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**Disposal Options For Bumer Ash
From Spent Graphite Fuel
(Final Study Report 11/93)**

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DISPOSAL OPTIONS FOR BURNER ASH FROM SPENT GRAPHITE FUEL

EXECUTIVE SUMMARY

This report presents the results of a scoping study of alternate means for processing and disposal of fissile-bearing particles and ash remaining after combustion of FSVR and PBR spent graphite fuels. Approximately 330 tonnes of spent graphite fuel having a minimum volume (close packed, unpackaged) of about 230 cubic meters require processing for final disposal. Combustion of fuel carbon could yield approximate reductions in spent graphite fuel mass and volume of 88 percent and 93 percent, respectively.

At present, there are no final repository WAC documents or other published criteria available to guide design, selection, and testing of final waste forms derived from spent fuel ash (SFA) or similar materials. Therefore, R&D efforts should focus on producing a waste form from SFA which is comparable to waste forms under development for high-level commercial and defense wastes.

Candidate SFA waste forms in decreasing order of estimated technical feasibility include glass-ceramics (GCs), polycrystalline ceramic assemblages (PCAs), and homogeneous amorphous glass.

Candidate SFA waste form production processes in increasing order of estimated effort and cost for implementation are:

- low-density GCs via fuel grinding and simultaneous combustion and waste form production in a slagging cyclone combustor (SCC).
- glass or low-density GCs via fluidized bed SFA production followed by conventional melting of SFA and frit.
- PCAs via fluidized bed SFA production followed by hot isostatic pressing (HIPing) of SFA/frit mixtures.
- High-density GCs via fluidized bed SFA production followed by HIPing of Calcine/Frit/SFA mixtures.

Near-term SFA waste form and process development efforts should focus on confirming the feasibility of one-step SCC vitrification, and on confirming the feasibility of incorporating SFA into Calcine/SFA glass-ceramics.

SFA waste forms must be selected on the basis of measured performance. Based on this study, laboratory and pilot scale development and characterization of all candidate SFA waste forms is warranted.

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APPENDIX 1: ASH COMPOSITION WORKSHEETS

DISPOSAL OPTIONS FOR BURNER ASH FROM SPENT GRAPHITE FUEL

1.0 INTRODUCTION

Three major disposal alternatives are being considered for Fort St. Vrain Reactor (FSVR) and Peach Bottom Reactor (PBR) spent fuels:

1. direct disposal of packaged, intact spent fuel elements,
2. removal of compacts to separate fuel into high-level waste (HLW) and low-level waste (LLW), and
3. physical/chemical processing to reduce waste volumes and produce stable waste forms.

For the third alternative, combustion of fuel matrix graphite and fuel particle carbon coatings is a preferred technique for head-end processing [1,2] as well as for volume reduction and chemical pretreatment [3] prior to final fixation, packaging, and disposal of radioactive residuals (fissile and fertile materials together with fission and activation products) in a final repository.

This report presents the results of a scoping study of alternate means for processing and/or disposal of fissile-bearing particles and ash remaining after combustion of FSVR and PBR spent graphite fuels.

1.1 ASH DESCRIPTION

Approximately 2,242 irradiated FSVR fuel elements containing about 1100 kg U and 22,600 kg Th, together with 1603 irradiated PBR fuel elements at ICPP, plus some additional PBR fuel elements at TAN containing about 325 kg U and 2,600 kg Th, will require conditioning for disposal.[4] This is equivalent to approximately 330 tonnes of spent graphite fuel having a minimum volume (close packed, unpackaged) of about 230 cubic meters.

Substantial reductions in the mass and volume of fuel requiring storage and disposal may be achieved by burning the carbonaceous components of the graphite fuel elements. It is estimated that combustion of all fuel carbon would yield approximately 15 cubic meters of ash material having a mass of approximately 40 tonnes. Combustion of fuel carbon, therefore, could yield approximate reductions in spent graphite fuel mass and volume of 88 percent and 93 percent, respectively.

The physical and chemical characteristics of the residual ash will depend on the types of fuel pretreatment and graphite combustion processes used. Whole-block (or rod) burning or "gentle" burning of crushed fuel elements in a bubbling fluidized bed will yield an ash comprised of intact pyrocarbon and SiC coated fuel particles (with U and Th carbide kernels) together

with a substantial fraction of broken particles, unburned graphite, and heavy metal oxide particulate and dust. Conversion of the reactive heavy metal carbides to their respective oxides would require breaking of the SiC particle coatings followed by combustion.

Ash from low temperature (<900 C') combustion of pulverized fuel or broken particles will be comprised of U and Th oxide dust and particles, broken SiC hulls, and less than one percent residual graphite. Ash from low temperature combustion would be suitable for subsequent chemical processing to separate heavy metals and fission products. Calculated compositions of ash from combustion of PBR and FSVR fuels are presented in Table 1-1. A more detailed listing of the major isotopes and their oxides is presented in Appendix 1.

TABLE 1-1: ASH COMPOSITION SUMMARY

	PBR kg	PBR %	FSVR kg	FSVR %
FP Oxides	35	1	240	<1
Thorium Oxide	2,945	88	25,270	70
Uranium Oxide	385	11	1,270	4
Other HM Oxides	5	<1	20	<1
Silicon Carbide	0	0	9,200	25
TOTAL	3,370	100	36,000	100

Both PBR and FSVR fuels yield ash which is substantially enriched in fissile uranium isotopes. FSVR ash uranium has a residual enrichment of about 45% U-235 and about 32% U-233. PBR Core I uranium has a residual enrichment of about 76% U-235 and 8% U-233; for PBR Core II, enrichment values are about 62% U-235 and 13% U-233. In addition, the potential commercial value of the ash uranium is virtually nil due to the presence of from about 8% (PBR Core I) to 13% (FSVR and PBR Core II) U-236.[5] Consequently, chemical processing of the ash to separate the heavy metals from the fission products would have the net effect of producing three waste streams (high-level liquid waste, heavy metal waste, and contaminated SiC hulls) all requiring further processing for stabilization and disposal. Reprocessing promises high costs for no benefit and is not judged to be part of a viable disposal option.

