

An Assessment of Applying Pyroprocessing Technology to Advanced Pebble-Type Fuels

Chemical and Fuel Cycle Technologies Division

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CONTENTS

1.	INTRODUCTION	1
2.	ISSUES TO BE ADDRESSED FOR APPLYING PYROPROCESSING TO PEBBLE-TYPE FUELS	4
3.	HEAD END PROCESSING METHODS	6
	3.1 Head End Processes Most Likely to be Suitable	6
	3.2 Head End Processes Potentially Suitable.....	7
4.	RECOMMENDED FEASIBILITY TESTS.....	9
	4.1 Laboratory Scale Pyroprocessing Unit Operations	9
	4.2 Acoustic Fracture of TRISO Fuel Pyrolytic Carbon	10
	4.3 Pyrochemical Breaching of TRISO Fuel Pyrolytic Carbon	11
5.	CONCLUSIONS AND RECOMMENDATIONS	12
	REFERENCES	13

FIGURES

1.	(a) Schematic drawing of TRISO particle and spherical fuel form and (b) photograph of fractured TRISO particle	1
2.	TRISO Fuel Assemblies (Source: US Department of Energy).....	2
3.	(a) X-ray cross section and (b) SEM cross section of TRISO particle after irradiation.....	2
4.	Generic Flowsheet for Application of Pyroprocessing to Pebble-Type Fuels.....	4

TABLES

1.	Survey of Advanced Reactors Being Developed to Use Pebble-Type Fuels	3
2.	List of Head End Processes being Assessed	6
3.	Test Matrix to Measure Effects of Graphite and SiC	10

1. INTRODUCTION

With an expected increased demand for high-assay low enriched uranium (HALEU) to supply advanced reactors, the possibility of recovering actinides from various used nuclear fuels (UNF) is being re-assessed. Pebble-type fuels such as tristructural isotropic (TRISO) fuel are intended to be directly disposed after use, but the HALEU remaining in each pebble is a resource with increasing value. An economically feasible recovery strategy would maximize the amount of HALEU recovered for reuse in new fuel without increasing the waste volume relative to the direct disposal of used TRISO fuel. We are evaluating the technical feasibility of recycling TRISO or pebble fuels to recover actinides for reuse as new fuel for advanced reactors by using pyrochemical methods.

Pebble-type fuels were developed for use in high temperature gas cooled reactors to securely retain fission products within the fuel particle. The small fuel kernel, which is usually composed of HALEU in the form of UO_2 , UCO , UC or UN , is encapsulated by several layers of pyrolytic graphite and silicon carbide, as pictured in Figure 1. The layered construction of pebble-type fuels effectively captures fission products during use, but makes subsequent recovery of actinides from the kernel difficult. The coated kernels, which are referred to as fuel particles, can either be used directly or a large number of particles can be embedded in a graphite matrix to create fuel forms including billiard ball-sized spheres (pebbles), pins, or rods referred to as compacts, pictured in Figure 2.

