DEVELOPMENT OF A GROUT-BASED STABILIZATION METHOD FOR SPENT HIGH-TEMPERATURE GAS-COOLED REACTOR FUEL

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ABSTRACT

The preliminary results of a study of potential predisposal stabilization methods for spent high-temperature gas-cooled reactor (HTGR) fuel are presented. The paper comprises (1) methods for reducing the volume of fuel prior to disposal, (2) an assessment of existing stabilization methods for use with spent HTGR fuel, (3) development of a potential reference HTGR fuel stabilization flowsheet, and (4) the results from laboratory preparation and testing of concrete stabilized, simulated HTGR fuel waste forms.

INTRODUCTION AND BACKGROUND

Nuclear fuel is manufactured in the U.S. by GA Technologies Inc. (GA) for use in high-temperature gas-cooled reactors (HTGRs). An HTGR fuel element consists of a prismatic hexagonal graphite block, containing fuel rods fabricated from graphite and fuel particles. The current reference fuel particles are coated fissile (UCO) and fertile (ThO_2) kernels.[1] The reprocessing of HTGR fuel results in the separation and recovery of fissionable materials and, together with refabrication of fuel, enables closure of the fuel cycle.

As shown in Fig. 1, a recently issued HTGR Spent Fuel Treatment Program Plan presents a decision tree approach to the HTGR fuel treatment.[2] Alternatives to reprocessing are delineated in the plan. This change in the HTGR spent fuel treatment is responsive to the many technological, political, institutional, and economic factors that impact the treatment decision process. One of the options for spent HTGR fuel treatment is the permanent disposal of whole elements or volume-reduced fuel. Economic studies conducted at GA show that volume reduction prior to storage and disposal is cost effective and simplifies handling and transportation requirements.[3] Significant reductions in the volume and weight of spent HTGR fuel are achievable due to large amounts of carbonaceous material contained in the fuel elements. Table I gives the relative numbers, weights, and volumes of the components of an HTGR prismatic fuel element.

In addition to the economic and handling advantages derived through volume reducing spent HTGR fuel, volume reduction also offers the potential for separating spent fuel elements into discrete high-level waste (HLW) and low-level waste (LLW) components. If an effective separation of HTGR fuel elements can be accomplished, the LLW component can be disposed of with little or no pretreatment. The relatively small volume of HLW produced would probably require some form of stabilization prior to geologic disposal. Stabilization is defined herein as a post-reactor nuclear material treatment that utilizes chemical and physical processes to effect beneficial changes in nuclear waste forms. The ultimate goal of stabilization is to protect the environment and to isolate mankind from the harmful effects of nuclear materials. To this end, governments and the nuclear industry have sponsored the development of methods for stabilizing nuclear waste and disposing of these stabilized wastes in geologic repositories.

METHODS FOR REDUCING THE VOLUME OF HTGR FUEL

Reference Volume-Reduction Method

GA has been developing methods, technology, and equipment for treating spent HTGR nuclear fuel since the late 1960s. This effort has resulted in the development of a volume-reducing reference reprocessing treatment, which includes the head-end processes (i.e., graphite matrix crushing, matrix burning, separation of fissile and fertile particles by

