Extended Summary

A NEW REPOSITORY WASTE FORM: GRAPHITE–CARBON HIGH–LEVEL WASTE

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A new repository waste form is proposed for the geologic disposal of high-level waste (HLW): a graphite-carbon high-level waste (GC HLW). Fission products from the processing of coated-particle graphite-matrix spent nuclear fuel (SNF) from high-temperature gas-cooled reactors (HTGRs) would be solidified using the carbon in various forms from the SNF assembly. Because the wastes generated in processing HTGR fuel are very different from those associated with traditional light-water-reactor SNF, it is appropriate to consider a new waste form based on the characteristics of the wastes generated during processing.

The oxide fuel (Table 1, Fig. 1) is in microspheres coated with several layers of different types of carbon and a layer of silicon carbide. The coated particles are incorporated into carbon-matrix cylindrical fuel compacts, which are then embedded in holes in a hexagonal graphite fuel block. The HTGR is being investigated for the destruction of minor actinides. For this application, the SNF contains driver fuel (DF) and transmutation fuel (TF). The DF is processed for recovery of fissile materials for recycle whereas the TF compacts are either directly disposed of or further irradiated in an accelerator.

Traditional approaches to processing graphite-matrix SNF are expensive because the graphite contains significant quantities of radioactive ^{14}C. The traditional first step in processing this SNF has been to burn the graphite to yield carbon dioxide (CO_2) and scrub the off-gas with a calcium hydroxide solution—thus producing a solid calcium carbonate waste form that contains the ^{14}C. A typical HTGR SNF assembly is 74 wt % graphite; thus, the largest processing operations are associated with burning the graphite and recovery of the CO_2.

A new processing approach is proposed that yields a new HLW form (Fig. 1). The fuel compacts are mechanically removed from the graphite block. The block, with its ^{14}C, becomes a low-heat waste form for direct disposal or consolidation and then disposal in a repository. The compacts are crushed and leached in nitric acid, which dissolves uranium, plutonium, higher actinides, and many fission products. The resulting solution is then sent to a processing plant for recovery of the fissile and fertile materials. The leaching process leaves a dissolver residue of graphite particles, silicon carbide hulls, and noble-metal fission products. The separations facility, which uses traditional aqueous separations processes, produces an aqueous HLW stream consisting of nitric acid, the soluble fission products, and trace quantities of actinides. The dissolver residue and the aqueous HLW stream are then mixed. Reagents are added to denitrate the slurry, with the subsequent precipitation of many fission products onto the dissolver residues. (Alternatively, the aqueous HLW stream may be thermally denitrated, with the resultant oxides mixed with the dissolver residue). The aqueous slurry is fed to a heated extruder, along with an organic binding agent. The heated extruder evaporates the water and mixes the organic binding agent with the solids. The solid product is heated at temperatures from 800 to 1000°C, a process that destroys the organic binder and produces the final GC-HLW. Each step in this process has already been used in one or more industrial applications. However, some of these industrial operations have not involved highly radioactive materials or the specific chemical compositions associated with this new process.

Table 1. Material balance for materials in one graphite-matrix fuel element

	Mass (kg)				
		Transmutation Fuel		Driver Fuel	
Component	**Fuel element**	**Compacts**	**Particles**	**Compacts**	**Particles**
Graphite	90.00	0	0	0	0
Filler carbon	13.97	3.14	0	10.83	0
Pyro carbon	4.58	0.72	0.72	3.86	3.86
Porous carbon	1.96	0.26	0.26	1.70	1.70
SiC	3.58	0.56	0.56	3.02	3.02
TF (TRU)	0.20	0.20	0.20	0	0
DF (Fuel + FPs)	0.80 + 1.88	0	0	0.80 + 1.88	0.80 + 1.88
TOTAL	116.97[b]	4.88	1.74	22.09	11.26

[a]Assumes a DF:TF ratio of compacts of 4:1.
[b]Without the graphite block, the total mass of carbon in compacts is 20.51 kg.

Thermodynamic analysis indicates that most of the radionuclides will be in the same chemical form as found in HLW glass. Cesium, strontium, and the rare earths will remain as oxides. However, some elements, such as iron, will be reduced to the metal state—rather than oxides, as is found in HLW glass. Selected radionuclides will embed themselves within the planer structure of the graphite and will be extremely resistant to being leached from the waste form under almost all potential repository environments. There is the potential that cesium compounds could intercalate between the planes of the graphite. Other radionuclides, such as the noble metals, will be embedded in the GC HLW. The waste loading is determined by the materials in the fuel compact. If higher waste loadings are viable, it may be feasible to add wastes from other processes. The matrix will consist of graphite, the carbon from the SNF compacts, and the degradation products of the organic binder.

