PARTITIONING AND DURABLE WASTE FORMS FOR HIGHLY RADIOTOXIC ISOTOPES

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Partitioning light-water reactor spent nuclear fuel offers the opportunity to dispose of the more troublesome radioisotopes in durable waste forms for the purpose of providing long-term isolation. These more troublesome radioisotopes can be identified by their radiotoxicity to determine which should be disposed of in a repository with long-term performance assurance. The purpose of this paper is to investigate the partitioning of isotopes into two groupings: one requiring long-term repository performance assurance with respect to disposal, and the other that does not. Once grouped in this way applicable separation technologies and durable waste forms are identified, and a material-balance analysis is used to estimate the disposal requirements of these groupings in terms of masses. Disposal masses are estimated for the cases of direct disposal, partitioning, and limited recycle where recovered plutonium and uranium are recycled once. Recovered uranium with adequate decontamination is expected to be disposed of or stored as the oxide. This investigation is not intended to imply that spent nuclear fuel should be partitioned solely for the purpose reducing the mass disposed in a single repository, but to illustrate the relative disposal masses for the identified components.

I. INTRODUCTION

Some long-lived troublesome radioisotopes could be partitioned from light-water reactor (LWR) spent nuclear fuel so that they can be disposed of in a repository with long-term performance assurance. These radioisotopes requiring partitioning from spent nuclear fuel can be put into durable waste forms that provide for robust isolation. By isolating these radioisotopes, the time frame of repository performance the remaining assurance for radioisotopes is reduced. Once those radioisotopes that require long-term disposal are identified, appropriate separations technologies to accomplish the partitioning can be identified along with further research and development needed to bring the separations technologies to economic, full-scale operation. A suitable waste form for the partitioned isotopes and all elements in the various separated waste streams can then be identified. However, a geologic repository will still be required for all elements, except possibly for recovered uranium. An important consideration then is the relative masses of these partitioned elements in final waste forms and the size in terms of disposal masses of the resulting repositories. It is the purpose of this paper to consider available information to accomplish partitioning and disposal, and to compare disposal masses for direct disposal of spent nuclear fuel, partitioning of the spent nuclear fuel mass and subsequent disposal, and limited recycle of plutonium and uranium in light water reactors where a reduction in the disposal mass of plutonium can be realized.

II. PARTITION BASIS

Considerable discussion on partitioning and transmutation to improve the utilization of a geologic repository and protect human health and safety can be found in the peer-reviewed literature. 1,2 measures determine which radioisotopes should be partitioned, or separated, and these measures are 1) curies, and 2) radiotoxicity. Consideration of curies is described in the Yucca Mountain Total System Performance Assessment for License Application.³ Based on this criterion, cesium, strontium, americium, plutonium would be partitioned because they are the major contributors to the total curie inventory of spent nuclear fuel out to 10,000 years. After this time period, the curie inventory has decayed to approximately one percent of the starting inventory.

Conversely, radiotoxicity of an isotope is the inventory weighted by an appropriate human dose conversion factor.⁴ For a mixture of isotopes, such as spent nuclear fuel, radiotoxicity is summed over all

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Despite having units of dose, radioisotopes. radiotoxicity does not provide a measure of potential dose from any nuclear waste material because it does not account for the quantity of the inventory causing humans or escaping into the exposure to Calculation of dose requires environment. knowledge of the interplay between waste management techniques and geologic-site characteristics. Hedin⁵ provides a discussion of radiotoxicity and accessibility (mobility) to illustrate the concept of risk in the context of the long-term safety of a deep repository by taking into account waste management techniques and geologic-site characteristics. Grambow⁶ describes the transport of mobile fission products and activation products in clay and the impact of the disposal environment on repository safety.

Westlén⁷ bases the radiotoxicity discussion on half lives of important actinides and long-lived fission products along their respective dose conversion coefficients. Westlén derives a plot of the time evolution of the radiotoxicity from spent nuclear fuel and also the radiotoxicity in this same fuel for individual actinides along with the total due to the actinides. The main observation from Westlén's results shows that the radiotoxicity of spent fuel approaches that of natural uranium at about 10⁷ years and that the transuranics are the reason for this very long time frame.