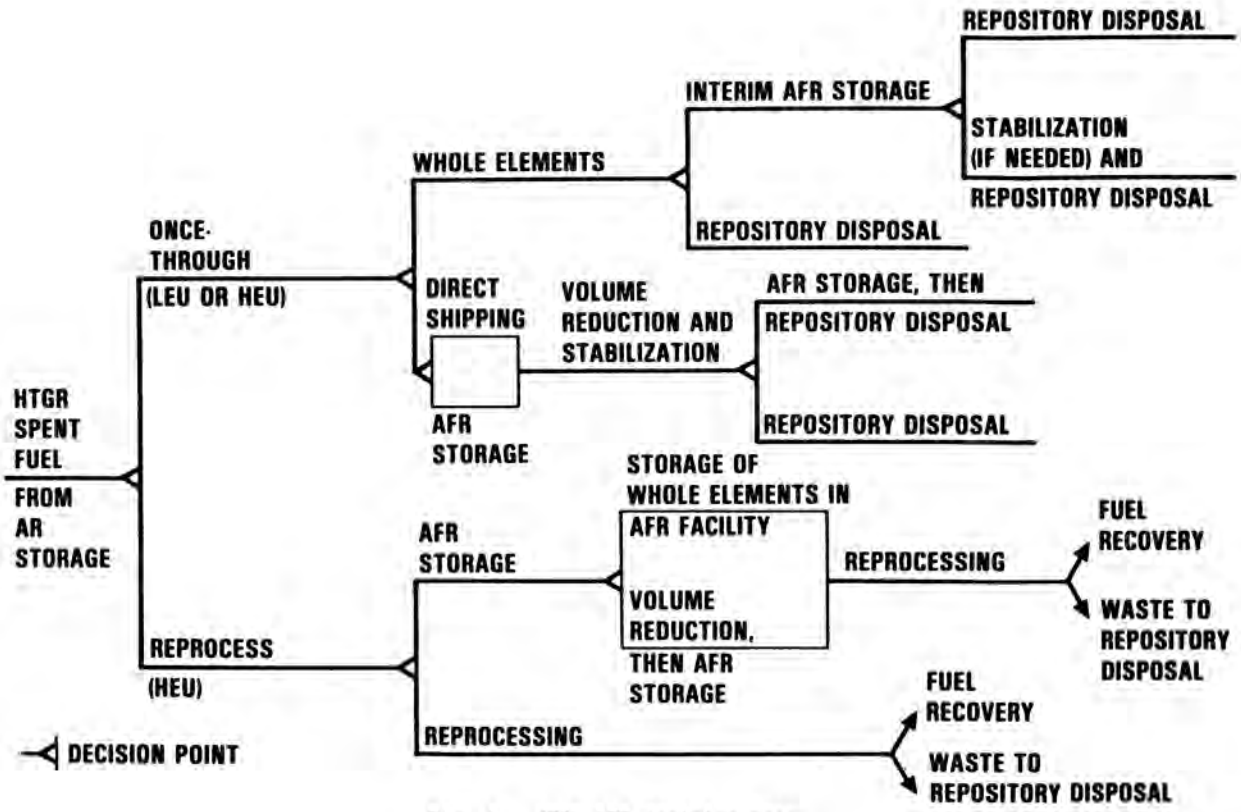

Fig. 1. HTGR-SFT decision tree.

TABLE I

Selected HTGR-LEU Fuel Element Properties

Block	
Volume	87 liters
Weight	99 kg (empty) 129 kg (full)
Fuel rod per block	
Number	2052
Fissile particles per block	
Number	9×10^6
Weight	5.1 kg
Volume	1.76 liters
Fertile particles per block	
Number	1×10^7
Weight	11.0 kg
Volume	2.65 liters

classification, particle crushing, and secondary burning of crushed coated fuel particles). If reprocessing is to be done, these volume-reducing operations are sequentially followed by fuel particle dissolution, feed acidity adjustment of dissolver solutions, and solvent extraction.

In the reference volume-reducing flowsheet shown in Fig. 2, the solid HLW residual from the head-end primary burner operation is much lower in weight and volume than the original fuel element and is composed primarily of fissile and fertile fuel particles. For an HTGR fuel element containing low-enriched uranium (LEU), primary burning yields a residual solid which is 15 wt % and 5 vol % of the original feed.

Alternative Volume-Reduction Methods

Mechanical volume-reduction methods have also been proposed for HTGR fuel treatment. Figure 3 gives the flowsheet for a rod-removal, volume-reduction process. This flowsheet contains a fuel rod removal operation in lieu of whole element crushing and burning. The removed fuel rods (representing a volume reduction of about five to one on a whole-rod-to-fuel-element basis) are fed to a waste stabilization process, or alternatively, crushed and burned back to the silicon carbide-coated particles in a fluidized bed combustor. The burned-back particles then are fed to reprocessing operations or storage/disposal, depending on the fuel treatment option exercised.

Volume-Reduction Flowsheet Material Characterization and Estimated Throughputs

If a volume-reduction treatment is used prior to reprocessing or waste disposal, the bulk quantity material throughputs for each of the process streams must be determined. Existing literature was used to determine the material throughputs for the process streams of Fig. 1. Annual processing of 50,000 HTGR-LEU fuel elements was asssumed. Table II provides the material characterization and throughput for the reference crush-burn flowsheet. These data are useful in

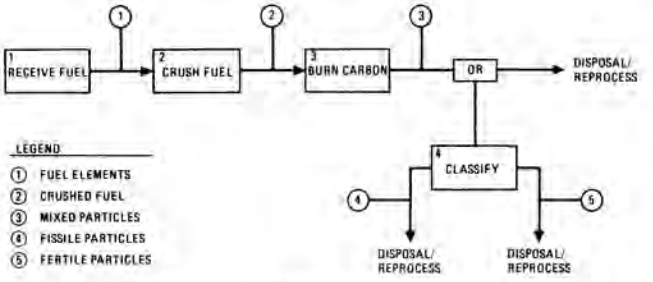

Fig. 2. Volume-reduction flowsheet for HTGR fuel crush-burn reference process.