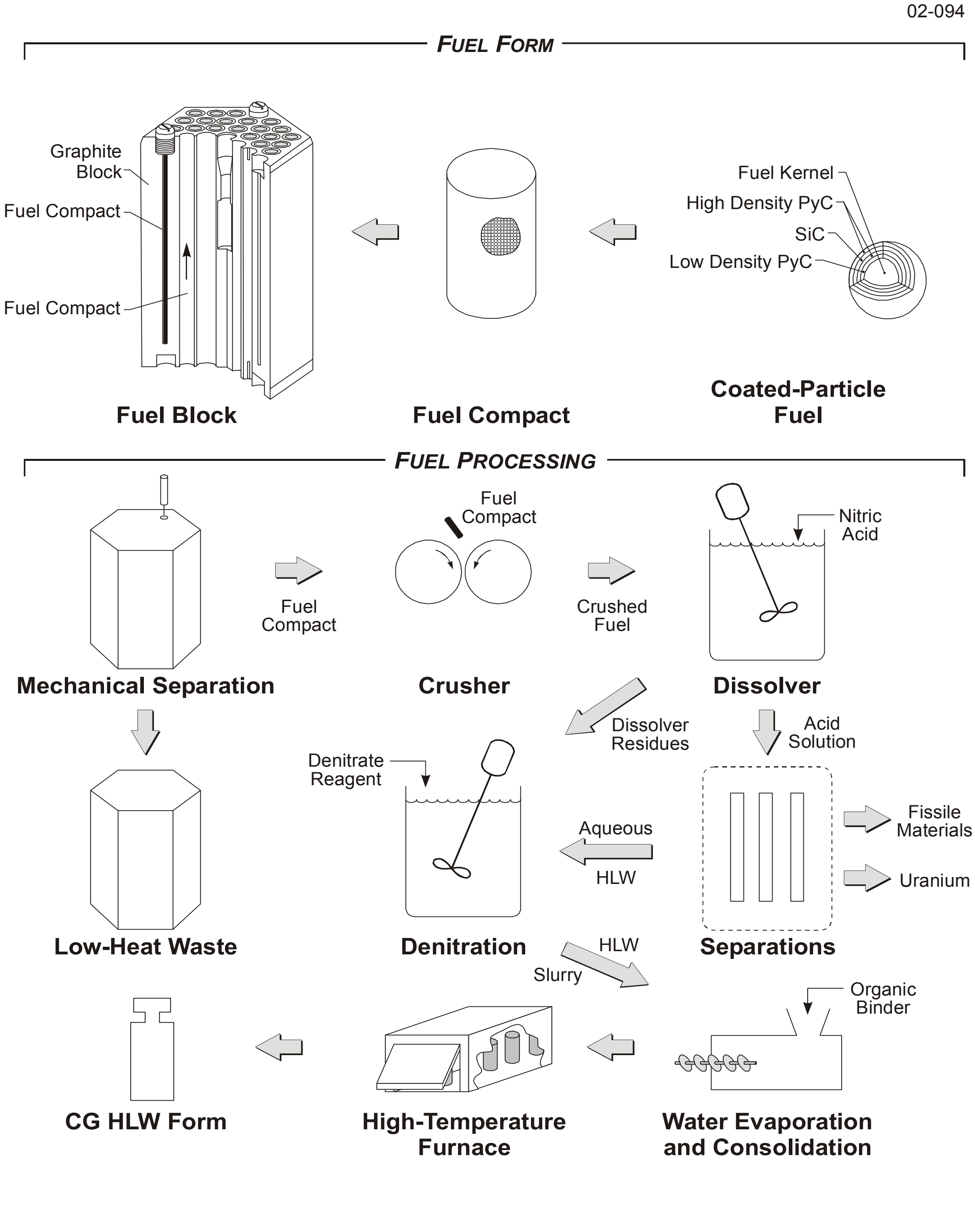

Fig. 1. Production of the graphite-carbon high-level waste form.

There is large scale industrial experience with the addition of binders to graphite and the subsequent bake-out to convert the organic binders to various forms of carbon. This method is used to produce impervious graphite, which is used for the fabrication of many graphite products. In France, this option has been investigated as a waste treatment process for irradiated graphite moderator blocks from gas-cooled reactors—to seal radionuclides into the graphite and reduce long-term leachability.

The choice of organic and temperature determines the specific morphology of the carbon produced from the pyrolysis of the organic. For this application, bake-out temperatures will be limited by the desire to avoid vaporization of cesium compounds from the graphite waste form. The option exists to place the waste form in a graphite cylinder before bake-out to produce a waste form with a relatively clean exterior.

A GC HLW has the potential to be an outstanding waste form. Coal and natural graphite have remained stable with various embedded impurities under a variety of geological conditions for hundreds of millions of years. Analysis of direct disposal of graphite-matrix SNF shows excellent performance under Yucca Mountain (YM) conditions. While carbon forms are not thermodynamically stable under oxidizing geochemical environments, measured oxidation kinetics at expected repository conditions imply lifetimes of hundreds of millions of years. The YM project at one time considered a graphite waste package (WP). Although this type of WP had outstanding geochemical durability, it was ultimately rejected because of the brittle characteristics of graphite that made it unsuitable as a package material to keep groundwater out. As a waste form, it is not required to be a single monolithic solid.

The GC HLW thermal performance is potentially better than traditional HLW glass. HLW glass typically has a maximum storage temperature of 400EC to avoid devitrification. A carbon waste form would be expected to maintain its integrity above 600EC. The thermal conductivity of the proposed GC HLW form is not known; however, the thermal conductivity of the components of the GC HLW are higher than glass. The thermal conductivity of SiC is 20 times that of glass and the thermal conductivity of graphite is 50 times that of glass. Combined, these factors should allow much higher heat loads than traditional HLW forms. This allows for larger HLW log sizes, reducing the number of HLW logs, and reducing the associated handling costs in storage, transport, and disposal.

Because GC HLW is a new HLW form, major uncertainties exist. However, there is much supporting information. Significant work has been conducted on how to prevent the release of radionuclides from irradiated reactor graphite moderator blocks. Graphite and many other forms of carbon are known to have excellent characteristics over long geological periods of time. There is geological, laboratory, and engineering data on inclusion of various materials in various carbon forms. Many materials have been added to various carbon products to modify their characteristics. Research is under way to further investigate the characteristics of this new GC HLW form.

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