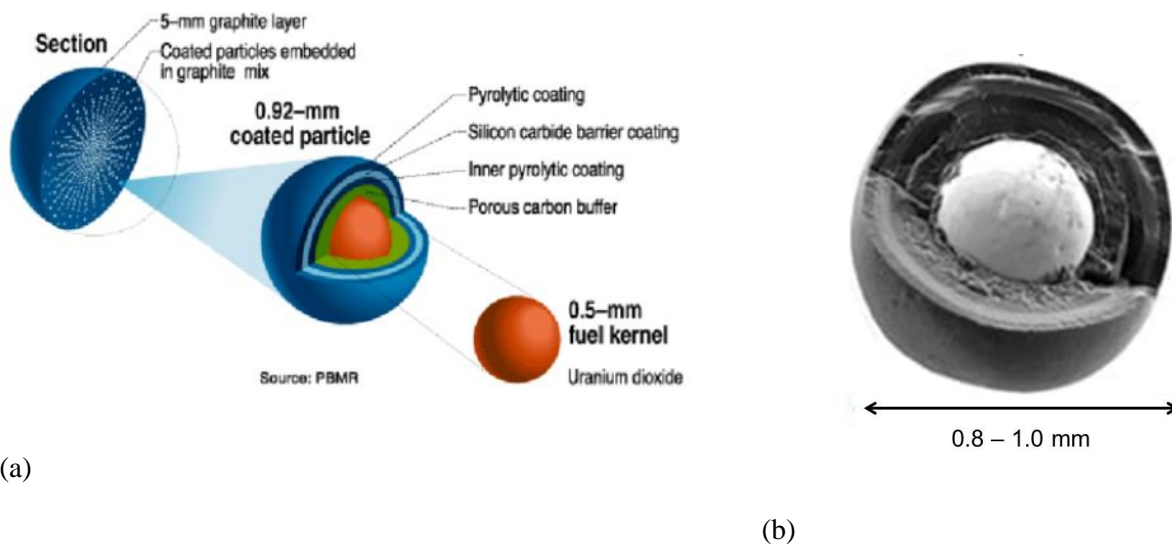


Figure 1. (a) Schematic drawing of TRISO particle and spherical fuel form [1] and (b) photograph of fractured TRISO particle [4].

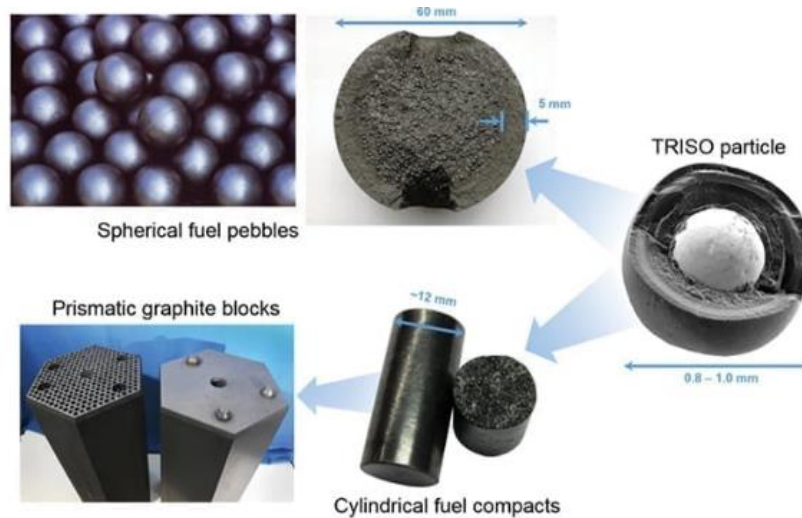


Figure 2. TRISO Fuel Assemblies [4]

The fuel particle is designed such that fission products generated during reactor operation can migrate into the porous graphite adjacent to the kernel and the surrounding layers, but are retained within the particle by the SiC layer. Figure 3 shows micrographs of an irradiated fuel particle imaged by X-ray tomography and a polished cross section analyzed by scanning electron microscope (SEM).

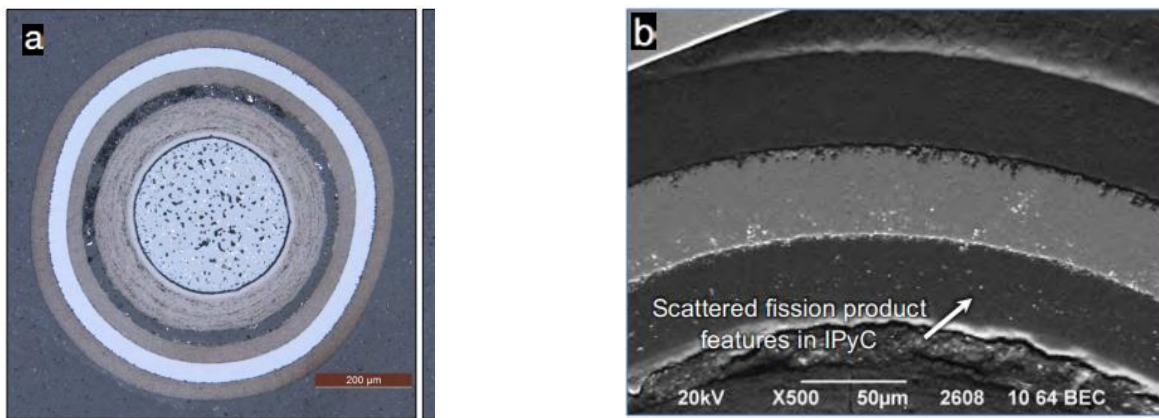


Figure 3. (a) X-ray tomography cross section and (b) SEM photomicrograph of metallurgical cross section of TRISO particle after irradiation (Figs. 32a and 33 from [2]).