The shapes of the total radiotoxicity curves for spent nuclear fuel as a function of time described by Piet⁴ and Westlén⁷ are approximately the same; both decrease with respect to time in about the same manner and show a slight change in slope at around 100,000 years. The shapes of the fission-product curves for each are also approximately the same, rapidly decreasing until about 500 years followed by little change with the final estimated radiotoxicity being below that of natural uranium. The shapes of the transuranic (TRU) curves are also approximately the same; the TRUs account for the major portion of the radiotoxicity out to about 100,000 years. The contribution of individual TRUs, or actinides, to the total actinide radiotoxicity as derived by Westlén show that plutonium and americium are the dominant contributors. Neptunium is never a major contributor to the total radiotoxicity and its contribution is always less than the radiotoxicity of natural uranium. Other references recognize radiotoxicity as the preferred criterion for determining which radioisotopes should be isolated; some of these are Nishirhara et al., 8 Gombert, 9 and Greneche et al. 1

The Yucca Mountain Repository as presented in the Total System Performance Assessment for the License Application³ for an oxidizing geologic environment provides an example of dose derived from geologic specific characteristics. The results of this work show that $^{239}\mathrm{Pu},~^{237}\mathrm{Np},~^{129}\mathrm{I},$ and $^{226}\mathrm{Ra}$ generally dominate the mean annual dose for the postclosure period from 100,000 to one million years. Greneche et al., derive evaluations of the radiological dose impact for specific fuel-cycle cases of the disposal of high-level radioactive waste in repositories sited in granite, clay, and salt, all of which are reducing environments. In this same discussion it is shown that the contribution to spent fuel toxicity due to fission products is quite small after about 500 years. The predicted doses derived by Greneche et al.. fall far below the dose constraint of 0.3 mSv/a (30 mrem/year) recommended by the International Commission on Radiological Protection. 10 Also concluded is that the impact of partitioning for all cases is that the maximum dose is rather limited because it is essentially due to mobile long-lived fission and activation products. mobile long-lived fission products are 129I, 79Se, ¹³⁵Cs, ⁹⁹Tc, and ¹²⁶Sn. Actinides contribute very little to the dose due to the reducing conditions of the selected geologies.

III. IDENTIFICATION OF CANDIDATE RADIOISOTOPES FOR PARTITIONING

Considering radiotoxicity, the following radioisotopes are candidates for partitioning to reduce the long-term radiotoxicity of spent nuclear fuel independent of the repository geologic medium: ⁷⁹Se, ⁹³Zr, ⁹⁹Tc, ¹⁰⁷Pd, ¹²⁶Sn, ¹²⁹I, and ¹³⁵Cs (Ref. 7). The fission products identified by Piet⁴ are (also) ⁹³Zr and ¹²⁶Sn. Based on radiotoxicity, the fission-product elements identified as candidates for partitioning are selenium, zirconium, technetium, palladium, tin, iodine and cesium. The transuranics identified by Westlén that are the major contributors to spent-fuel radiotoxicity are plutonium, americium and curium; neptunium is not a major contributor. transuranics identified by Piet⁴ are also the major contributors to spent fuel radiotoxicity. Based on radiotoxicity, all of the transuranic elements are candidates for partitioning.

Using the criterion of repository-specific geology with respect to the long-term dose contributors rather than radiotoxicity, different isotopes are identified for partitioning. For a repository sited in oxidizing tuff, the major dose contributors identified by McNeish et al., are 239 Pu, 237 Np, 129 I, and 226 Ra. Because 226 Ra is identified, 238 U must also be included. 226 Ra will be

partitioned with uranium. Based on dose the transuranic elements identified for partitioning are plutonium, neptunium, uranium, along with the fission-product element iodine. For repositories sited in reducing geologic environments such as granite, clay, or salt, the major contributors to dose are determined by Greneche et al., to be the long-lived fission products which are 129 I, 79 Se, 135 Cs, 99 Tc, and 126 Sn. These are also identified by Westlén as noted above, along with 93 Zr and 126 Sn by Piet. No transuranics are identified because of their very low solubility and high sorption capacity in reducing systems. Based on dose the fission-product candidate elements identified for partitioning are iodine, selenium, cesium, technetium and tin.

