrolyte using zeolyte would only double the storage requirement. Therefore, there is no reason to separate the fission products into different custody-duration categories.

3 Reagent costs

The cost of lithium ferrocyanide could not be found. $10/kg assumed.

Reagent	Moles	Molecular Weight	Total Amount	Price	Cost
K_2CO_3	103.55	138.205	14.31 kg	$1.05/kg	$15.03
HCl 11.65 M	207.1	36.46	17.78 L	$0.94/L	$16.67
$Li_2Fe(CN)_6$	5.76	239.8	1.38 kg	$10.00/kg	$13.80
$LiNO_3$	40.46	68.946	7.79 kg	$1.00/kg	$2.79
Li_2SO_4	12.76	109.94	1.40 kg	$10.00/kg	$14.03
Total	369.63				$61.22

4 Potassium and lithium nitrate, sulfate, and ferrocyanide

Potassium or lithium nitrate, lithium sulfate, and lithium ferrocyanide, are used to remove barium, europium, samarium, strontium, and tin. If they remain in solution after those steps, they cannot be directly re-used as electrolyte.

They can be converted to chloride using barium chloride:

$$2\,KNO_3 + BaCl_2 \rightarrow 2\,KCl\,(aq) + Ba(NO_3)_2\,(ppt)$$
$$2\,LiNO_3 + BaCl_2 \rightarrow 2\,LiCl\,(aq) + Ba(NO_3)_2\,(ppt)$$
$$Li_2SO_4 + BaCl_2 \rightarrow 2\,LiCl\,(aq) + BaSO_4\,(ppt)$$
$$Li_4Fe(CN)_6 + 2BaCl_2 \rightarrow 4LiCl\,(aq) + Ba_2Fe(CN)_6\,(ppt)$$

Barium nitrate and barium chloride remaining in solution can be removed using zeolyte.

5 Anode basket contents

5.1 Cadmium

Cadmium constitutes 0.36% of the mass of fission products and produces 0.044% of the total radiotoxicity of all fission products in ten-year-old spent fuel. ^{113m}Cd has a half life of 14.10 years. Cadmium remains in the anode basket of the electrorefiner; it does not appear in the electrolyte. Its boiling point is several thousand degrees Fahrenheit below the boiling point of other metals in the anode basket. It can therefore be separated from them by distillation.

5.2   Ruthenium

Ruthenium constitutes 6.6% of the mass of fission products and produces 0.13% of the total radiotoxicity of all fission products in ten-year-old spent fuel. ^{106}Ru decays to ^{106}Rh with a half life of 1.020 years. ^{106}Rh decays to stable ^{106}Pd with a half life of 30 seconds. The market value of ruthenium is \$6.50/gram, or \$22,522.50 per tonne of spent fuel. Ruthenium requires custody for less than ten years.

5.3   Other elements

None of the other elements in the anode basket require custody.

- Molybdenum is 9.7% of fission products, and is not radioactive. The market value for molybdenum is 2.7¢/gram, or \$136.59 per tonne of spent fuel.

- ^{107}Pd has a half life of 6.5 My; it is essentially nonradioactive. The market value for palladium is \$47.72/gram, or \$112,576 per tonne of spent fuel.

- ^{102}Rh has a half life of 2.902 years, but only 141.1 μg are present in ten-year-old fuel, and it is essentially gone after another ten years. The market value of rhodium is \$141.54/gram, or \$86,535 per tonne of spent fuel.

- ^{99}Tc has a half life of 214.0 ky, so its radioactivity is very low. The market value of technetium is \$60.00/gram, or \$68,160 per tonne of spent fuel.

- ^{93m}Nb has a half life of 16.13 years, but only 4.153 mg are present (7.06 mg total Nb), and its market price is about \$0.165/gram.

- ^{93}Zr has a half life of 1.530 My, so its radioactivity is very low. The market value of zirconium is \$0.926/gram, or \$5,045.77 per tonne of spent fuel. Zirconium is also an activation product (fuel pin cladding) of which 237.9 kg accompany each tonne of spent fuel. At \$0.96/gram, its market value is \$220,295.40.