Pyroprocessing uses electrochemical reactions to extract fissile material from used fuels and generate source material suitable for recycle. The processes were originally developed to treat used metallic fuels and have since been applied to traditional LWR oxide fuel by including a preceding electroreduction operation. Pyroprocessing flowsheets typically begin with a head end treatment tailored to the fuel type by either chopping clad metallic fuel or electroreducing oxide fuel to metal. The main separation step in a pyroprocessing flowsheet is electrorefining, wherein constituents in the used nuclear fuel are oxidized and dissolved into molten salt and then actinides are selectively deposited as alloys that can be used as source materials for new fuel. The majority of the fuel is recovered as separate alloys of by-product uranium and

mixed actinides. Waste from the process consists of process salt and metallic waste streams with fission products and fuel cladding. Both the volume of HLW and its activity are decreased by pyroprocessing compared to direct disposal.

An assessment of the suitability of pyroprocessing methods to pebble-type fuels is being performed to identify technological gaps that must be addressed. This report summarizes the head end processing necessary to prepare fuel for pyroprocessing. Addressing these issues is deemed essential to justify further consideration of using pyroprocessing methods to recover HALEU from pebble-type fuels.

Other issues that remain to be addressed include regulations for disposal of by-product wastes that may contain amounts of ^{14}C and other radionuclides resulting in the waste being classified as Class C or structural wastes. Requirements for handling and disposal of coatings removed from the fuel kernel before or during processing will depend on the amounts and distributions of fission products in the coating materials. Pebble-type fuels are designed to retain all fission products within the protective coatings, including gaseous species, and are generally considered to be indestructible during use. However, post-irradiation examinations (PIE) indicate breaches of the coatings do occur leading to release of radionuclides. Results of PIE of TRISO particles performed previously provide insights into the levels and distributions that can occur within a particle. However, no information is available on the total radionuclide contents in coatings that have been removed from fuel particles, which will be an important aspect of HALEU recovery. Those issues are being addressed in on-going assessments.

A survey of advanced-pebble or TRISO fueled reactors was done to determine the prevalence of different fuel forms in planned designs. Attributes of reactor designs collected include the reactor developer, reactor name, fuel type, fuel kernel material, and coolant type. This survey is not comprehensive but demonstrates the variability in fuel type and kernel chemistry being employed. Table 1 summarizes this survey.

Table 1: Survey of Advanced Reactors Being Developed to Use Pebble-Type Fuels [14-19]

Company	Reactor Name	Fuel Type	Fuel Kernel	Coolant Type
HTR-PM	HTR-10	Pebble	Unknown	helium
PBMR Ltd.	PMBR	Pebble	UO ₂	helium
BWXT	BANR	TRISO Pebbles in cylindrical compacts	UCO or UN	helium
Xenergy	Xe-100	TRISO Pebbles	UCO	helium
KAIROS	Hermes Reactor	TRISO Pebbles	UCO	Salt (FLiBe)
General Atomics	EM ²	Pebble	UC	helium
Westinghouse	Evinci	TRISO	UCO	air
Adams Atomic Engines'	Adams Engine	TRISO in a fixed annular bed	UCO	nitrogen
Star Core Nuclear	StarCore HTR	TRISO in prismatic carbon matrix	UCO	helium
Ultra Safe Nuclear Corp	USNC MMR	TRISO in prismatic carbon matrix	UCO or UN	helium
Holosgen	Holos Quad HTR	TRISO in hexagonal compacts	UCO	air

2. ISSUES TO BE ADDRESSED FOR APPLYING PYROPROCESSING TO PEBBLE-TYPE FUELS

In pebble-type or TRISO fuels the fuel kernel containing the fissile material is encapsulated within several carbon and silicon carbide layers that mitigate loss of fission products and fissile material during reactor operation. In addition, a large number of coated particles are often embedded in a matrix of graphite to create spherical or cylindrical fuel elements that can be stacked into beds or assemblies. The coating that microencapsulates each kernel must be breached so that, during pyroprocessing, molten salt electrolyte can penetrate that coating and make contact with the fuel kernel. A head end process that first separates the particles from the matrix material and then breaches the coatings is required. Figure 4 is a generic flowsheet for pyroprocessing pebble-type fuels.