Based on cooling to April 1993, decay heat from the ash is estimated to be approximately 1.1 watts per kilogram. Similarly, specific activity for the ash is estimated to be approximately 200 Ci per kilogram.

Burning of crushed, unirradiated FSVR fissile particles at General Atomics Co. yielded an ash having bulk and tap densities of 2000 kg/m³ and 2600 kg/m³ respectively. FSVR fissile particle ash exhibited a very fine size distribution with 99 percent less than 300 microns and 50 percent less than 40 microns. The ash contained less than 0.2 percent carbon and had an angle of repose of approximately 44 degrees.[6]

Burning of crushed, unirradiated FSVR fertile particles yielded an ash having bulk and tap densities of 2670 kg/m³ and 3430 kg/m³ respectively. FSVR fertile particle ash was coarser than fissile particle ash with 99 percent less than 500 microns and 50 percent less than 100 microns. The ash contained about 1.4 percent carbon, 1 percent unbroken particles, and had an angle of repose of approximately 37 degrees.[7]

1.2 ASH DISPOSAL CRITERIA

DOE's Office of Civilian Radioactive Waste Management has only recently documented its general requirements for establishing a system for acceptance of waste for repository disposal.[8] Prior to disposal in a Federal Repository, PBR and FSVR spent fuel ash (SFA) must be rendered into a form which meets the Waste Acceptance Criteria (WAC) for the designated repository. WAC documentation will be prepared by the repository operator following construction and detailed characterization of the repository. At present, there are no final repository WAC documents available to guide design, selection, and testing of final waste forms derived from SFA or similar materials.

Existing published criteria [9-11] focus on borosilicate glass waste forms for fixation of high-level defense wastes and for acceptance of wastes at the Waste Isolation Pilot Plant (WIPP). These criteria do not specifically address high-level wastes containing significant quantities of fissile isotopes and heavy metal.

Some very general disposal criteria may be inferred from study of currently published EPA [12-16] and NRC [17-20] regulations and other guidance documents [21,22] for radioactive waste and fissile material management. An excellent review of these documents has been included in WINCO's evaluation of the direct disposal option for graphite fuels.[23] From these documents it may be inferred that:

1. The waste form must be a consolidated (monolithic) solid in a compatible sealed container having minimum free volume.

2. The waste form must remain subcritical under all credible conditions likely to be encountered.
3. The waste form must not be characteristically ignitable, corrosive, or reactive, and the waste form's package must not contain organic materials, free liquids or materials which are explosive, combustible, pyrophoric, or chemically reactive.
4. The waste form must be well characterized (and well documented) with respect to chemical, physical, and radiological properties.
5. The waste form, container, overpack, and repository system must meet specified performance requirements for containment of radionuclides and toxic materials over time.

Additional repository-specific criteria concerning heat generation, radioactivity, package configuration and dimensions, retrievability, surface contamination, labeling, shipping and handling, etc. will also apply.

2.0 CANDIDATE WASTE FORMS

Although the waste form, container, overpack, and repository are considered together to form an integrated waste disposal system, the waste form itself must be viewed as the key element of the system. Current regulations [12] require that the total system must limit cumulative releases of specified radionuclides for a period of 10,000 years following emplacement, and must do so with a specified probability of success. Since it is virtually impossible to attain a level of predictability, approaching certainty, for geologic phenomena far into the future, the isolation elements (container, overpack, repository) of the waste disposal system cannot provide a credible guarantee of containment.[24] On the other hand, preparation and accelerated laboratory testing of candidate waste forms can provide reliable and reasonably rapid (days to perhaps a decade) information concerning potential radionuclide releases under any credible future repository environment. Therefore, R&D efforts should focus on producing a waste form from SFA which is comparable to waste forms under development for high-level commercial and defense wastes.

Promising waste forms under development for high-level commercial and defense wastes include:[25]

1. Glasses via calcination, frit addition, blending, and melting;

2. Polycrystalline ceramic assemblages (PCAs) via calcination, frit and chemical addition, blending, and hot isostatic pressing (HIPing);
3. Glass-Ceramics (GCs) via calcination, frit addition, blending, and HIPing.

2.1 SFA GLASSES

Glass waste forms are produced by intimately mixing solid or slurried waste together with a low-melting glass frit. The resulting mixture is then heated and fused into a homogeneous glass. If the waste loadings are low enough (10-20 wt%) most of the fission product waste elements, as oxides, will dissolve into the glass and either help to form the glass network or be bound within it. Often, however, selected elements (such as Fe, Ru and Mo) may be present at concentrations above which they can be completely dissolved. If this occurs, additional undesirable phases may form in the glass matrix.

As shown in Table 2-1, glasses may be formed from a rather large selection of compounds traditionally designated as glass formers, intermediates, and modifiers.[26,27] As the name implies, glass formers usually comprise the dominant oxide species in the glass matrix. Intermediate oxides are also capable of contributing to the glass matrix but do not easily form glasses by themselves. High concentrations of intermediate oxides may even render a glass unstable and subject to devitrification. Glass modifiers are predominantly alkali or alkaline earth oxides added to control the melting, forming, and fabrication properties of the glass.