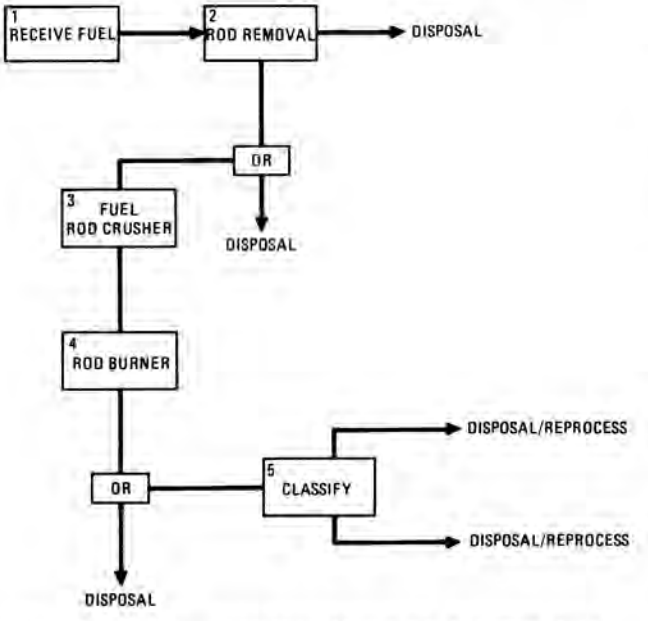

Fig. 3. Volume-reduction flowsheet for HTGR fuel rod removal process.

TABLE II

Volume-Reduction HTGR Fuel Crush-Burn Reference Process Flowsheet Stream Characterization and Throughput[a]

Material Handled	Stream No.	Throughput (kg/yr)	Est. Material Density (g/cm^3)
Fuel Elements	1	6.45×10^6	1.49
Crushed Fuel	2	6.45×10^6	--
Mixed Particles	3	8.1×10^5	--
Fissile Particles	4	2.6×10^5	2.92
Fertile Particles	5	5.5×10^5	3.86

[a]Based on processing ~50,000 HTGR-LEU fuel elements annually (unirradiated fuel).

determining chemical, equipment, facility, and operating requirement for a reprocessing or stabilization plant.

REVIEW OF NUCLEAR WASTE STABILIZATION REQUIREMENTS AND PROCESSES

While the disposal package criteria for HLW have not been completely defined, a stabilization process will probably be required for volume-reduced spent HTGR fuel and possibly for whole spent fuel elements. At a minimum, the stabilization process will (1) provide structural stability, (2) reduce dusting and gassing, (3) reduce void space within the waste and between the waste and its package, and (4) impart groundwater leach resistance to the final waste form.[4] The stabilization process must ultimately contribute to the overall Nuclear Regulatory Commission HLW disposal package requirements described in 10CFR60.

The literature was surveyed for existing processes for nuclear waste stabilization.[5-9] Several processes were identified which have been developed for nuclear wastes. Five processes were initially judged to be potentially applicable as stabilizing methods for HTGR fuel: vitrification, concreting, metal matrix casting, synthetic rock formation, and individual particle coating. The apparent merits and disadvantages of each of these stabilization methods for use with HTGR fuel are discussed below.

Vitrification

In the vitrification process, nuclear waste is reacted with molten glass and solidified. Process temperatures are generally from 800° to 1100°C. The process can accept a liquid or particulate feed, providing the maximum particle size is less than about 50 microns. Comparative evaluation of the vitrification process product with other waste-stabilized forms shows that the material has very low actinide and fission product leach rates.[9] The feed to the vitrification process is generally derived from liquid chemical process materials which have been evaporated and calcined to a finely divided oxide powder.

The vitrification process does not appear to be directly applicable to the treatemnt of reference flowsheet volume-reduced HTGR fuels because of the relatively large particle size of intact fuel particles (nominally 800-μm diameter) and the possible effect of silicon carbide and residual unburned carbon on the process. Carbon not oxidized in the burning operations is expected to react with molten glasses, and these reactions could result in adverse physical and chemical changes in the waste form. Size reduction of the fuel particles to a powder is difficult due to the abrasiveness of silicon carbide hulls and sintered ThO_2 kernels. This process has yet to be attempted.