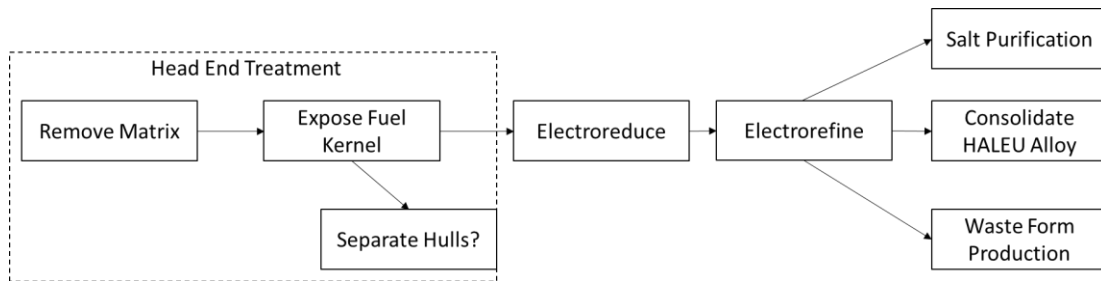


Figure 4. Generic Flowsheet for Application of Pyroprocessing to Pebble-Type Fuels

An optional step is shown in the schematic for separation of the graphite and SiC hulls from the fuel kernels during head end treatment. The effects of graphite and SiC residue on the efficiency of pyroprocessing and the product quality are not known. It remains to be determined the extents to which pieces of graphite and SiC layers and fines in the electrorefiner will detrimentally affect either electroreduction of the fuel kernel, subsequent process operations, or product quality before the requirements for a head-end process can be identified. Previous experience using graphite anodes to electroreduce fuel showed carbon fines suspended in the molten salt created electrical bridges between the electrodes that decreased the process efficiency [3]. Large amounts of fines may similarly inhibit the deposition of actinides during electrorefining. The effects of SiC fines on electrochemical operations are expected to be similar to the effects of graphite. Although it will be necessary to remove the matrix material surrounding the particles in the fuel form prior to processing, it may not be necessary to completely remove the coatings from the fuel. Breaching the coating material to expose the kernel may be sufficient for processing, in which case the coating material would be recovered after electrorefining the kernels.

Fuel kernels are commonly made of UO_2 , UC, a mixture of UO_2 and UC (referred to as UCO), or UN. The conversion of UO_2 to U metal for subsequent electrorefining is an established process, but processes for recovering uranium from UCO, UC, and UN need to be demonstrated [3]. Electrochemical reduction of UO_2 to metallic uranium occurs as a solid state reaction that proceeds from the surface inward and results in densification. Porosity in the generated metal will provide access to the interior of the kernel. Oxygen will be generated simultaneously on the anode during reduction of UO_2 and probably also during the reduction of UCO to UC. The retention of UN and UC during electroreduction will not affect further

processing because both are electrically conductive and will dissolve during electrorefining. If graphite and SiC are stable in the electrorefiner salt and do not impede electroreduction or electrorefining of the fuel kernel, the coating materials could be separated from the small dense metallic kernels after processing.

Head end processing to breach or remove the pyrolytic graphite and SiC coatings in preparation for pyroprocessing may result in a release of small amounts of volatile fission products. An off-gas capture system will likely need to be included with any head-end process that breaches the coating to capture volatile fission products. Analyses of irradiated TRISO fuel particles by using SEM/EDS detected fission products, Cs, I, Ba, and Xe in the SiC layer and fission products Pd, Cd, and Ag and a small amount of uranium in cracks formed between the fuel kernel and inner-most coating layer [12,13]. Analyses of both the separated fuel kernels and hulls from irradiated pebble-type fuels are needed to quantify the fission product and actinide contents in the SiC and graphite coatings and estimate the amounts volatilized. For example, it needs to be determined if the small amount of actinides known to collect between the fuel kernel and the first inner coating remain with the fuel kernel, are retained in the coatings, or are released during head end processing.

Significant amounts of ^{14}C are likely to be present in the graphite and SiC because carbon is activated in the reactor. The graphite and SiC coatings directly adjacent to the fuel kernel will likely contain more ^{14}C than graphite matrix materials further from the fuel kernel. If the coating is removed as a separate waste stream, the total amounts of ^{14}C and other fission products will impact the waste classification governing disposal of those materials. It will be advantageous to generate a technical basis for classifying the recovered matrix material as contaminated structural material to limit the volume of HLW requiring geologic disposal to only include the pebble hulls and processed U-depleted fuel kernels.