TABLE 2-1: PRIMARY GLASS CONSTITUENTS

GLASS FORMERS	INTERMEDIATES	MODIFIERS
B ₂ O ₃	Al ₂ O ₃	Li ₂ O
SiO ₂	Sb ₂ O ₃	Na ₂ O
GeO ₂	ZrO ₂	K ₂ O
P ₂ O ₅	TiO ₂	MgO
V ₂ O ₅	PbO	CaO
As ₂ O ₃	BeO	SrO
	ZnO	BaO
	Fe ₂ O ₃	

Calcined HLW from spent fuel reprocessing operations contains few components which are not true glass formers, intermediates, or modifiers or chemical analogues of these species. Consequently, it is relatively easy to make quality glasses having relatively high loadings of calcined HLW.[28] Making a quality glass from SFA promises to be a significantly more challenging task.

As shown in Table 1-1, oxides of thorium and uranium together with SiC comprise the majority of the SFA. These compounds are very refractory and are not chemically analogous to any of the primary glass constituents listed in Table 2-1. Consequently, incorporation of thorium, uranium, and SiC into a glass would likely require high dilution with frit and high-temperature, long-duration processing. Such processing would complicate capture and incorporation of volatile and semi-volatile fission products and would significantly reduce melter throughput rates. In addition, it is highly probable that residual thorium and SiC would remain undissolved and serve as nucleation sites to promote devitrification of the product glass.

Another vitrification alternative would be to incorporate only the SFA fission product oxides into a borosilicate glass and simply encapsulate the thorium, uranium, and SiC particulate in the resulting glass matrix. The product would probably be classified as a low-density glass-ceramic with only minor participation of waste constituents in formation of the glass matrix. Such a possibility is foreseen in the WAC document for the WIPP which allows waste material to be immobilized in borosilicate glass so that less than 1% by weight of the composite is particulate material less than 10 microns in diameter, or less than 15% by weight of the composite is particulate material less than 200 microns in diameter.[10]

Under these criteria, waste loading in the borosilicate glass is determined by the size distribution of encapsulated particles following the vitrification process. Unfortunately, this parameter cannot be accurately predicted using available information. The particle size distribution of the feed ash will change as smaller particles dissolve to extinction into the glass and as larger particles shrink via partial dissolution. In the worst case, if irradiated SFA has a sufficiently fine particle size distribution and dissolution of thorium, uranium, and SiC is negligible, achievable waste loadings may be as low as 1 or 2 percent SFA in glass. On the other hand, if dissolution of the major SFA constituents is negligible and the SFA particle size distribution is similar to that given in Section 1.1 above for crushed, burned-back fissile and fertile particles, then waste loadings of from 15 to 20 percent by weight might be acceptable.

Figure 2-1 shows total product glass volume as a function of waste loading over a reasonable range of expected waste loadings. As shown on the figure, waste loadings below about 6.4 percent

would rapidly result in generation of product glass volumes much greater than the volume of the unburned PBR and FSVR spent fuels. Likewise, waste loadings above about 6.4 percent would result in a net volume reduction with respect to the unburned fuel. At the extreme shown, a waste loading of 20 percent would result in a volume reduction of approximately 68 percent with respect to the unburned fuel volume.

At a SFA waste loading of 20 percent, approximately 120 standard canisters (2-ft diam. x 10-ft long, 85% full) would be required to contain about 75 cubic meters of product glass.[29] Each canister would hold about 1700 kg of glass containing about 11.2 kg fissile uranium (U-233 and U-235) and having total activity and decay heat of approximately 68,000 curies and 375 watts respectively. Slightly lower (18 percent) waste loadings might be required to meet the most restrictive single parameter mass limit (for $^{235}\text{UO}_2$) of 10.1 kilograms per standard size canister.[22]

Vitrification is probably the simplest and best developed means of producing suitable high-level waste forms. Properly sized SFA, glass frit, and chemical additives are thoroughly blended, charged to a waste canister, and melted "in-can" using either an inductively or resistively heated electrical furnace. Alternatively, the sized and blended charge is fed to a joule heated ceramic melter (JHCM) or arc melter. The molten glass product is then sampled, tested, and adjusted as necessary before being drained into a disposal canister or being fabricated into an alternative (buttons, marbles, etc.) waste form configuration.

In addition, it may be possible to simultaneously produce and vitrify the SFA using a slagging cyclone combustor (SCC).[3] In this process, pulverized graphite fuel and frit are fed tangentially, together with primary combustion air to one end of a horizontal, cylindrical furnace cavity. Fine graphite particles burn immediately in suspension and partially burned coarser particles are propelled by centrifugal force to the wall where they are trapped in a film of molten glass covering the wall. Secondary air is added tangentially along the length of the furnace to ensure that the graphite particles trapped in the glass film are burned to extinction before the molten material reaches the end of the combustion chamber and is drained to a JHCM "fining tank" or standard waste canister. SCC vitrification utilizes the graphite fuel values to produce a vitrified or low-density glass-ceramic waste form in a single processing step.

