

Alternative Breaching Methods of the TRISO Fuels

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Abstract

The head-end processes of spent TRISO fuel have been reviewed to understand the current status and the limitations of the reported processes. The main concerns in the TRISO treatment are to effectively breach and separate the carbon and SiC layers composing the TRISO particles. The crush-burn scheme which was considered in the early stages of the development has been replaced by the crush-leach or CO₂ burning and the succeeding CO decomposition process because of a sequestration problem of CO₂ containing ¹⁴C. However there are still many obstacles to overcome in the reported processes. Hence, innovative thermomechanical and pyrochemical concepts to breach the coating layers of the TRISO particle with a minimized amount of second waste are proposed in this paper and their principles are described in detail.

Key words : TRISO, HTGR, thermal shock, breaching

1. Introduction

1-1. Review of the Head-End Process of TRISO Fuel

High Temperature Gas Cooled Reactor (HTGR) is being developed as one of the Generation IV nuclear energy systems. The fuel used in this reactor consists of a small particle designated as TRISO(usually less than 1mm) which has a center kernel of fissile fuel coated with layers of pyrolytic carbon and silicon carbide. The silicon carbide layer is sandwiched between the inner and outer pyrolytic carbon layers to be laid in a compression during an operation. Hence, TRISO fuels are capable of sustaining a high pressure build up by a very high burnup but they require a good fuel design and fabrication. In the case of the USA the inventory of the spent HTGR fuel had already exceeded 24 MT by 2002 and the quantity all over the world will be rapidly increased after the commercialization of the HTGR.

There are three options for a spent fuel treatment, namely (1) reprocessing, (2) long-period repository and reprocessing (intermediate storage) and (3) a direct disposal[1]. Last two

Table 1. Summary of the head-end treatment procedures of the TRISO fuel

Process	Chemical					Mechanical		Electrical
	Thermochemical	Electrolytical Disintegration[3]	Pure chemical disintegration[4]	Halogen gas oxidation[5]	Decomposition in molten salt[6]	Jet milling[7]	Crushing/Grinding and burning[8-10]	
<p>Thermal shock[2]</p> <p>Treating whole fuel elements at 2500~3000 °C and a succeeding chemical separation treatment</p>	<p>Forming graphite intercalation compounds with anions of the electrolyte. The higher the acid concentration the more efficient the disintegration effect</p>	<p>Applying vapors of potassium, cesium, bromine and FeCl₃</p>	<p>Decomposing SiC into SiCl₄ and Silicate in a molten chloride and carbonate</p>	<p>Jet grinding of burned TRISO-particles to remove the SiC layer and a second burning to remove the inner carbon layers</p>	<p>Crushing/grinding the fuel element including the broken-open fuel particles or Separating the fuel particles from the element and coring a kernel</p> <p>Burning crushed blocks under oxygen at ~875 °C</p>	<p>Passing high-voltage electrical pulses through a compact in water</p>		
<p>Description of the process</p>	<p>Forming graphite intercalation compounds with the anions of the electrolyte(e.g., sulfate or nitrate)</p> <p>Oxidation of the carbon atoms along the grain boundaries of the graphite body, forming CO₂ at the anode</p>	<p>Causing a strong lattice deformation followed by a crystal breakup</p> <p>Formation of graphite Interlaminar Compounds (K, Cs, Br, FeCl₃ gas)</p>	<p>Fast reaction rate</p> <p>Dust free</p> <p>Collectable silicon component</p>	<p>Easy scaling-up</p>	<p>Explosion by the pressure of the evaporated graphite built up as a result of the direct conversion of solids into a gas during the high-voltage pulses</p>			
<p>Main effects</p>	<p>Formation of soluble carbides by the reaction between the PyC and SiC coatings</p> <p>Embrittlement of the graphite matrix and the particle coatings</p>	<p>Efficient process by K, Ce, Br, FeCl₃ gas</p>	<p>Corrosive, difficulties in handling molten salt</p>	<p>Contamination of FPs to crushed fines due to possible Kernel breakage</p>	<p>Reduction of the maintenance works of the roll-gap clearance of the roll grinder</p>			
<p>Advantages</p>	<p>Simple process</p>	<p>Low temperature process</p>	<p>Corrosive, sophisticated process</p>	<p>Corrosive, sophisticated process can be adopted</p>	<p>Reducing the number of processing steps</p> <p>Low temperature, minimized 2nd contamination</p>	<p>Easy separation of the coated particles from the graphite fines</p>		
<p>Disadvantages</p>	<p>U, FPs migration to PyC, SiC layers (2nd contamination) and a reaction with the graphite part of the furnace</p>	<p>U, FPs co-dissolution(2nd contamination), vessel corrosion, low reaction rate</p>	<p>Kernel breakage and Off-gas treatment including ¹⁴C, CO₂</p> <p>Difficulties in maintenance works of roll-gap clearance of roll grinder</p>	<p>Difficulties in a technological optimization due to the lack of phenomenological understandings</p>				