The key issues for applying pyroprocessing to pebble-type or TRISO fuels are determining the effect of graphite and SiC hulls on pyroprocessing unit operations, and the availability of a head end process that exposes the fuel kernel to salt without generating unacceptable amounts of carbon fines. The tolerance of pyroprocess operations to residual carbon will dictate whether the coating materials must be removed from the kernel during head end operations or only be breached. If the coating materials must be removed prior to pyroprocessing, the cost of separating coating materials will be an important factor in determining the benefits of reprocessing. If the kernels can be processed in the presence of breached hulls, the hulls and U-depleted kernels can be immobilized together in an engineered waste form.

3. HEAD END PROCESSING METHODS

An initial survey of methods for removing the carbon and SiC layers from the fuel kernel that have been described in the literature was performed to assess suitability of those methods to preparing pebble type fuels for pyroprocessing. Considerations included the compatibility of the product with pyroprocessing, the extent to which the process generates carbon fines, the waste volume generated relative to direct disposal, and the complexity of the process. The results of the survey are indicated in Table 2 with methods divided into three categories: (1) those most likely to be suitable for use in a pyroprocessing flowsheet, (2) those that are potentially suitable for use, and (3) those that are least likely to be suitable with pyroprocessing. Technological gaps for applying each method to pebble-type fuels and subsequent pyroprocessing were identified. Summary assessments for methods that are most likely or potentially suitable for use are provided in Sections 3.1 and 3.2. Processes determined least likely to be suitable with pyroprocessing are not discussed further.

Table 2. List of Head End Processes being Assessed

Most Likely to be Suitable	Potentially Suitable	Least Likely to be Suitable
Mechanical Separations [5, 7, 8]	Thermal Shock [4, 8, 10]	Electrolytic Dissolution [8, 10]
Acoustic Fracture [4]	Pyrometallurgical Dissolution in non-halide salts [4, 11]	Chemical Dissolution or Disintegration [4, 5, 6, 10]
Pyrometallurgical Dissolution in halide Salt [4, 9, 10]	Combustion [7, 10]	Gas Halogenation [4, 5, 10]

3.1 Head End Processes Most Likely to be Suitable

Three processes were deemed most likely to be suitable for preparing TRISO fuel for pyroprocessing. Those methods are simple, directly compatible with the molten salt electrolyte used in pyroprocessing and decrease the amount of HLW relative to direct disposal of the used fuel. The three methods are discussed in detail below and the technological gaps in their application to this fuel are identified.

Mechanical Separations

Physical or mechanical separation methods crush or crack the carbon and SiC coatings to expose the fuel kernel [4, 5, 7, 8, 11]. The fuel kernels can be separated from the fractured coatings pieces by sifting the smaller fuel kernels from the hulls and sweeping carbon fines from the sifted fuel kernels. The sifting and sweeping operations do not provide complete separation and result in a product stream of fuel kernels with a small amount of retained carbon fines and a waste stream of SiC and graphite hulls.

The process as currently developed includes multiple separation stages using off-the-shelf equipment. The use of off-the-shelf equipment lowers design and operating cost. Consolidating separation stages or simplifying the separation stages should be considered to further decrease operating costs. Because the presence of retained carbon fines may be detrimental to pyroprocessing operations, alternative physical methods of exposing the fuel kernel that minimize generation of carbon fines should be considered, such as shearing or laser ablation.

Acoustic Fracture

Another head end process likely to be suitable is acoustic fracture. In this process, fuel is submerged in a salt and ultrasonic sound waves are applied to fracture the coatings [4]. The product stream consists of fuel kernels with cracked coatings. These fuel kernels would then be pyroprocessed to produce a single waste stream of hulls and densified U-depleted kernels. The suitability of this process depends on several factors. First, that the presence of hulls in the molten salt does not adversely impact pyroprocessing unit operations of electroreduction and electrorefining. Second, that the fracturing does not produce an unacceptable amount of carbon fines. Third, that the degree of fracture provides adequate access for molten salt to contact the fuel kernel. A major uncertainty for applying this technology to pebble-type fuels is that it has never been demonstrated in molten salts, only in weak nitric acid solutions.