Concreting

In an evaluation made at the Savannah River Plant, a concreting process, known as FUETAP (Formed Under Elevated Temperature and Pressure) was rated as a close second to a borosilicate glass vitrification process for stabilizing Savannah River Plant HLW.[10] The FUETAP concreting process has some very attractive characteristics which appear to be compatible with reference flowsheet volume-reduced HTGR fuels. In general, the temperature and pressure requirements are modest and the FUETAP process products are concrete

waste forms with high density, low porosity, good mechanical strength, high thermal and radiation stability, and relatively low actinide and fission leachability. The highest temperature required is ~250°C (for removal of unbound water), with required pressures ranging from 0.1 to 1.5 MPa. Silicon carbide and carbon will not react with the cement and can be tolerated. No feed particle size limit is stated, and neutron poisons may be added to control criticality in cement forms. The leachability of actinides and cesium and strontium fission products (typically the most difficult to contain) is generally higher from cement forms than from glasses, but work at Oak Ridge National Laboratory (ORNL) shows that leachability can be lowered by using tailored grouts containing components that increase the retention of these elements (e.g., clays and fly ash).[5]

Concrete has been used extensively for several years to stabilize low-level and intermediate-level radioactive wastes. More than 400 m^3 of intermediate-level radioactive waste has been disposed of in geologic shale formations at ORNL by a grout injection process (hydrofracture).[11] Concrete technology is well developed.

Other Potential HTGR Waste Stabilization Methods

Metal matrix casting,[7] individual particle coating,[8] and Synroc processes are relatively complex methods which would require extensive development prior to use with HTGR fuels.[9] All of these methods require operation at high temperatures (700°C or higher), a decided process operational disadvantage. In addition, these methods have stated limits on feed particle sizes and would probably be more costly than the vitrification or concreting methods. Therefore, these methods were not considered to be readily adaptable to the stabilization of HTGR fuels.

SELECTION AND PRESENTATION OF A REFERENCE STABILIZATION PROCESS FOR HTGR FUEL

Concreting was selected as the most promising of the existing processes for stabilizing volume-reduced HTGR fuels. The concreting process has versatile feed requirements and yields an acceptable solid waste form with good physical and chemical properties for subsequent waste disposal. The encapsulation of whole particles in a concrete host yields a waste form in which the inert silicon carbide particle coatings enhance the retention of nuclear materials in the waste package, even in the unlikely event of a geologic repository groundwater ingress. Use of tailored grouts should result in adequate waste-form fission product and actinide leach rates, even with broken particles. The concreting process is relatively simple, is based on well-established technology, and has a comparatively low cost.

Waste Stabilization Flowsheet for HTGR Reference Process Development

Figure 4 gives the proposed flowsheet developed for the reference stabilization process. In the reference process, mixed fissile and fertile silicon carbide-coated fuel particles are fed to the grout (grout is defined as a fluid concrete mixture that has not yet set) preparation process along with Portland cement, water, fly ash, sand, clay, and water reducer. The fly ash and clay are added to the grout to reduce strontium and cesium fission product leachability rates, respectively. The water reducer minimizes the amount of water required to maintain a pourable grout.