options have been considered as an application to reprocess and dispose of TRISO because its three layers are able to serve as pressure vessels to contain the fission products and the fuel. However, the reprocessing concept has been accepted by most countries to get rid of the possible fission products released through coating failed particles and to reduce the volume of the highly toxic wastes. The key technology to treat spent HTGR fuel is how to breach the coating layers and separate the kernel which consists of the fuel and fission products from the breached coating layers.

Thermochemical, chemical, mechanical and electrical methods have been attempted for the breaching of the spent TRISO particle. Each process has unique features as shown in Table 1, however the procedure should be readily performed, provide a highly efficient disintegration of the fuel balls and a separation of the components and generate a minimum amount of waste material.

In the case of the thermochemical process, a very high temperature ranging from 2500~3000 °C induces a thermal shock in the PyC and SiC and causes the fuel kernels to react with the layers, including graphite, to yield soluble carbides. As a consequence of the high temperature treatment, a succeeding chemical separation treatment is possible. However, FPs form intercalation compounds with the graphite components in the furnace during an operation which have been recognized as insurmountable amounts of material and corrosion problems [2]. Formation of graphite intercalation compounds with the anions of the electrolyte (e.g., sulfate or nitrate) lead to a lattice growth on the c-axis and thus to a destruction of the lattice structure during the electrolytical disintegration process[3]. Similarly vapors of potassium, cesium, bromine and FeCl₃ are used to form graphite inter laminar compounds during the pure chemical process[4]. Although the effect is strong and efficient, its practical application is of limited value due to the enormous corrosion problems along with the very sophisticated engineering process. The other chemical processes are to decompose the carbon and silicon carbide by the formation of gaseous halogens compounds in Cl₂ and F₂ at a high temperature[5] or in a molten chloride or carbonate[6]. These processes are advantageous in that crushed fines and a large volume of the CO or CO₂ gases are not produced, whereas the difficulties in the handling of corrosive gases and a molten salt should be considered. Mechanical process consists of two substantial approaches, thoroughly grinding the fuel elements[7] and coring a kernel[8,9]. Former has potential drawbacks due to the formation of a mellithic acid (C₁₂O₁₂H₆) or similar compounds from the interaction of a strong nitric acid with ground graphite. Their presence can affect the solvent extraction process by a complex Pu(IV) and probably Th(IV) and by increasing the coalescence times

under the uranium backwashing conditions. The latter was introduced to overcome the formation of an organic acid. This process involves the elimination step of the graphite during each crushing step, then it obtains the pure fuel kernel as an oxide powder[8]. To realize a coring without a failure of the kernel and a contamination of the FPs to the crushed fines, a sophisticated hard crusher is used. However the operation condition of the hard disk crusher is very limited, i.e. the clearance of the crushing gap should be kept to within $\pm 0.15\text{mm}$ which is too risky in terms of a commercial scale. Hence an alternative crushing process which guarantees a soundness of the fuel kernel is necessary while minimizing the contamination of the FPs to the breached fines. Latest attempts to eliminate the carbon are with an electrical disintegration process[6,10]. In this process high-voltage electrical pulses are passed through a graphite compact. This results in an electrical breakdown of the substances with graphite and an explosion of the compact. The explosion is initiated by the pressure of the evaporated graphite built up as a result of the direct conversion of the solids into a gas during the high-voltage pulses. In this step, the coated particles should be intact while the size of the graphite is reduced to much lower than that of the coated particles. Hence the separation of the breached graphite from the coated particles could be easily achieved.