2.2 SFA CERAMICS

Polycrystalline ceramic assemblage (PCA) waste forms require partitioning of waste elements either into specific crystal phases in the host crystal structure or into solid solution within the host lattice. The objective is to formulate and

intimately mix a combination of chemical additives and waste components which can be densified at elevated temperature to form an assemblage of predominantly crystalline phases in which each waste radionuclide is incorporated into its most stable host lattice. The dense PCA resulting from such a process should simulate an assemblage of sedimentary-resistate minerals (rock) and exhibit leaching characteristics limited only by the inherently low solubility of the crystal phases comprising the PCA. [25]

Previous work has shown that it is a straightforward if somewhat tedious task to do the molecular engineering necessary to design a formulation which will allow incorporation of the approximately 40 major fission product radionuclides into various desirable assemblages of five to ten phases which constitute an appropriate polyphasic PCA waste form. [30-32] Typical examples of PCA waste forms include "supercalcine" in which the desired mineral species include apatite, nepheline, pollucite, scheelite, and sodalite; and "Synrock" in which fission product isotopes are incorporated into the high-titania minerals hollandite, zirconolite, and perovskite. Similarly, monazite has been proposed as the prototypic example of a mineral which survives "wet and warm" environments, and which can accommodate actenides and other nuclear waste ions. [34]

Calculations indicate that actenide waste loadings of approximately 70 percent should be achievable for synthetic monazite. Since actenides and SiC comprise about 99 percent of the SFA, overall waste loadings above 70 percent might be feasible if SiC hulls may be incorporated as separate-phase inclusions in the PCA waste form product. A predominantly monazite (sp. gr. 5.0-5.3) waste form having a SFA waste loading of 70 percent would yield approximately 11.5 cubic meters of PCA waste form having a mass of approximately 60 tons.

PCA blocks would likely be stacked in wide-mouth standard canisters having the interstitial volume backfilled with granular inert material. In this fashion, approximately 65 percent (0.57 cubic meters) of the canister's free volume could be occupied by the PCA waste form. [29] Under these assumptions, approximately 20 canisters would be required for disposal of the PCA waste form. Each canister would hold approximately 2850 kg of PCA waste form containing approximately 65 kg of fissile uranium isotopes and having a specific activity and heat generation of 400,000 curies and 2200 watts respectively. Single-parameter mass limits for the fissile oxides range from 10.1 kilograms for $^{235}\text{UO}_2$ to 51.2 kilograms for $^{238}\text{UO}_2$. [22] Therefore, it is likely that significantly lower waste loadings would be required in order to meet WAC limits based on standard canister size and single-parameter lower mass limits.

Production of PCA waste forms requires very fine grinding, proportioning, and dry mixing of SFA and chemical additives. The SFA/additive mixture is charged to a stainless steel can which is then evacuated and hermetically sealed. After sealing, the can and its contents are heated (1100-1150 °C) and subjected to high pressure (100-300 MPa) under argon to densify the softened mixture and form the desired PCA phases.[40,41]

2.3 SFA GLASS-CERAMICS

Glass-ceramic (GC) waste forms are essentially an hybrid of the glass and PCA waste forms. GCs incorporate inclusions of stable waste components into an amorphous, resistant glass matrix. High alumina or zirconia waste calcines, in particular, can be used to produce good GC waste forms.[35-39] Calcine is intimately blended with powdered frit and chemical additives and the mixture is densified by HIPing to produce the finished GC waste form. To date, high quality GCs have only been produced from calcines which contain large fractions of glass formers, intermediates and modifiers. Consequently, relatively small amounts of chemical additives and frit are sufficient to produce an abundant borosilicate glass matrix to encapsulate undissolved calcine components. An added benefit of using calcine components to form the glass matrix is that relatively high waste loadings of from about 65 to 85 percent are achievable. Typical GC densities range from 2900 to 3200 kilograms per cubic meter.

As described in Section 2.1, SFA contains virtually no components which may be classified as glass formers, modifiers, intermediates, or their chemical analogues. Consequently, efforts to produce quality GCs from SFA would likely be frustrated by very high requirements for borosilicate glass frit and would be subject to the particle size distribution and waste loading restrictions for glasses and low-density GCs as described in Section 2.1. Therefore, preparation of GCs from pure SFA is probably not a desirable option because it would entail the difficult processing associated with PCAs while achieving only the modest waste loadings associated with vitrified SFA.

However, high quality GCs might be made from a blend of calcine and SFA. More than 3000 cubic meters of alumina and zirconia calcine are currently in retrievable storage at the Idaho Chemical Processing Plant (ICPP). This inventory is equivalent to approximately 4,200 tonnes at an estimated bulk density of 1400 kilograms per cubic meter. Uniform blending of the SFA with this inventory would yield a concentration of SFA in calcine of approximately 0.95 percent. It is very unlikely that this small concentration of SFA would adversely affect the quality of GC waste forms produced from the blended calcine and SFA.

Approximately 3,330 canisters would be required to contain about 1,900 cubic meters of GC product from the combined calcine and

SVA. Each canister would hold approximately 1825 kg of GC waste form containing approximately 0.4 kg of fissile uranium isotopes and having a specific activity and heat generation of 109,000 curies and 340 watts respectively. Fissile content would be well below the most restrictive single parameter mass limit (for $^{235}\text{UO}_2$) of 10.1 kilograms per standard size canister.[22]
About 32 of the 3,330 required canisters may be attributed to disposal of the SFA fraction of the calcine/SFA blend.

Process requirements for fabricating GCs are similar to those for making PCA waste forms and typically include fine grinding, proportioning, dry blending, and HIPing.

2.4 SUMMARY OF WASTE FORM CHARACTERISTICS

Table 2-2 presents a summary of the major characteristics for each waste form based on maximum SFA loading (assuming no criticality constraints) and use of standard (2-ft diam. x 10-ft long) repository canisters.