The total value of nonradioactive metals in the anode basket, per tonne of spent fuel, is \$487,703.

6   Rare-earth cathode

Elements in the rare-earth cathode constitute 25.9% of all fission products and produce 0.086% of radiotoxicity.

- ^{144}Ce has a half life of 285 days, but only 55.2 mg are present. ^{141}Ce has a half life of 32.5 days; less than a picogram is present. The remaining 3607 grams of cerium are not radioactive. Cerium needs custody for about seven years.

- ^{147}Pm has a half life of 2.623 years, but only 11.02 grams are present. Promethium does not need custody.

- Neodymium is 11.8% of fission products, and is not radioactive.

- Lanthanum is 3.5% of fission products. ^{138}La has a half life of 102 billion years, or about 7.4 times the age of the universe, and only 7.637 mg are present. The remaining 1849 grams are not radioactive. Lanthanum does not need custody.

- Praseodymium is 3.2% of fission products and produces 2.1 ppm of radiotoxicity. ^{144}Pr and ^{144m}Pr have half lives of 17.28 and 6.9 minutes, respectively; they are decay products of ^{144}Cm. They constitute 2.331 μg and 11.65 ng, respectively, out of a total of 1689 grams. Praseodymium does not need custody.

Elements from the rare-earth cathode have essentially identical chemical properties; they should not be processed in the same stream as the electrolyte. They might precipitate as ferrocyanides with europium, increasing the amount of material requiring custody for $\sim$100 years by about nine times. Otherwise, they would precipitate with strontium, increasing the amount of material requiring custody for 300 – 400 years by nearly 13 times..

Appendix: Fission products decay curves

The graphs below show the relative radiotoxicities of fission products that have half lives greater than ten years. The units are Sieverts per gram of total fission products, not Sieverts per gram of each fission product individually.

Individual isotopes, top to bottom at 24 years:

Sr-90	Cs-137	Eu-154	Cd-113m	Eu-155	Sm-151	Tc-99	I-129
Eu-152	Zr-93	Sn-126	Cs-135	Se-79	Pm-146		

Total Radiotoxicity, Sv/g

Appendix: Flowsheet

Electrolyte XCl → Mixer ← H_2O

Mixer → Cross-flow Filter → 192 µg/100 g → **AgCl AgI AgBr** SnI_2 **precipitate**

Cross-flow Filter → XCl in H_2O → Mixer ← K_2CO_3

Mixer → Cross-flow Filter → **CsCl** KCl RbCl solution → Zeolyte

Zeolyte → **CsCl** RbCl Sodalite

Zeolyte → KCl Solution

Cross-flow Filter → Li_2CO_3, X_yCO_3 precipitate → Mixer ← HCl

Mixer → $H_2CO_3 \rightarrow H_2O + CO_2$

$H_2CO_3 \rightarrow H_2O + CO_2$ → All except AgCl CsCl KCl RbCl solution → Mixer ← $LiNO_3$

Mixer → Cross-flow Filter → 9.02 g/100 g → **Ba(NO$_3$)$_2$** precipitate

Cross-flow Filter → All except AgCl $BaCl_2$ CsCl KCl RbCl solution → Mixer ← $Li_4Fe(CN)_6$

Mixer → Cross-flow Filter → **Eu$_2$Fe(CN)$_6$** **Sb$_4$(Fe(CN)$_6$)$_3$** **Sm$_2$Fe(CN)$_6$** **Sn$_2$Fe(CN)$_6$** precipitate

Cross-flow Filter → Se_2Cl_2 $SrCl_2$ $TeCl_4$ YCl_3 Minor elements → Mixer ← Li_2SO_4

Mixer → Cross-flow Filter → 0.0132 g/100 g → **SrSO$_4$** precipitate

Cross-flow Filter → Se_2Cl_2 $TeCl_4$ YCl_3 Minor elements → Zeolyte

Zeolyte → **Se$_2$Cl$_2$ TeCl$_4$ YCl$_3$** **Minor elements** Sodalite

Zeolyte → LiCl solution