Among the reported head-end treatment processes of a coated fuel, only a few options have been continuously investigated. The main point that should be considered in the treatment process is how to minimize the carbon component combined with the other elements such as

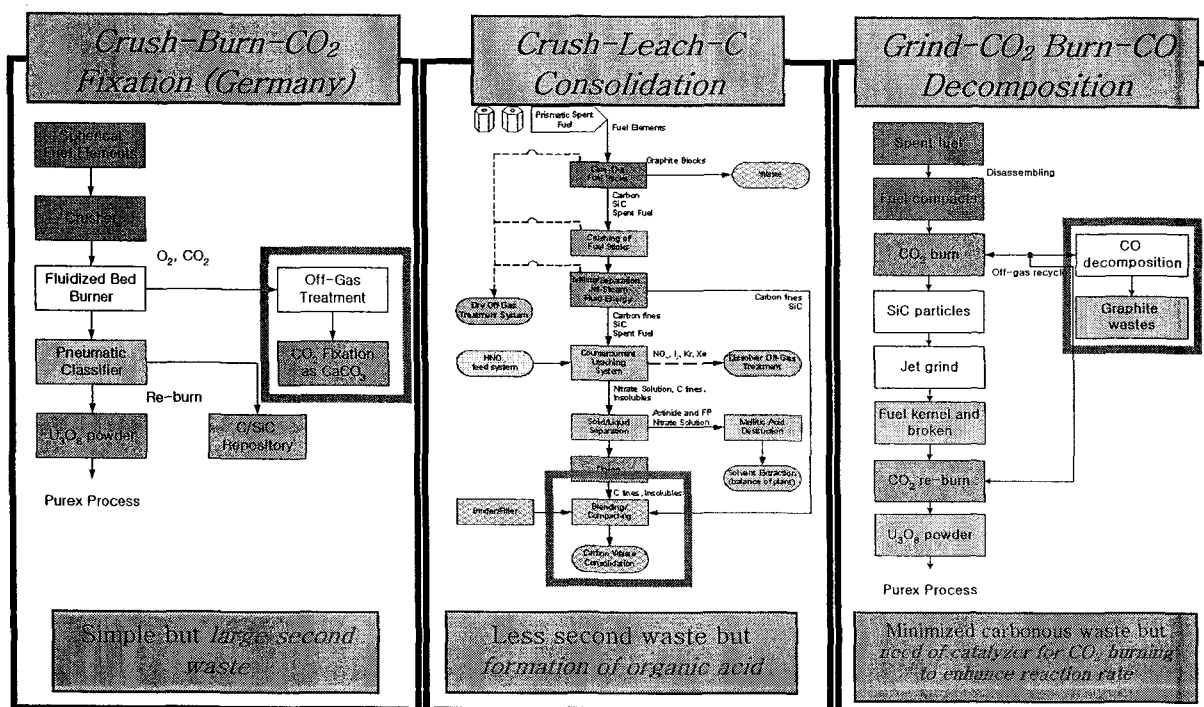


Fig. 1. Comparison of the reported TRISO fuel treatments

CO₂, CF₄ and CCl₄ etc. Hence, a dry mechanical crushing and grinding cycle is regarded as a promising method for a practical head-end processing of the HTGR fuels as shown in Fig. 1. The current dry crushing concepts of spent TRISO fuels are classified into three large groups; crush-burn-CO₂ fixation, crush-leach-C consolidation and grind-CO₂ burn-CO decomposition. The two processes being developed in ORNL(USA) and JAERI(JAPAN) are different from the German concept in that they don't generate CO₂ gas any more. However, these processes still have many obstacles to overcome such as the formation of organic acid during the leaching process and an efficiency enhancement of a CO₂ burning etc.

Hence, the objective of this study is to develop innovative methods for breaching the protective outer layers (porous carbon, inner pyrolytic carbon, silicon carbide barrier coating, and outer pyrolytic carbon) of the spent TRISO nuclear fuel. The innovative technologies include a radio frequency (RF) induction heating and a microwave heating connected with a rapid quenching to accelerate the internal gas expansion in the fuel kernel and thereby cause a stress cracking of the outer layers surrounding the fuel kernel.

1-2. Stability of the TRISO Particle

In the high temperature condition, the SiC layer in TRISO is laid under a compression by the interaction between the IPyC and OPyC layers. Hence, the structural integrity of the TRISO particle such as it's pressure vessel makes it difficult to be breached especially by a pressure build up with a high temperature. The high temperature durability of the ceramic coated fuel particles were investigated by Schenk et al.[11] and Ogawa et al. [12]. Usually a SiC layer without an outer PyC layer can suffer a serious degradation by an irradiation at about 1400 °C for less than two days. Even with the intact outer PyC, it decomposes so rapidly above 2300 °C that the coated fuel particles lose their diffusion barrier for the metallic

Table 2. Published results on the stability of TRISO [12].