Considering radiotoxicity or dose, it appears that iodine, as ¹²⁹I and its half-life of 16 million years, is the most important fission-product element to partition because of its accumulation in the thyroid and relatively high dose-conversion coefficient. The other fission-product radioisotopes are not uniquely important with respect to biological uptake, and these radioisotopes are in the elements selenium, cesium, technetium, tin, palladium and zirconium. Since the radioisotopes in these elements contributed relatively little to dose and have relatively low radiotoxicities compared to natural uranium, it may not be worthwhile to partition these and put them into a durable waste form.

In summary, it appears that partitioning could be a benefit for the transuranics and iodine based on radiotoxicity and/or geologic repository performance. The next consideration is available separation processes to effectively partition these elements.

IV. SEPARATION TECHNOLOGIES

Reprocessing relies on separation technologies to allow the partitioning of energetic elements for recycle and a significant reduction in high-level waste volume and radiotoxicity prior to final disposal. The only process that has operated at an industrial scale is PUREX (Plutonium - Uranium Extraction), which includes several variations of flow sheets, as evidenced by examples from Schulz and Benedict. 11 For example, the PUREX process at Hanford operated differently than PUREX at Savannah River. Advances to PUREX have been proposed and demonstrated at the pilot scale. Called UREX (for Uranium Extraction) or advanced PUREX, these processes are considered industrially feasible. As with PUREX, UREX has many process possible variations. 12

In some variations, UREX consists of many steps that partition spent-fuel elements into nine product groupings.1 Uranium and technetium are recovered in one process stream and then separated by pertechnetate ion exchange. The raffinate is treated by fission-product extraction (FPEX) to remove the alkali and alkaline earth components. The FPEX raffinate is separated into transition metal fission products and lanthanides and actinides by the transuranics extraction process. The actinides (TRUs) are separated from the lanthanides using trivalent actinide separations by phosphorus-reagent extraction from aqueous complexes. Radioactive gases from the front end can be treated to recover iodine by capture on silver mordenite.¹³

For PUREX, Advanced PUREX, and UREX, and their variations, no specific separation of all fission-product elements exists except for the volatiles that occur as off-gas components, and technetium. At present, further partitioning of fission products into element groupings of interest as identified above, i.e., selenium and tin, is not being pursued. Note that a variety of processes have been investigated for the recovery of cesium from acidic wastes. ^{14,15}

V. WASTE FORMS

Considerable effort has been expended on the development of waste forms. 16 In some cases, the waste form is developed without a consideration of the disposal environment. However, regardless of the disposal environment, the waste forms currently produced at an industrial scale are primarily borosilicate glass and iron phosphate. It is to be recognized that borosilicate glass is not a singular waste formulation, there are many glasses whose main components are B_2O_3 and SiO_2 (Ref. 17). Borosilicate glass is not universally accepted as being the best or most appropriate waste form. Indeed Laverov et al., 18 state that "Glasses are insufficiently stable for isolation of long-lived radionuclides, including actinides." Considering the disposal environment, borosilicate glass experiencing a high (aqueous) flow rate can be substantially altered in relatively short periods of time. 16

A waste form that has received considerable attention, following borosilicate glasses, are the SYNROCs, of which there many formulations;¹⁷ for example, SYNROC-C was developed for immobilization of commercial waste from the reprocessing of spent power reactor fuel, SYNROC-D for defense waste, SYNROC-E with improved long-term stability, and SYNROC-F for unprocessed

spent fuel containing significant amounts of uranium and plutonium. While SYNROCs have been extensively studied, there is no commercial application of the immobilization of high-level radioactive waste using this waste form.

There are primarily two attributes of a waste form for consideration for long-term disposal; these are chemical durability and radiation resistance. Chemical durability pertains essentially to wasteform aqueous dissolution under conditions relevant to disposal to minimize release of radiotoxic species.¹⁹ Also, Ewing²⁰ defines durability as referring to a wide variety of properties: mechanical strength, thermodynamic stability, slow kinetics for corrosion processes, (and) retention of trace elements due to low diffusivity. Considering radiation resistance, or resistance to radiation damage, of a waste form, many alteration processes can occur as described by Ewing et al.²¹ This particular property is sometimes termed "radiation stability" as described by Donald et al. 17 Radiation damage can induce important changes in physical and chemical properties, most notably the from the crystalline-to-aperiodic transition (amorphous or metamict) state can occur. 21 Donald et al., 17 conclude that, in practice, radiation effects in glass do not seem to be particularly detrimental to the longer-term integrity or chemical durabilities of glasses, at least over the time scales so far examined. However, this conclusion is not universally accepted because Ewing et al.,²¹ state that the post-disposal radiation damage to waste-form glasses and crystalline ceramics can be significant.