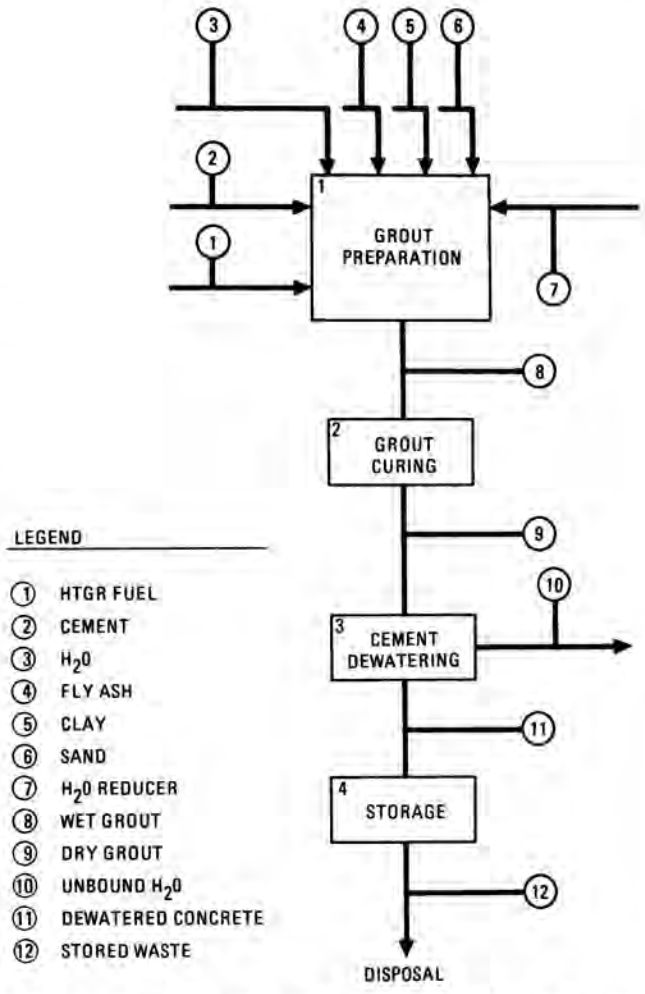

Fig. 4. Waste stabilization flowsheet for HTGR fuel reference process.

After preparation, the grout is cured at elevated temperatures and pressures (nominally 200°C and 0.1 MPa). The cured grout is dewatered at elevated temperature (~250°C) under vacuum. The dewatering step is necessary to remove free (unbound) water and thereby reduce gassing due to radiolysis of water. Following the dewatering step, the stabilized waste may be stored or sent to disposal.

Estimate of Material Throughputs for Reference HTGR Fuel Stabilization Flowsheet

Table III contains a formulation development at ORNL for the preparation of cement-based, simulated Savannah River Plant waste forms.[5] This formulation was the basis for the material characterization and throughput for the reference stabilization process in this work. Figure 5 shows relative component quantities required in the stabilization processes. Annual processing of 50,000 HTGR-LEU fuel elements was assumed.

Data derived in flowsheet development indicate that the ~1000 tonnes of mixed particles produced during annual processing of 50,000 fuel elements through the reference crush-burn, volume-reduction operation could be converted to ~4500 tonnes of stabilized concrete-based waste.

TABLE III

Concrete Mix for Simulated Savannah River Plant Waste[a]

Component	Fraction of Total (wt %)
Type I Portland cement	22.0
Fly ash	11.0
Savannah River Plant simulated waste solids	20.0
Sand	27.75
D-65 water reducer	1.25
Water	18.00

[a]Reference 5.

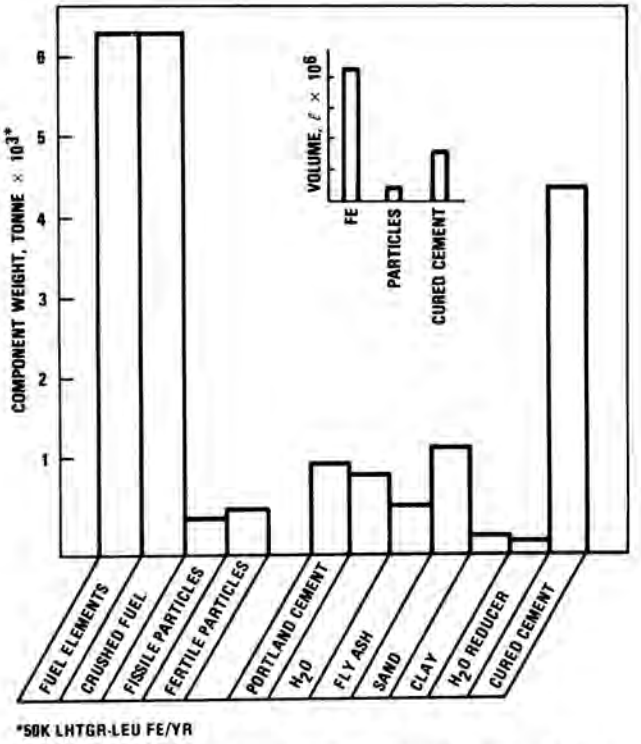

Fig. 5. Relative component quantities for volume reduction/stabilization of spent HTGR fuel.