Temp, °C	SiC-TRISO (conventional)	ZrC-TRISO	ZrC/(Th,U) O ₂	Biso
1850	Intact	Intact	Intact	Intact
2100	Intact	Intact	Intact	Intact
2450	Pressure-failed	Swelling without U migration outside of ZrC	Intact	Swelling and U migration into PyC
2500	-	5/8 pressure failed U migration	Intact	Pressure-failed
2550	-	-	Intact	-

FPs. Furthermore a pressure failure starts only after 2450 °C in the case of conventional TRISO, and more for the ZrC coated fuel particles, see Table 2. Consequently, a breaching without a contamination of the FPs to the breached fines by a conventional heating method appears to be difficult from a previous report on the stability of TRISO in a high temperature. There is a fundamental temperature limit in the thermal breaching method regardless of the heating scheme, because the temperature should not exceed more than ~2500 °C which is the onset temperature of a migration of the FPs through the diffusion barrier.

2. Thermomechanical breaching under consideration

2-1. Critical ΔT for Inducing a Fracture (Simple Geometry)

When each face of a continuum is placed in a different temperature condition, there is a temperature gradient as shown in Fig. 2. This temperature difference causes a thermal stress which is expressed by Eq. 1.

$$\sigma_{\max} = \frac{E\alpha\Delta T}{1-\nu} f(\beta) \quad (1)$$

where

σ_{\max} : maximum stress

E : Young's modulus

α : thermal expansion coefficient

ΔT : quench temperature differential

ν : Poisson's ratio

$f(\beta)$: function of the heat transfer conditions

$$\beta = \frac{ah}{k}$$

a : characteristic length, h : surface conductance,

k : thermal conductivity of sample

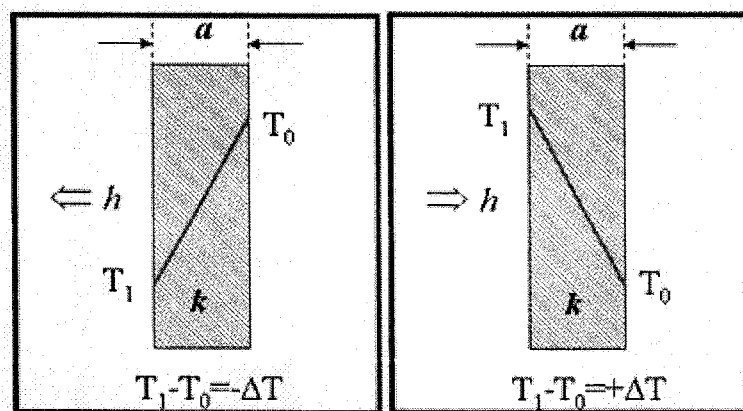


Fig. 2. Temperature gradient for a cooling(left) and a heating(right)

When the maximum stress, σ_{\max} exceeds the tensile stress of the material, there will be a crack. This temperature difference to induce a crack in the material is defined as the critical temperature difference, ΔT_c as depicted in Eq. 2.

$$\Delta T_c = \frac{\sigma_i(1-\nu)}{E\alpha f(\beta)} \quad (2)$$

where σ_i : tensile strength

ΔT_c : critical temperature difference

The critical temperature is a function of the mechanical property of a material and also the dimension of the material as shown in Fig. 3. In the case of silicon nitride which has a good toughness comparable to SiC, the critical temperature reaches 1400 °C, in TRISO size[13].

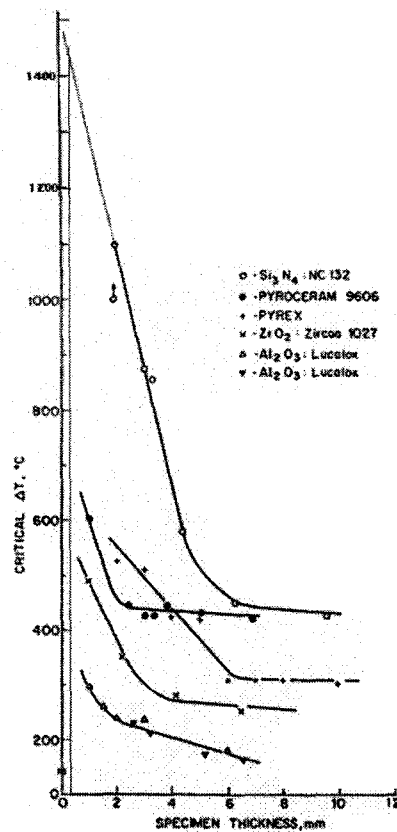


Fig. 3. Effect of a specimen size variation on the ΔT_c for various ceramics. Note: Biot number is proportional to the specimen size (red line is interpolated)[13]