TABLE 2-2: SUMMARY OF WASTE FORM CHARACTERISTICS

WASTE FORM PARAMETERS	Glass	PCA	GC
SFA Loading, percent	20	70	<1
Waste Volume, m ³	75	11.5	1900
Waste Mass, Mg	204	60	6077
Waste form Density, Mg/m ³	2.7	5.2	3.2
Volume Reduction ^a , percent	67	95	92 ^b
No. of Canisters Required	120	20	3300
Waste Inventory, kg/canister	1700	2850	1825
Activity, kCi/canister	68	400	109
Decay Heat, Watts/canister	375	2200	340
Fissile U, kg/canister	11.2	65	0.4

a. Volume reduction with respect to intact fuel volume.

b. Based on proportionate volume of SFA in SFA/calcine GC.

3.0 EVALUATION OF PROCESS OPTIONS

Figure 3-1 presents a summary of the major process steps required for each of four candidate SFA disposal options. Two options are shown for preparation of a vitrified waste form and one option each is shown for preparation of a PCA (predominantly monazite) and a GC (SFA/calcine blend) waste form:

OPTION 1: SCC VITRIFICATION -- Fraize mill and double roll crushers are used to grind fuel to a top size of about 250 microns in order to ensure that all SiC particle coatings are broken. Finely ground fuel and frit are fed to a SCC where carbon combustion, carbide conversion, and vitrification occur simultaneously in the combustion chamber.[3] Glass composition may be adjusted in an adjacent "fining tank" prior to canister filling, annealing, and sealing before shipment to the repository.

OPTION 2: CONVENTIONAL VITRIFICATION -- Following fuel size reduction, graphite burning and carbide conversion are performed in a circulating fluidized bed combustor (CBC).[3] CBC ash is packaged and placed in interim storage awaiting vitrification processing. SFA from interim storage is sized and blended with appropriate glass frit and chemical additives. The SFA/frit mixture is charged to a stainless steel canister, which is heated as required to vitrify the contents. Alternatively, the SFA/frit mixture may be vitrified in a JHCM or other melter, and the molten product discharged to the disposal canister which is then sealed and shipped to the repository.

OPTION 3: PCA PROCESSING -- Following fuel size reduction, graphite burning and carbide conversion are performed in a circulating fluidized bed combustor (CBC). CBC ash is packaged and placed in interim storage awaiting PCA processing. SFA from interim storage is sized and blended with appropriate mineral frit and chemical additives. The SFA/frit mixture is charged to stainless steel cans which are evacuated, sealed, and densified by HIPing under high-pressure argon. Densified PCA waste forms are stacked in disposal canisters which are backfilled with inert material before being sealed and shipped to the repository.

OPTION 4: SFA/CALCINE GC PROCESSING -- SFA from interim storage is sized and blended together with ICPP calcine, frit, and chemical additives. The mixture (containing <1 wt% SFA) is charged to stainless steel cans which are evacuated, sealed and densified by HIPing. The resulting GC waste forms are stacked in disposal canisters which are backfilled with inert material before being sealed and shipped to the repository.

3.1 DEVELOPMENT CONSIDERATIONS

Substantial development work will be required in order to formulate, produce, and characterize acceptable waste forms for all listed options. The waste form development program for Option 1 will require the most effort since it must proceed virtually "from scratch". Development of waste forms for the other options may draw substantially from ongoing glass, PCA, and GC development programs for commercial (West Valley) and defense (INEL, HANF, SRL, etc.) wastes. For all options, appropriate "molecular engineering" must be performed to establish a number of candidate formulations which are likely to yield an acceptable waste form. Each formulation must be produced in the laboratory or pilot plant and tested for a host of physical, mechanical, and chemical properties. Finally, the radiation stability of the most promising formulations must be determined by long-term testing of waste form samples made from actual radioactive waste. Specific development considerations for each option are presented below:

Option 1: SCC technology has been adapted for vitrifying heavy metals found in contaminated soils, and as the first step in a combustion/vitrification process for treating spent graphite potliner from aluminum smelting.[42,43] Since the SCC processing environment provides much higher temperature and turbulence during vitrification than other melters, refractory species are more likely to dissolve and become part of the amorphous glass network. High dissolution of refractory species will allow increased waste loading. Likewise, the higher temperatures of the SCC should permit preparation of high-silica glasses which have superior mechanical properties and leach resistance when compared to low-temperature borosilicate glass. Although high temperature vitrification might permit production of a superior, durable glass, it will also complicate problems associated with retention, capture, and recycle of semivolatile fission products. SCC processing of spent graphite fuels to produce an acceptable final waste form is a novel concept which will require an integrated program including waste form, process, and equipment development.

Option 2: In-can and JHCM melting of calcined waste and glass frit are well demonstrated technologies.[44-46] However, quality glasses prepared by these techniques usually have relatively low (10-20 wt%) waste loadings and are often degraded by formation of second phases (free metal, spinels, crystals, undissolved oxides, glass-in-glass, etc.) which may promote devitrification or otherwise compromise desirable physical and chemical properties of the glass. Because of their refractory nature -- urania and thorium melt at about 2880 °C and 3220 °C respectively, and SiC decomposes at about 2700 °C -- and their high concentrations in SFA, incorporation of these materials into a homogeneous,

amorphous, low-temperature, borosilicate glass will be virtually impossible at reasonable waste loadings.

Encapsulation of undissolved urania, thoria, and SiC in a borosilicate glass matrix may also be difficult to accomplish using conventional melter technologies. Urania and thoria are three to four times as dense as the calcine or glass frit normally used to form an amorphous glass matrix. Consequently, these species will begin to settle out as the glass components soften, melt, and become less viscous. In-can melting of SFA/frit mixtures would likely yield a relatively thin but very dense bottom layer comprised of undissolved urania, thoria, and SiC particulate covered by a thick upper layer of glass having composition virtually identical to that of the original frit. Since the particulate species in the bottom layer would be in contact with one another or separated by a minimal amount of interstitial glass matrix, this layer would likely exhibit very poor structural properties and leaching behavior. Similarly, if processed in a JHCM, the denser materials would probably settle to form an intractable bottom sludge with substantially different rheological and electrical properties than those of the bulk glass. Development of new melting and forming techniques may be required to produce an acceptable low-density glass-ceramic or particle-in-glass waste form.