Pyrometallurgical Dissolution

A pyrometallurgical approach using halide salts consists of submerging coated fuel particles in a molten salt containing MgCl_2 and then applying a potential between the fuel and an anode to generate Mg metal [4, 9, 10]. The Mg metal reacts with SiC to form Mg_2Si and C. The graphite-coated fuel kernels are then removed from the salt containing MgCl_2 , drained, and then immersed in a salt containing LiCl. A potential is applied between the fuel and an anode and Li metal is formed. This Li metal intercalates the inner graphite coatings, causing them to slough off of the fuel kernel as fine carbon particles. This results in a product stream of kernels completely separated from their hulls and two waste streams of carbon-contaminated Mg_2Si and Li-containing salts.

Separation efficiency of this process is excellent, but several steps are required and a large amount of carbon contaminated salt waste is produced [4,9,10]. The fuel must be treated in two salts because MgCl_2 will react with metallic lithium formed during graphite intercalation and inhibit the Li from interacting with the graphite. One technical gap is whether salts contaminated with carbon, Mg_2Si , and Li can be cleaned and recycled without generating a large waste stream. If contaminated salt must be directly disposed of, then this head-end process would not reduce waste relative to direct disposal of pebble type fuels. A second technical gap is a means of determining the amount of Mg needed to react with all of the SiC and leave no available Mg for reaction with Li metal in the subsequent step. If this can be achieved, a single salt can be used to decrease the waste volume and the required number of steps.

3.2 Head End Processes Potentially Suitable

The three head end processes determined to be potentially suitable are compatible with the molten salt electrolyte used in pyroprocessing, but may produce unacceptably large waste streams, require subsequent processing steps, and may result in fission product loss. The three methods are discussed here briefly and the technological gaps in their application to this fuel are identified.

Thermal Shock

Thermal shock at 1350–1650 °C can be used to fracture the graphite and SiC layers of the pebble fuel [4, 8, 10]. This would likely produce a significant amount of carbon fines and a mixed product of hulls and fuel kernels. Sifting and sweeping operations can be applied to decrease the quantity of carbon fines and separate hulls from fuel kernels, but these are additional processing steps. Volatile actinides or fission

products may be lost from the fuel kernel at these elevated temperatures. This process was deemed to be potentially suitable because it does not increase waste volume or introduce materials that are incompatible with molten salts, but it may require a follow-on process to separate the fuel kernels from the hulls and has a risk of actinide or fission product loss due to vaporization.

Pyrometallurgical Dissolution

A pyrometallurgical method for removing graphite and SiC is to electrochemically oxidize the graphite to generate carbon dioxide and the SiC to generate silicate in a nitrate or hydroxide salt [4, 11]. The separation of fuel kernels from the coating materials is efficient, but the waste volume is dramatically increased. Waste streams include carbon dioxide, which must be sequestered, and a nitrate or hydroxide salt contaminated with silicate. The separated fuel kernels may have adhered nitrate or hydroxide salt, which is incompatible with pyroprocessing salts. Therefore, thorough removal of these salts from the fuel kernels would be required prior to pyroprocessing.

Combustion

The graphite and SiC can be removed through combustion by heating the fuel under oxygen gas to generate CO_x gas [7, 10]. A pre-treatment step of crushing the fuel to a powder may be required for efficient combustion. The crushing process can result in breakage of fuel kernels and potentially loss of FPs and fissile material. The generated CO_x gas must be sequestered, which results in a large increase in waste volume. The silicon released from the SiC may remain with the fuel as a contaminant. It is unclear what effect this retained silicon would have on pyroprocessing.

4. RECOMMENDED FEASIBILITY TESTS

Technological gaps that must be addressed to assess application of pyroprocessing to pebble fuels include uncertainty in the effect of separated coating materials on electrochemical operations. The quantity of graphite and silicon carbide layers that can be tolerated in the feed stream is unknown. The effect of graphite and SiC hulls and fines on pyroprocessing unit operations must be determined to establish head end requirements. Once the head end requirements have been established through study of the effect of retained coating materials on unit operations, then head end processes must be developed which meet the established requirements. Pyrochemical processes for recovering uranium from UCO, UC and UN kernels have not been demonstrated, though pyroprocessing is expected to be effective as they are both electrically conductive. Specific technical gaps remain in the application of head end processes as well. Acoustic fracture has never been demonstrated in molten salts and it needs to be determined if pyrochemical dissolution can be conducted in a single salt instead of two to reduce the volume of the waste stream. Feasibility tests are recommended here to address these identified technical gaps.