2-2. Skin Effect during an Induction and Microwave Heating

The radio frequency and microwave are used for a heat-treating and a higher frequency which causes a higher concentration of the induced current at the surface by the skin effect. Shorter gap between the coil and the conductor causes an induced current and a

coil current which attract each other stronger at the surface by the proximity effect during the RF induction heating. The induced current flow within the object is most intense on the surface, and decays rapidly below the surface. This is described as the "skin depth", d_s of the object as shown in Eq. 3. The skin depth decreases when the resistivity decreases(ρ), permeability(μ) increases or the frequency increases(ω).

$$d_s = 5.03 \sqrt{\frac{\rho}{\mu\omega}} \quad (3)$$

Figure 4 shows the critical frequency as a function of the diameter for round bars[14]. Higher frequencies are needed to efficiently heat small bars, but once the critical frequency is reached, increasing the frequency has very little effect on the relative efficiency. Hence, a careful choice of the frequency as well as the power is essential to achieve a maximum efficiency.

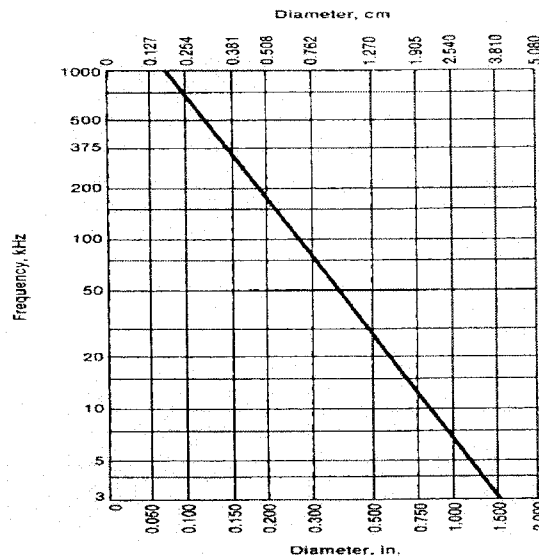


Fig. 4. Critical frequency as a function of the diameter for round steel bars[14].

2-3. Evaluation of the RF-Induction and Microwave Heating for a TRISO Heating

The alternative heating processes considered in this study have unique characteristics in the heating of a coated particle, but a common denominator is to heat up the TRISO fuel with a very high heating rate. The primary difference between the RF and microwave heating is the frequency range for deciding the heating mode. When we heat up a several centimeters sized sample as a fuel compact, the critical frequency should be chosen at below 10 kHz as shown in Fig. 4. The application of a higher frequency leads to only a surface heating of the compact. In the case of the heating of a TRISO particle, however, a much higher frequency is necessary to heat up an individual particle. Hence, the frequency range should be decided upon

according to which type of fuel, compact or coated particle, is to be treated. It is required that the filler carbon in the fuel compact is removed by a mechanical method before a breaching of the TRISO particle, so a microwave heating would be more preferable to heat up the particles.

2-4. Application of a Plasma to a TRISO Treatment

Plasma heat transfer processes are significantly more complicated than that for an unionized gas since the heat transfer phenomenon is greatly affected by the flow of the electrons and the ions. An electrical boundary layer is formed around the particle apart from the hydrodynamic and thermal boundary layers. The electrical, thermal and hydrodynamic effects must be considered simultaneously with the surface phenomenon to accurately determine the heat transfer from the plasma to the item being heated. Due to the heat transfer from the plasma, the particulates heat up and eventually melt when the retention time is excessive. The temperature history of TRISO during its time in a plasma flame was preliminary calculated by the commercial FEM code, AnsysV.5.5.1. The material properties of each component in TRISO and their thermal boundary conditions are summarized in Table 4.

Table 4. Material properties of the components in the TRISO fuel for the calculation of the temperature distribution.

	Thickness(m)	Density(kg/m ³)	Thermal conductivity(W/mK)		Specific heat(J/kgK)		Heat transfer coefficient(W/m ² K)
			300K	9.76	300	235	
Kernel	500E-6(dia)	10000	1773K	2.52	1773	338.4	-
			3120K	3.959	3120	792	
			300K	~200	300K	~540	
Buffer layer	100E-6	1000	300K	~200	300K	~540	4500
IPyC	35E-6	1850	300K	340	300K	711.72	Thermal contact
SiC	40E-6	3200	300K	250	300K	720	Thermal contact
OPyC	35E-6	1850	300K	340	300K	711.72	10000

The FEM mesh indicating the temperature measuring points and temperature profiles are depicted in Fig. 5. The initial and environmental temperature of TRISO were set to 300K and 8000K (as a plasma temperature) respectively. It was calculated that the OPyC temperature rapidly increased to the environmental temperature within 0.2 second while the rate decreased in the inner layers. One interesting result is the temperature of the kernel which was kept below its melting point within the first 0.6 sec. This is because the gas gap between the kernel and the buffer carbon layer and the porous buffer carbon layer itself with a lower

thermal conductivity acted as thermal barriers. Hence, with a carefully considered design of the plasma flame, an extremely rapid heat up of the coating layers without a considerable heating of the kernel should be possible.