Option 3: PCA waste form development can be accomplished using well demonstrated bench-scale equipment and techniques for sizing, blending, and HIPing of fine particles.[34-41] Process design and scale-up to achieve reasonable throughput while maintaining criticality safety will be a major development issue for this option.

Option 4: All development tasks for this option may be included as supplementary tasks under the overall development program for permanent disposal of ICPP stored calcine.

3.2 FACILITY CONSIDERATIONS

For each option, facilities and equipment will be required to perform the top-level functions shown on Figures 3-2 through 3-5. To the extent that the number of top-level functions reflects expected facility and equipment costs, Options 1 and 4 enjoy a considerable advantage over Options 2 and 3. Option 1 (SCC Vitrification) is the only continuous process and offers high throughput using relatively simple and compact equipment. In addition, Option 1 is the only option which does not require facilities for interim storage of packaged SFA. Option 4 appears attractive because it is shown as a "head-end" for the planned ICPP Calcine Stabilization Facility. If Option 4 were to be evaluated as a "stand-alone" option for disposal of SFA it undoubtedly would require the greatest investment in facilities and equipment of all options presented.

3.3 OTHER CONSIDERATIONS

Systems engineering and design of facilities to accomplish any of the SFA treatment options will be guided by a predetermined set of principal plant factors which specify facility performance requirements (lifetime, throughput, availability, reliability, maintainability, etc.), safety and environmental requirements, (NEPA, RCRA, TSCA, CWA, CAA, OSHA, etc.) and nuclear materials management requirements (accountability, containment, ALARA, criticality safety, accident dose-risk, etc.). Since the principal plant factors have yet to be established for any of the SFA disposal options, comparative evaluation at present must be based on more general empirical criteria. Experience has shown that effort and costs for design, construction, licensing, and operation of nuclear processing systems and facilities are reduced when the following guidelines or rules-of-thumb are applied:

1. Primary containment of radioactive materials is provided by the process equipment or vessels. Uncontained material transfers into, out of, between, or within processes are avoided.
2. Process equipment and vessels are maintained at negative pressure with respect to the hot cell and the hot cell is maintained at negative pressure with respect to the facility. Flows required to maintain necessary pressure differentials are minimized.
3. Exposure of pressure boundaries to heat, corrosion, or cyclic stress is avoided.
4. High-pressure and/or high-temperature processing operations in-cell are avoided.
5. Processing equipment and operations are simple and amenable to remote operation. Operations requiring in-cell manipulation of materials or equipment are avoided.
6. Processes are dynamically stable, control functions are minimized, and equipment is designed to "fail safe". Recovery from failures is easy.
7. Production or handling of radioactive powders, dusts, aerosols, or volatile species are avoided.
8. Criticality control utilizes simple physical methods with minimum reliance on sampling, analyses and administrative controls.
9. Total power and potential energy of unit operations are minimized.

10. Production of secondary wastes and contaminated equipment is minimized.

11. Use of raw materials and energy resources are minimized.

Table 3-1 presents an extremely subjective forced ranking evaluation of the four SFA disposal processes with respect to the aforementioned eleven guideline criteria. The top level functional flow diagrams (Figures 3-2 through 3-5) were vetted against the criteria to arrive at a forced ranking of how well the four options complied with each criterion. For each criterion the options were given a numerical ranking from 1 to 4 in order of their perceived compliance (best to worst) with the guideline criterion.

The sum of the rankings for each option taken over all criteria provides a relative estimate of the cost and effort required to design, construct, license, and operate facilities for each option. Options 1 and 4 appear to exhibit a clear advantage over Options 2 and 3. However, it should be noted that the low score for Option 4 is attributable to its being considered only as an "head-end" process for the planned ICPP Calcine Stabilization Facility as shown on Figure 3-5. Also, the difference in ranking between Options 2 and 3 is an artifact of the forced ranking procedure which does not allow for a "tie" ranking.

TABLE 3-1: FORCED RANKING EVALUATION OF PROCESS OPTIONS
RANKING FROM 1 TO 4: BEST TO WORST COMPLIANCE WITH CRITERIA

	Option 1 SCC Vit.	Option 2 Glass	Option 3 PCA	Option 4 GC/Calc.
1. Containment	1	2	4	3
2. Delta P & Flow	1	2	4	3
3. Boundary Exposure	1	3	4	2
4. High P, T, Ops.	2	3	4	1
5. Simple, Remote	1	3	4	2
6. Stable, Fail Safe	4	3	2	1
7. Rad. Dusts	1	3	4	2
8. Criticality	1	3	4	2
9. Power, Energy	3	2	4	1
10. Wastes	1	3	4	2
11. Resources	1	4	3	2
TOTAL	17	31	41	21

4.0 CONCLUSIONS AND RECOMMENDATIONS

This report presents the results of a scoping study of alternate means for processing and/or disposal of fissile-bearing particles and ash remaining after combustion of FSVR and PBR spent graphite fuels. Conclusions and recommendations from the study are:

1. There exists very little technical data or published information to guide development of final waste forms derived primarily from refractory actenide oxides and SiC as found in FSVR and PBR spent fuel ash (SFA).
2. To ensure the highest probability of acceptance at a final repository, SFA waste forms must meet or exceed the performance specifications for defense or other commercial waste forms derived from high-level liquid wastes, sludges, salts, and calcines.
3. Candidate SFA waste forms in decreasing order of estimated technical feasibility include glass-ceramics (GCs), polycrystalline ceramic assemblages (PCAs), and homogeneous amorphous glass.
4. Candidate SFA waste form production processes in increasing order of estimated effort and cost for implementation are:
 - low-density GCs via fuel grinding and simultaneous combustion and waste form production in a slagging cyclone combustor (SCC).
 - glass or low-density GCs via fluidized bed SFA production followed by conventional melting of SFA and frit.
 - PCAs via fluidized bed SFA production followed by hot isostatic pressing (HIPing) of SFA/frit mixtures.
 - High-density GCs via fluidized bed SFA production followed by HIPing of SFA/Calcine/Frit mixtures.
5. Near-term SFA waste form and process development efforts should focus on confirming the feasibility of one-step SCC vitrification, and on confirming the feasibility of incorporating SFA into Calcine/SFA GCs.
6. SFA waste forms must be selected on the basis of measured performance. Based on this study, laboratory and pilot scale development and characterization of all candidate SFA waste forms is warranted.

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Figure 2-1: Estimated SFA Glass Volume
Glass sp.gr. = 2.7

NOTES:

1. Volumes are for wasteform only based on maximum waste loading, without regard for backfilling, overpacking, or criticality requirements.
2. PCA = Polycrystalline Ceramic Assemblies
3. Off-gas treatment, material transport, and other major auxiliary and support systems are omitted for clarity.

DRAKE ENGINEERING INCORPORATED

INITIAL	DATE	SUMMARY OF SPENT FUEL ASH (SFA) DISPOSAL OPTIONS		
DES.	<i>AND</i> 11/22/93			
RD				
DRAW	<i>AND</i> 11/22/93			
RND				
CHECK	<i>VMD</i>			
VMD	11-22-93			
PROJECT		SCALE	SKETCH NUMBER	REV.
GFCS		NA	FIGURE 3-1	0

BOG = BURNER OFF-GAS

SCC = SLAGGING CYCLONE COMBUSTOR

VOG = VESSEL OFF-GAS

ORAKE ENGINEERING INCORPORATED

INITIAL	DATE
DES. RND	11/22/93
DRAW RND	11/22/93
CHECK VMD	11-22-93

FUNCTIONAL FLOW
DIAGRAM FOR
OPTION 1
SCC VITRIFICATION

PROJECT	SCALE	SKETCH NUMBER	REV.
GFCS	NA	FIGURE 3-2	0

BOG • BURNER OFF-GAS
 MOG • MELTER OFF-GAS
 SFA • SPENT FUEL ASH
 MOG • MELTER OFF-GAS

DRAKE ENGINEERING INCORPORATED				
INITIAL	DATE	FUNCTIONAL FLOW DIAGRAM FOR OPTION 2 CONVENTIONAL (IN-CAN) VITRIFICATION		
DES.	<i>RND</i>			
RND	<i>11/22/93</i>			
DRAW	<i>RND</i>			
CHECK	<i>VMD</i>			
VMD	<i>11-22-93</i>			
PROJECT		SCALE	SKETCH NUMBER	REV.
GFCS		NA	FIGURE 3-3	0

BOG : BURNER OFF-GAS
 HOG : HIP PROCESS OFF-GAS
 PCA : POLYCRYSTALLINE CERAMIC ASSEMBLAGE
 SFA : SPENT FUEL ASH
 VOG : VESSEL OFF-GAS

DRAKE ENGINEERING INCORPORATED

INITIAL	DATE	FUNCTIONAL FLOW DIAGRAM FOR OPTION 3 PCA PROCESSING			
DES.	AMD				
RND	11/22/93				
DRAW	AMD				
RND	11/24/93				
CHECK	VMD				
VMD	11-22-93				
PROJECT		SCALE	SKETCH NUMBER	REV	
GFCs		NA	FIGURE 3-4	0

BOG : BURNER OFF-GAS
 SFA : SPENT FUEL ASH
 VOG : VESSEL OFF-GAS

DRAKE ENGINEERING INCORPORATED

INITIAL	DATE	FUNCTIONAL FLOW DIAGRAM FOR OPTION 4 SFA/CALCINE GC PROCESSING		
DES.	<i>AMD</i>			
RND	11/22/93			
DRAW	<i>AMD</i>			
RND	11/22/93			
CHECK	VMD			
VMD	11-22-93			
PROJECT		SCALE	SKETCH NUMBER	REV.
GFCS		NA	FIGURE 3-5	0