4.1 Laboratory-Scale Pyroprocessing Unit Operations

Removal or fracture of pebble fuel coating materials will likely produce graphite fines and particulates. The effects of retained graphite, silicon carbide hulls, and carbon fines on the pyroprocessing unit operations of electroreduction and electrorefining, as well as salt removal and metal consolidation steps, remain to be determined. Insights can be gained through simple experiments measuring the effects of controlled amounts of graphite particles and fines on lab-scale pyroprocessing operations. If carbon fines have an appreciable effect, head end processes must be designed to limit the amount of carbon fines produced or include steps to efficiently remove carbon fines prior to processing.

To determine the effect of graphite and silicon carbide materials on the molten salt electrolyte and on pyroprocessing operations, both powders and discrete pieces of graphite and SiC should be added to fuel materials and electroreduction of oxide containing fuels (UO_2 and UCO) and electrorefining of conductive fuels (U, UC and UN) with these contaminants should be performed. The current and voltage requirements for each unit operation should be determined to assess process efficiency and be compared to process efficiencies for those materials in the absence of graphite. Open circuit potential measurements and cyclic voltammetry should be used to monitor changes in the salt conditions during each unit operation. Table 3 summarizes suitable amounts of the different feed materials to be tested and the relative amounts of coating material contaminants to determine the effects. Tests without contaminant materials are included to demonstrate the capacity of pyroprocesses to reduce or refine fuel materials unique to pebble type fuels, UCO, UC and UN.

The fuel baskets and salt crucible ingots should be examined after the tests. Examination of the residual material in the basket should include SEM/EDS imaging of residual fuel pieces. The salt hold up and amount of material retained in the basket should be estimated from the weight difference between pre- and post-test and from SEM/EDS imaging. Examination of the salt ingot from the crucible should include sectioning, photographing and conducting SEM/EDS at a variety of locations across the sectioned ingot to provide a mapping of the location of any accumulated graphite or SiC.

Table 3. Test Matrix to Measure the Effects of Graphite and SiC

Unit Operation	Salt	Feed Material	Contaminant	Relative Amount of contaminant (% of total feed mass)
Electrorefining	LiCl-KCl	U metal	Graphite fines	1, 5, 10, 20
			Graphite pieces	10, 20, 50
			SiC pieces	10, 20, 50
		UC	None	n/a
		UN	None	n/a
Electroreduction	LiCl + 1wt% LiO ₂	UO ₂	Graphite fines	1, 5, 10, 20
			Graphite pieces	10, 20, 50
			SiC pieces	10, 20, 50
		UOC	None	n/a

4.2 Acoustic Fracture of TRISO Fuel Pyrolytic Carbon

Breaching of TRISO particles by acoustic fracture has been performed in acidic media [20] but has not been attempted in molten salts. High temperature ultrasonication equipment utilizing molten salts has been designed and used for Al-Sc alloy preparation, but an off the shelf system is not readily available [21].

It is recommended to build and test a high temperature ultrasonication system with molten LiCl-KCl and to test the breakup of pyrolytic carbon (PyC) and silicon carbide (SC) using this system to demonstrate feasibility. The design would be largely based on Liu et al. [21], using a tube furnace with an ultrasonic probe coupled to the bottom of a crucible within the hot zone of the furnace. Once the system is verified to operate with molten LiCl-KCl at 500 °C and a safe maximum applied ultrasonic power determined, four sets of experiments should be conducted.

The first two experiments would test the breakup of PyC in LiCl-KCl with applied ultrasonication. In the first, a small piece of PyC should be examined by SEM and serve as the baseline reference for future characterization. This PyC should then be added to the salt and the maximum ultrasonic power applied to the system. The salts with the PyC should be cooled, the PyC retrieved, and post-ultrasonic characterization in the SEM should be performed to assess damage to the material. The second experiment should repeat this characterization and ultrasonication process with PyC powder.

The second set of experiments should similarly test the breakup of SiC in LiCl-KCl with applied ultrasonication. In the first, a piece of SiC should be characterized by SEM, exposed to ultrasonication in the LiCl-KCl, and then characterized post-ultrasonication by SEM. Next, the test should be performed with SiC powder.