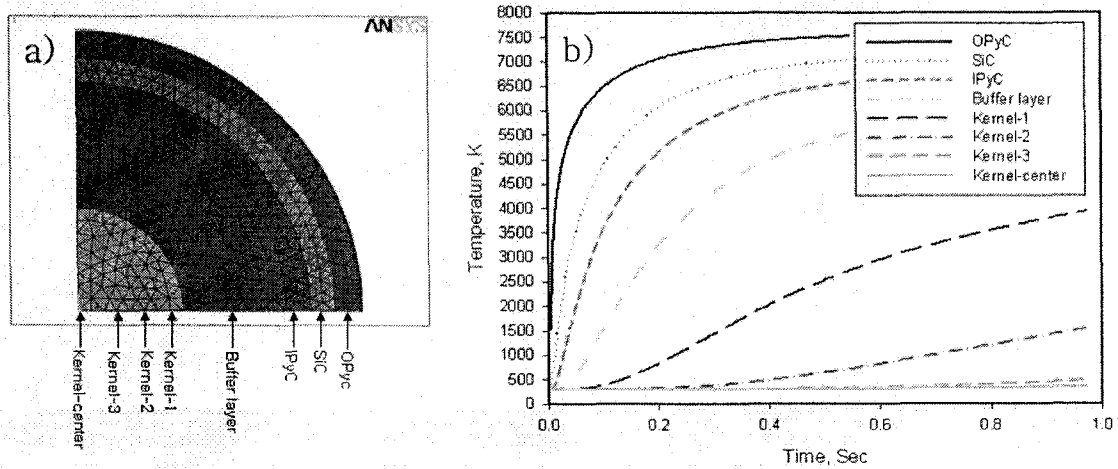


Fig. 5. FEM mesh indicating the T.C. points a) and the temperature profiles b).

2-5. Application of a Quenching to Proposed Heating Methods

An example of the experimental apparatus utilizing a electromagnetic and a plasma

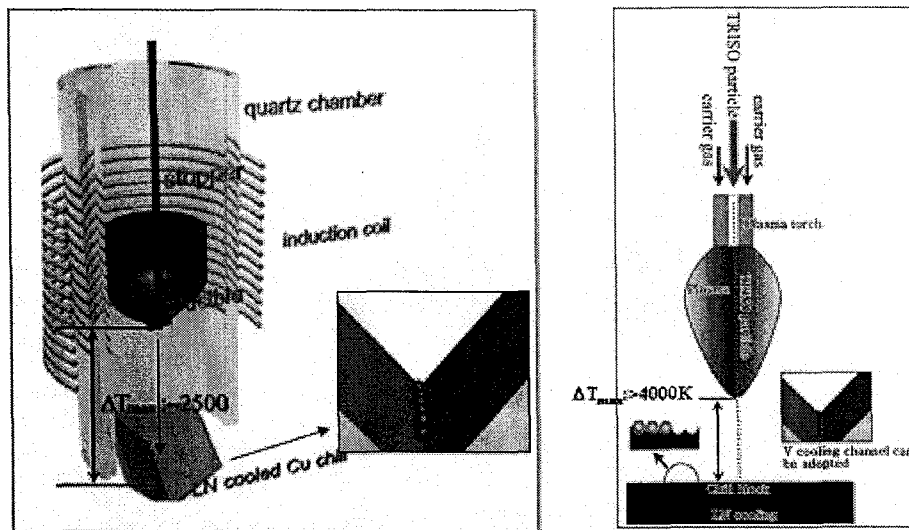


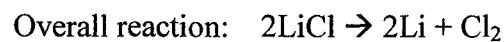
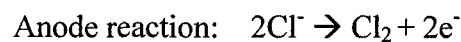
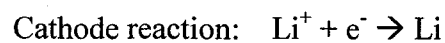
Fig. 6. Schematic diagram of the plasma (left) and induction heating (right) assisted quenching processes of the spent TRISO particle.

heating with a subsequent quenching is shown in Fig. 6. The heat up particles are impinged on the chilled surface, which is designed to wrap the particles. Liquid nitrogen could be chosen as a quenching medium, however gasified nitrogen would be an additional burden that would need to be treated. Heat transfer from the particles to the surface results in a rapid cooling and the formation of a temperature gradient on the surface. This induces a thermal crack in the

coated layers which are laid under the critical T. At this moment, the temperature change of the kernel would be milder than in the coated layers because of the porous carbon layer and the gas gap in the buffer carbon layer of the spent TRISO particle. Hence macro and micro cracks would only be induced in the coating layers.