APPENDIX 1

ASH COMPOSITION WORKSHEETS

Conversion of Fuel Isotopes to Oxides

	-----Isotope-----					-----Oxide-----		CURIES	Cu %
	MW	g mols	grams	wt %	Ratio	grams	wt %		
H	3	0.28	0.85	0.00%	0.5	3.12	0.00%	8,221	0.10%
C	14	1.94	27.13	0.00%	2.0	89.15	0.00%	121	0.00%
Kr	85	10.21	867.85	0.00%	0.0	867.85	0.00%	340,472	4.31%
Rb	87	125.59	10925.90	0.04%	0.5	11930.58	0.04%	0	0.00%
Sr	90	203.05	18274.68	0.07%	1.0	21523.51	0.07%	2,547,513	32.22%
Zr	93	266.53	24786.92	0.09%	2.0	33315.75	0.11%	622	0.01%
Yc	99	233.66	23132.54	0.09%	3.5	36217.61	0.12%	393	0.00%
Ru	106	0.41	43.78	0.00%	2.0	56.99	0.00%	144,583	1.83%
Sb	125	0.34	42.88	0.00%	2.5	56.60	0.00%	44,962	0.57%
I	129	51.53	6647.63	0.03%	0.0	6647.63	0.02%	1	0.00%
Cu4	134	5.66	757.77	0.00%	0.5	803.01	0.00%	980,512	12.40%
Cu5	135	61.58	8313.03	0.03%	0.5	8805.65	0.03%	8	0.00%
Cu7	137	223.85	30666.90	0.12%	0.5	32457.67	0.11%	2,665,488	33.71%
Ce	142	358.59	50919.78	0.19%	2.0	62394.66	0.21%	0	0.00%
CePr	144	2.35	338.69	0.00%	2.0	413.95	0.00%	1,078,066	13.63%
Nd	144	303.51	43706.02	0.17%	1.5	50990.35	0.17%	0	0.00%
Sn47	147	46.57	6845.79	0.03%	1.5	7963.47	0.03%	3	0.00%
Sn51	151	10.16	1533.56	0.01%	1.5	1777.30	0.01%	40,341	0.51%
PPSUM		1,906	227,832	0.86%		276,315	0.92%	7,851,306	99.29%
Th	232	106881.30	24796461.60	93.79%	2.0	28216663.20	93.54%	3	0.00%
U32	232	0.32	74.70	0.00%	2.7	88.61	0.00%	1,672	0.02%
U33	233	1582.01	368609.26	1.39%	2.7	436192.90	1.45%	2,305	0.03%
U34	234	201.91	47247.88	0.18%	2.7	55873.64	0.19%	294	0.00%
U35	235	3034.52	713111.03	2.70%	2.7	842745.51	2.79%	1	0.00%
U36	236	738.07	174183.34	0.66%	2.7	205713.48	0.68%	27	0.00%
U38	238	394.50	93891.48	0.36%	2.7	110744.60	0.37%	0	0.00%
Np	237	48.84	11576.03	0.04%	2.0	13139.04	0.04%	8	0.00%
Pu38	238	12.17	2895.75	0.01%	2.0	3285.09	0.01%	49,572	0.63%
Pu39	239	7.73	1846.75	0.01%	2.0	2094.02	0.01%	115	0.00%
Pu40	240	3.79	910.56	0.00%	2.0	1031.97	0.00%	207	0.00%
Am	241	2.41	579.85	0.00%	2.0	656.84	0.00%	1,987	0.03%
TRUSUM		112,908	26,211,388	99.16%		29,888,229	99.08%	56,191	0.71%
TOTAL		114,813	26,439,220	100.00%		30,164,544	100.00%	7,907,497	100.00%
ThSUM		106,881	24,796,462	93.79%		28,216,663	93.54%	3	0.00%
USUM		5,951	1,397,043	5.28%		1,651,270	5.47%	2,627	0.03%
PuSUM		24	5,653	0.02%		6,411	0.02%	49,894	0.63%

Spent Graphite Fuel Composition Summary
HMSUN.WQ1

	FSVR	PB-1	PB-2	Total	Total
FSVR Equivalent FK	2208	165.5	139.2	2512.7	

Mols kg

Mols Th	232	95720.4	6901.4	4259.5	106881.3	24796.4616
Mols Th		89.56%	6.46%	3.99%	100.00%	

Mols

U-232	232	0.31	0	0.01	0.32	0.07424	0.01%
U-233	233	1440.71	74.63	66.67	1582.01	368.60833	26.58%
U-234	234	174.95	13.04	13.92	201.91	47.24694	3.39%
U-235	235	2048.08	674.91	311.53	3034.52	713.1122	50.99%
U-236	236	601.88	67.69	68.49	738.06	174.18216	12.40%
U-238	238	300.92	55.44	38.14	394.5	93.891	6.63%

Mols U	4566.85	885.71	498.76	5951.32	
Mols U	76.74%	14.88%	8.38%	100.00%	
U-235 Enrichment	44.85%	76.20%	62.46%	50.99%	
U-233 Enrichment	31.55%	8.43%	13.37%	26.58%	

kg U	1071.65709	208.21356	117.24422	1397.11487	
kg U	76.71%	14.90%	8.39%	100.00%	

	PB kg	PB %	FSVR kg	FSVR %	TOTAL kg	TOTAL %	CI	WATTS
FP OXIDES	33.16	12.00%	243.14	88.00%	276.30	0.70%	7,843,404	41,533
THORIUM OXIDE	2945.82	10.44%	25270.88	89.56%	28216.70	71.69%	3	0
URANIUM OXIDE	384.09	23.26%	1267.20	76.74%	1651.29	4.20%	4,299	414
OTHER HM OXIDES	3.43	17.00%	16.77	83.00%	20.20	0.05%	53,131	1,758
SiC	0.00	0.00%	9192.20	100.00%	9192.20	23.36%	0	0
TOTAL	3366.50	8.55%	35990.19	91.45%	39356.69	100.00%	7,900,837	43,705

	PB kg	PB %	FSVR kg	FSVR %
FP OXIDES	33.16	0.98%	243.14	0.68%
THORIUM OXIDE	2946.48	87.51%	25270.22	70.22%
URANIUM OXIDE	384.09	11.41%	1267.20	3.52%
OTHER HM OXIDES	3.43	0.10%	16.77	0.05%
SiC	0.00	0.00%	9192.20	25.54%
TOTAL	3367.16	100.00%	35989.53	100.00%

DATE

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END