VI. SELECTION OF RADIOELEMENTS FOR PARTITIONING AND WASTE FORMS FOR LONG-TERM DISPOSAL

The radioelements selected for illustrating partitioning and disposal in durable waste forms for long-term repository performance assurance are iodine, plutonium, and the minor actinides, as they account for most of the radiotoxicity. Radioelements that are to be disposed of otherwise in a manner not requiring long-term performance assurance are all the remaining fission products and uranium. The reason for this selection is that the radiotoxicity of fission products decreases to less than that of natural uranium in centuries. Uranium, appropriately decontaminated, can (possibly) be disposed of (or stored) in a manner similar to that being pursued for the converted uranium hexafluoride tails.²²

The selection of the following waste forms for the indicated elements is based on available engineering physical-property parameters necessary to calculate a final waste-form mass. There are compilations of waste-form information such as that by Trocellier, ²³ but compositions of the waste form with respect to radioelement composition and waste-form densities are not always available. Therefore for the purpose of this illustrative investigation, waste forms are selected for which the engineering parameters of composition and density are available as referenced or can be reasonably estimated. This observation on the lack of pertinent information points out the need for a detailed review and compilation of information on this topic.

The waste form selected for iodine is lead Naturally occurring apatite, generally apatite. Ca₁₀(PO₄)₆(F, OH, Cl)₂, has high chemical, radiation thermal durability. Iodine-apatite Pb₁₀(VO₄)_{4.8}(PO₄)_{1.2}I₂ is synthesised through solidstate reaction of PbI₂ and Pb₃(VO₄)_{1.6}(PO₄)_{0.4}. This iodine-rich core is then coated with a layer of Pb₃(VO₂)_{1.6}(PO₄)_{0.4} during a sintering stage at a pressure of 25 MPa and a temperature of 700°C, to form a composite ceramic.²⁴ The pertinent physical property for estimating the iodine waste form mass is an iodine loading of 1.3 wt% based on stoichiometry and the composite-core geometry.

The waste form selected for plutonium is a zirconolite-rich ceramic with a waste loading of 12 wt% plutonium as the element as described by Ebbinghaus. The manufacturing process is stated as cold pressing and sintering at 1325 - 400°C.

The waste form selected for actinides, with or without plutonium, but not including uranium, is SYNROC-C as described by Lee et al.¹⁹ The main minerals in SYNROC-C are hollandite, zirconolite and perovskite. Zirconolite and perovskite are the major hosts for long-lived actinides, such as plutonium. Physical properties for SYNROC-C are given by Lee et al.;¹⁹ the pertinent information is a waste loading of 20 wt%.²³ The synthesis route is that of calcining precursor powders under reducing conditions, hot pressing, and then sintering at 1100 – 1170°C.

The waste form selected for the bulk of the fission products is borosilicate glass because the radiotoxicity of these decreases to less than that of natural uranium in centuries and thus long-term repository performance assurance is not necessarily required. The fission-product waste loading in borosilicate glass used by Rowe et al., is approximately 10 to 12 wt% fission-product elements in a high-level radioactive waste stream that comprises a 22 to 23 wt% oxide loading in the glass.

This information points out the reality that there is no such thing as a pure fission-product waste stream; there will always be "process and additive" chemicals introduced in the separations process. A summary of

the radioelements selected for partitioning, waste forms and pertinent physical properties is given in Table I.

Waste Elements			Density, gm/cc	Elemental Loading, wt %	
Iodine	Lead apatite	l apatite Reactive sintering		1.3	
		under 25 MPa at 700°C			
Plutonium	Ceramic,	Cold pressing and sinter	4.5	12	
	zirconolite rich	at 1325 - 1400°C			
Actinides	SYNROC-C Hot pressing and sint		4.3	20	
		at 1100 - 1170°C			
All Other	Borosilicate	Melting	2.7	~10%	

1000 - 1200°C

Table I. Selected Partitioned Elements, Waste Forms and Physical Properties.