3. Electrolytic molten salt carbon conversion(EMS-CC) process

For an efficient separation of the heavy metal fuel kernel from the TRISO coating layers composed of carbon (graphite), the recent research results by Chen G.Z. et al.[15] and Hsu W.K. et al.[16] upon the preparation of carbon nanotubes and nanoparticles were utilized. Carbon nanotubes and nanoparticles were electrolytically prepared from normal graphite in molten alkali chlorides, namely LiCl, NaCl, KCl. Visual observations showed that the carbon consumption for the nanotube formation occurs at the graphite cathode where the alkali metal forms during the electrolysis. The EMS-CC reaction which is proposed in this study is based on the above mentioned literature results of a carbon erosion reaction by the intercalation of the alkali metal ion for the removal of the TRISO coating layers. The electrochemical formation of CNTs in the molten salts is related to the erosion of the cathode, such behavior of a carbon cathode is, as was first pointed out by Fray D.J., common in the Hall-Heroult cell where the intercalation of alkali metals, particularly sodium, into the carbon cathode is known to have caused a cathode expansion, cracking and erosion[15]. Based on the fact that the CNT yield depended strongly on the electrolysis current and temperature, it was proposed that the alkali metal ion (M^+ : Li, Na, K) intercalates, under the influence of a sufficiently negative electrode potential, into the graphite lattice where it is reduced in situ. The alkali metal then expands the lattice and more strain is put on the lattice as the amount of metal increases, until the lattice fragments.



The carbon based fragments may then enter the molten salt and, without the protection of the graphite lattice, undergo through an inter- and/or intra-fragment recombination process, the formation of various carbon nano-materials in the electrolyte. For an application of the EMS-CC process to a TRISO fuel particle treatment it is required to obtain the conditions of a good disintegration of graphite thus avoiding the formation of carbon nano-materials.

3-1. Removal of the OPyC layer in the TRISO fuel particles

Fraction of the outer pyrolytic carbon layer (OPyC) volume in the TRISO coating layers is significant at around more than 30 %. Also, OPyC is not contaminated with the fission products due to the barrier of a SiC layer assuming that this layer was not damaged during the irradiation in the HTGR. It is possible that OPyC layers are disintegrated and removed by the EMS-CC treatment which is a carbon erosion reaction by the intercalation of the alkali metal ion if the TRISO fuel particles covered with OPyC layers are used as a cathode in the molten salt electrolysis cell. According to the intercalation mechanism, the erosion results from the fast intercalation of the Li^+ ion and its reduction inside the graphite lattice of the cathode. After a removal of the OPyC layers by an application of the EMS-CC process, the remaining SiC coated TRISO particles can be transported to the next breaching unit such as RF and EMIH which are described in this paper.

3-2. Removal of the IPyC and porous PyC layers in the TRISO fuel particles

The porous PyC coating layer, called the buffer layer, attenuates fission recoils and provides a void volume for gaseous fission products and carbon monoxide. The IPyC coating layer acts as a containment for the gases during an irradiation and protects the fuel kernel from a reaction with the coating gases during the SiC coating process. Therefore, the porous PyC and IPyC coating layers are always contaminated with the fission product gases and metals. After an application of the alternative breaching methods under development in this study and the separation of the breached particles such as SiC, porous PyC and IPyC from the fuel kernel, the recovered fuel kernel can be applied by the EMS-CC process for a further removal of the remaining porous PyC and IPyC layers on the kernel.

Until now, the feasibility of the EMS-CC process has been proven from the viewpoint of a disintegration of the graphite material by the intercalation mechanism of the alkali metal, but the scale of the electrolysis cell is small. For a future application of the EMS-CC concept for the treatment of the TRISO coated fuel particles, many studies on the EMS-CC process development are needed by using a graphite block and TRISO type PyC coating layered surrogate particles.