LABORATORY PREPARATION OF SIMULATED HTGR CONCRETE-STABILIZED WASTE FORMS AND TEST RESULTS

Several small, cylindrical (2.5 cm length and diameter), simulated spent HTGR fuel-concrete host waste forms were prepared in GA laboratory tests of limited scope. In these tests, unirradiated fuel (in the form of rod section containing TRISO-coated ThO_2 particles, burned-back whole ThO_2 TRISCO-coated particles, and crushed unburned and burned ThO_2 particles) was used as feed stock to the concreting process. The percentage of fuel in the prepared waste forms ranged from 5 to 80 wt %. All preparations were cured in the formulation container at ambient temperature and atmospheric pressure. Several of the cured waste forms were dewatered at 250°C.

The physical dimensions of all cement waste forms prepared in this work were measured to enable the volume and density of the final form to be calculated and, in the case of whole particles, the cement host density to be compared to values obtained in earlier work.[5] Comparable results were achieved for the concrete hosts containing HTGR fuel.

Some of the laboratory-prepared HTGR fuel/concrete waste forms were doped with stable oxides of cesium and strontium (to simulate fission product content) and later subjected to material characterization leaching tests. The fission product retentive effects of fly ash and clay in the concrete are shown graphically in Figs. 6 and 7.

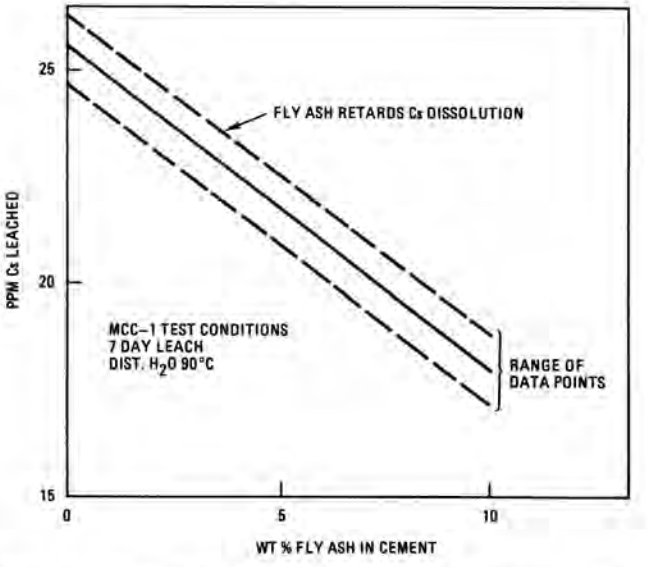

Fig. 6. Effect of fly ash on HTGR fuel/cement waste form cesium leachability.

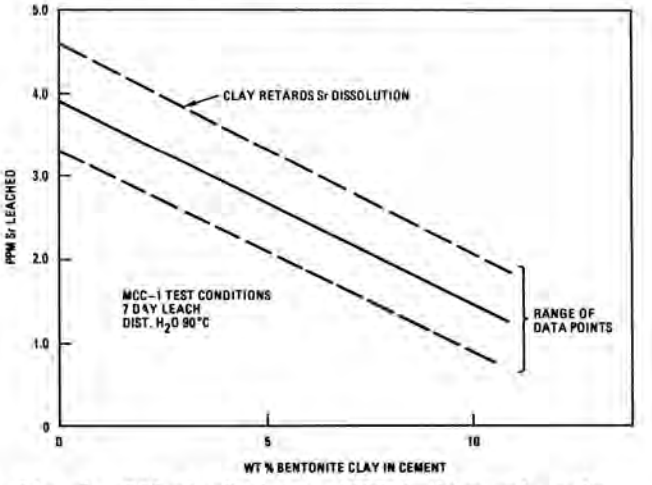

Fig. 7. Effect of clay on HTGR fuel/cement waste form strontium leachability.

CONCLUSIONS AND FUTURE WORK RECOMMENDATIONS

1. Preliminary test results show that concrete stabilized waste forms with acceptable physical properties can be prepared from whole particle, crushed, or crushed-burned volume-reduced HTGR fuel.

2. Relatively high-waste loadings can be achieved in a concrete waste host. Experimental test results indicate that up to ~80% by weight of HTGR fuel may be encapsulated in a concrete waste host and that whole or crushed fuel particles can be substituted for all or part of the concrete sand component.

3. Significant volume-reduction factors can be maintained when concrete is used as the host material for HTGR fuel.

4. Companion studies are required to assess the effects of decay heat, radiation, and criticality limitations on the reference stabilization process and products.

ACKNOWLEDGMENT

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