VII. COMPARISON OF DIRECT DISPOSAL, COMPLETE PARTITIONING, AND LIMITED RECYCLE OF MOX AND UOX

glass

Fission

Products

The basis for comparison is a repository size of 70,000 metric tons (MT) of heavy-metal equivalent waste³ as spent nuclear fuel from light water reactors with a burn-up of 50 GWd/ton. This burn-up is chosen to reflect the possibility of the future average burn-up but does not affect the 70,000 ton capacity. If the burn-up changes then the fission-product mass in the waste will change accordingly. The fission-product mass concentration in spent nuclear fuel is very close to one wt% per 10 GWd/ton.²⁷ The mass of fission products is the same for all three comparison cases illustrated here.

All three cases produce the same mass of fission products based on a burn-up of 50 GWd/ton, or there will be 3,500 metric tons of fission products. Clearly, for the case of direct disposal the fission products and the actinides all remain in the spent nuclear fuel. For the case of partitioning these masses also are the same. The actual disposed mass of spent nuclear fuel is approximated by that of uranium dioxide so that 70,000 metric tons of heavy metal becomes 79,411 metric tons of oxide equivalent.

Limited recycle, or once-through recycling of recovered uranium and plutonium from reprocessing, occurs when discharged fuel derived from enriched natural uranium (eNatU) is reprocessed to recover uranium and plutonium and these elements are subsequently used, or recycled, to produce fuel. The

recovered, or reprocessed, uranium is enriched (eRepU) to a slightly higher enrichment relative the eNatU initially charged due to ²³⁶U. The recovered plutonium is fabricated into a mixed oxide of plutonium and uranium (MOX). The eRepU and MOX are used only once as fuel and upon discharge stored or disposed, but for the purpose of this illustration the discharged radioelements partitioned for disposal. The concept of limited recycle is sometimes used to illustrate that a savings in natural uranium consumption can be realized that is on the order of 25%. This savings appears to be derived from the observation that approximately eight LWRs produce sufficient plutonium to yield MOX fuel for one LWR core, 28 and also that the eRepU yields fuel for approximately one LWR core. Hence the initial eight LWRs fueled with eNatU upon discharge and reprocessing yield two LWR cores, or natural uranium consumption for the next cycle has been reduced by 25%. However, the next cycle does not have eight LWRs fueled with eNatU, but only six, and these six LWRs are the source of recovered plutonium and uranium for the next cycle; clearly the natural uranium savings will diminish as will the masses of recovered plutonium and uranium. As the recovery and recycling goes on it can be shown (with some simplifying assumptions) that this recycling reaches an equilibrium state, and it is this equilibrium state that is of interest with respect to the reduction in plutonium mass for disposal from the once-used MOX in limited recycle.

Given these two parameters, the number of LWRs required to produce sufficient plutonium to obtain one MOX core and the fission-product mass as

a function of burn-up, a parameterized simplified equilibrium-state material balance for the case of limited recycle can be derived with the following additional parameters. These additional parameters are the initial uranium enrichment required to achieve a specified burn-up, the discharge ²³⁵Û assay, and the required incremental enrichment for the eRepU to compensate for the presence of ²³⁶U (Ref. 29). The independent variables for this illustration are eight LWR cores produce sufficient plutonium for one MOX core, 50 GWd/ton, 4.6 wt% eNatU, 1.0 wt% fission products per 10 GWd/ton, ²³⁵U discharge assay of 0.77 wt%, and eRepU at 5.0 wt% 235U. This vields an equilibrium state for limited recycle where the MOX feed mass is 0.818 core (not 1.0), the eRepU feed mass is 0.637 core (not 1.0), and an eNatU mass of 6.545 cores; these add up to 8.0 cores. This calculation can be done for any range and combination of parameters. The displacement of natural uranium required relative to no recycle is approximately 18%. The net plutonium concentration reduction in discharged MOX is estimated as 25% from calculated results by Bays et al.³⁰ Using this plutonium concentration change and an initial MOX plutonium concentration of 10% (or 8%), the net reduction in plutonium disposal mass compared to no recycling is calculated as approximately 28%. For the mass of minor actinides use the results of Bays et al., 30 as an approximation, of 0.5 wt%, and for the plutonium concentration in discharged eNatU and eRepU use 1.2% (or 1%) throughout this illustration. The mass of stable iodine produced at the indicated burn-up as a fraction of the discharge mass is approximately 0.00273 (Ref. 30). These numbers are approximate, and there appears to be no single equilibrium-state material balance for limited recycle with a consistent set of assumptions and computations carried through the calculation. An example of this calculation was done by Rowe et al., ²⁶ using the material-balance simulator NUWASTE where the annual production of spent nuclear fuel in the United States reactor fleet was approximately matched by the reprocessing capacity; the number of LWRs required to yield sufficient MOX for one reactor core was determined to be approximately 8.3 which compares reasonably with the numbers used in this study.