3-3. Removal of the SiC layer in the TRISO fuel particles

There are two kinds of options for breaching the SiC coating layer under consideration in this study. One is a thermomechanical breaching method such as RF and microwave heating which was mentioned earlier. The other is a chemical method for

breaching the SiC layer by the reaction of SiC with Mg metal which is produced by the electrochemical reduction of $MgCl_2$ in the molten salt phase. The Gibbs free energy of the $MgCl_2$ formation is -57.7 kcal/g-equiv.Cl. And the Gibbs free energy of the LiCl formation is -78.8 kcal/g-equiv.Cl. Electrolytic reduction of the Mg ion occurs prior to the reduction of the Li ion. This chemical breaching of the SiC layer can simultaneously occur during the course of the EMS-CC reaction for the removal of the OPyC layer. Thus a breaching of the TRISO coated particles for the removal of four layers, such as the porous PyC, IPyC, SiC and OPyC layers can occur in an EMS-CC reactor with a single step operation. According to the thermodynamic calculation result for the following SiC removal reaction by Mg metal, it is feasible that a chemical reaction of SiC with Mg moves in a forward direction because the Gibbs free energy of the Mg_2Si formation is -5 kJ/mol.

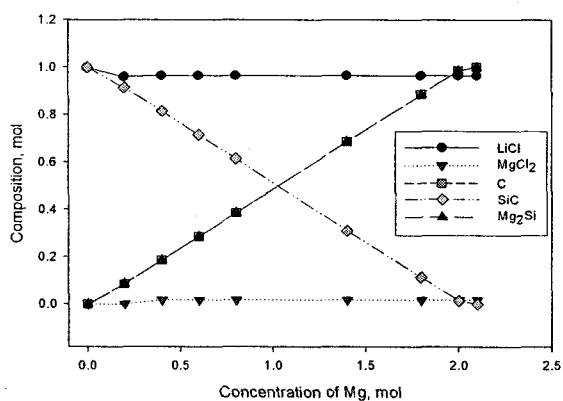


Fig. 7. Calculated equilibrium composition of $LiCl+xMg+SiC$ system according to the x ratio at 1000K.

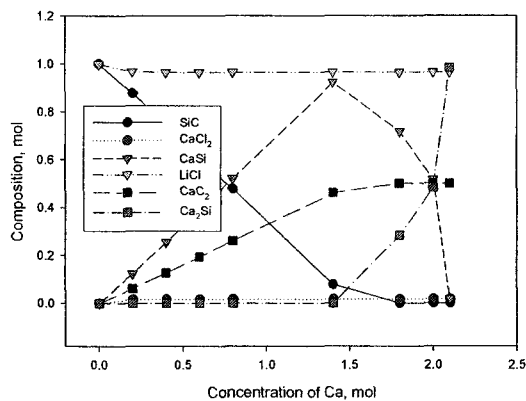
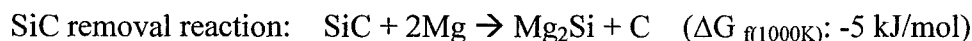


Fig. 8. Calculated equilibrium composition of $LiCl+xCa+SiC$ system according to the x ratio at 1000K.

It is believed that the reaction would be possible below 1000K in which any composition of $LiCl-MgCl_2$ is a liquid phase. The equilibrium compositions of the $LiCl+xMg+SiC$ system according to the mole ratio of Mg are calculated by a commercial thermodynamic program, THERMO[®] and the results are presented in Fig. 7. The concentration of Mg_2Si increases as the Mg content increases and the SiC is completely converted to Mg_2Si and the free carbon at 2.1 mol of Mg, is a little more than stoichiometry.



By introducing calcium chloride, the SiC layer can also be removed as shown in Fig. 8. Furthermore, the carbon also forms intercalates with magnesium as well as calcium[17]. This means a single step reaction would be possible to exfoliate a pyrolytic carbon layer as well as

a SiC layer in MgCl₂ and CaCl₂ or LiCl-KCl-MgCl₂/CaCl₂ and any combination of them. Furthermore, the Li as well as Mg might be recyclable from the reaction product such as C₆Lix and Mg₂Si by an anodic dissolution which means a closed treatment cycle is theoretically possible without a second waste generation.

4. Conclusions

The conventional treatment technologies of spent TRISO fuel were reviewed. It was found that the existing processes still have many obstacles to overcome in terms of a second waste generation as well as a proliferation resistance. Preliminary investigations have shown that the electromagnetic waves accelerate a heating then an internal gas expansion in the fuel kernel and thereby cause a stress cracking of the outer layers surrounding the fuel kernel. Also, a subsequent quenching process would enhance the formation of a microcracking in the coating layers. The innovative technologies such as a RF-induction, microwave and plasma heating with a rapid quenching under consideration in this study could be alternatives to treat spent TRISO fuel. In addition, the proposed electrochemical process was also promising due to a proliferation resistance and a compatibility with existing pyrochemical processes of spent fuel.

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