If no breakup of the material occurs, this system can alternately be used to assist in the electrolytic breakup of TRISO materials. The crucible or a basket containing the PyC or SC material and immersed in the salts can be anodized by a power supply, with a cathode inserted into the molten salt electrolyte from above. This would allow for the electrolytic deconsolidation of the PyC by intercalation of the anions, which has

been proposed as the mechanism of fracture in aqueous media [22, 23]. The breaching of PyC and SiC by electrochemically formed lithium metal is proposed in section 4.3.

4.3 Pyrochemical Breaching of TRISO Fuel Pyrolytic Carbon

Preparing TRISO-type fuels for pyroprocessing by breaching the pyrolytic carbon layers using a molten salt electrochemical technique is highly compatible with the current pyroprocessing flowsheet. This technique was reported by researchers in Korea [24] and involved electrochemically forming lithium metal from a molten salt onto the carbon surface, which intercalates and destroys the graphite.

Tests are recommended to determine the effects of electrochemically generated lithium metal on glassy carbon and reaction-bonded silicon carbide and demonstrate a method for breaching the pyrolytic carbon and silicon carbide layers surrounding the fuel kernel in TRISO fuel.

A total of six experiments are recommended using the Molten Salt Furnace-IV in an argon atmosphere glovebox at INL. All six experiments would be performed at 500 °C in LiCl-KCl eutectic salt with a small amount of additional LiCl added to the mixture. This additional LiCl is to compensate for the LiCl that is reduced to Li metal during the experiments and to minimize deviation from the LiCl-KCl eutectic point.

Three of the experiments should be performed with a glassy carbon rod cathode shrouded by a MgO tube to contain Li metal and carbon fines. Migration of Li metal to the anode could damage the anode and dispersion of carbon fines through the salt could cause shorting issues between the electrodes. This shorting behavior should be investigated as described in section 4.1. The anode would be a second glassy carbon rod also shrouded with a MgO tube to help protect it from Li and fines. Total charge passed (directly affecting the quantity of Li metal formed) will vary between the three tests, with the starting point based on the charge passed in the published work [24]. Applied current density should be minimized in order to maximize the amount of time the glassy carbon and Li are in contact. Following each test, the electrodes should be removed from the salt, visually examined for damage, and weighed.

Three additional experiments should be performed with a reaction-bonded SiC rod as the cathode to determine the compatibility of the TRISO fuel SiC layer with Li metal. These experiments should be performed at the same current densities and total charge passed as was done with the glassy carbon cathodes. Following each test, the electrodes should be removed and visually examined.

Before and after each of the six tests salt samples should be taken and analyzed to assess the depletion of LiCl and migration of carbon dust from the shrouded cathode. Nominally, 2-3 samples should be taken at each point and placed into individual containers. Between 1-2 samples from each set should be analyzed for Li, K, and C concentration, most likely via ICP-OES. Between 14 and 21 salt samples would be generated for analysis during these experiments to understand the feasibility of pyrochemical dissolution as a head-end process for TRISO fuel.

5. CONCLUSION

Due to an expected increase in the demand for HALEU to supply advanced reactors with fuel, the application of pyroprocessing to pebble-type fuels was assessed. The main challenge for pyroprocessing this fuel type is determining the effect of coating materials on pyroprocessing unit operations and the availability of a head end process that exposes the fuel kernel to salt without generating unacceptable amounts of carbon fines. The tolerance of pyroprocess unit operations to carbon will dictate whether the coating materials must be removed from the kernel during head end operations or only be breached to allow salt contact with the fuel kernel. This will indicate head end processing requirements. The use of several head end processes was evaluated based on their suitability for this fuel type and pyroprocessing. The three processes deemed most suitable included mechanical separations of coating materials from fuel kernels, acoustic fracture and pyrometallurgical dissolution using halide salts. A set of recommended tests to demonstrate feasibility of applying pyroprocessing to pebble-type fuels were laid out to address the identified technological gaps.

Other issues related to waste disposal to be addressed include likely regulatory controls, suitable waste forms for processing wastes, and benefits relative to direct disposal of used pebble-type fuels. The feasibility of pyroprocessing pebble-type fuels can be better assessed after head end process requirements are identified and waste disposal paths are assessed. The benefits of pyroprocessing can then be assessed by comparing the costs of required head end process operations, implementing a pyroprocessing flowsheet, and waste disposal with the value of the recovered HALEU fuel.

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