The disposal masses for the three cases are summarized as follows. The mass of direct-disposal spent nuclear fuel is 70,000 tons as heavy metal, or approximately 79,411 tons as the oxide. The mass of radioelements from partitioning the entire spent-fuel mass is 3,500 tons of fission products as elements, 191 tons of iodine (included in the fission-product mass, but partitioned), 840 tons of elemental

plutonium, and 350 tons of other elemental actinides. The mass of radioelements from partitioning the discharge mass for limited recycle is the same as that for the entire mass except for the plutonium mass which is now 605 tons. These partitioned masses now have to be put into appropriate waste forms to obtain the respective disposal masses according to waste-form loadings. Calculated results for the disposal masses are summarized in Table II.

VIII. CONCLUSIONS AND OBSERVATIONS

The material-balance scenarios illustrated in Table II are i.) direct disposal of spent nuclear fuel, ii.) partitioning of the direct-disposal spent nuclear fuel mass, and iii.) limited recycle with partitioning. Comparison of the partitioning cases with direct disposal illustrates that the partitioned waste-form masses destined for a repository with long-term performance assurance total approximately 28% of the direct-disposal mass with no recycle, and 26% with limited recycle. Iodine contributes the most mass to the partitioned waste form mass.

Limited recycle of MOX from LWR spent nuclear fuel does not offer any significant advantage with respect to the disposal of plutonium because irradiation of MOX does not significantly decrease the plutonium concentration at discharge. The plutonium concentration is only reduced by about 25%, and this is not expected to change much for this particular limited recycle case in LWRs. Using more sophisticated reactor and material balance predictions are not expected to make any significant changes due to the presence of the even-numbered plutonium isotopes in the fuel composition, these will only increase in concentration upon irradiation.

The radiotoxicity basis for selecting radioisotopes to partition appears to be quite well documented, but there needs to be an integration of the selected radioisotopes as elements with the various separation processes so that these processes can yield the selected elements in a chemical form that can be used to manufacture the desired waste form. Most of the fission products will not be available in pure form but in a mixture of "process and additive" chemicals that will be part of the waste loading in the final waste form. Process studies will identify the various separations product streams that the selected radioelements will occur in and the additional chemical processing that will yield a waste form containing the radioelement and these processing chemicals. An example of this is the capture of iodine on silver mordenite, and the subsequent incorporation of this iodine in lead

Table II. Disposal Mass Summary.

Waste	Long-term	Total-Partitioned	Limited-Recycling	
Identification	performance	As Elemental,	As Waste Form,	Waste Form
	assurance?	MT	MT at % Loading	Disposal Mass, MT
Direct-Disposal	Yes	70,000 MT	79,411 MT, as oxide	n/a
Spent Nuclear			equivalent	
Fuel				
Iodine	Yes	191	14,692 at 1.3%	Same
Plutonium	Yes	840	7,000 at 12%	4,900
Minor Actinides	Yes	350	1,750 at 20%	Same
Fission Products	No	3,309	33,090 at 10%	Same
Recovered	No	65,310	74,091 as oxide	Same
Uranium				
	79,411			
To	56,532			
Total par	23,442			
Total par	21,342			
Percent of	28%			
Percent of	26%			

apatite. Additional processing has to occur, or another recovery technology used, to make the iodine available to be incorporated into the apatite.

Based on the extensive literature on nuclear waste forms, there has been a significant focus on the fundamental chemistry, durability and radiation stability of these materials. As such there appears to be a lack of pertinent engineering parameters describing waste forms in terms of waste loading and density (used to estimate waste volumes). These parameters will be required to complete a material balance to predict the final waste masses that will be subject to disposal.

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