Feasibility Study on the Development of Alternative Methods for the Treatment of TRISO Fuels

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Abstract

In this study, conventional head-end processes of spent TRISO fuel have been reviewed to develope more effective treatment methods. The main concerns in the TRISO treatment are to effectively separate the carbon and SiC contained in the TRISO particles. The crush-burn scheme which was considered in the early stages of the development has been replaced by the crush-leach process because of ^{14}C problems as a second waste during the process. However there are still many obstacles to overcome in the reported processes. Hence, innovative thermomechanical 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, reprocessing, 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 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. The key technology to treat spent HTGR fuel is how to separate the kernel which consists of the fuel and fission products from the coating layers.

Thermochemical, chemical, mechanical and electrical methods have been attempted for the reprocessing of the spent TRISO particle. Each process has unique features as shown in

Table 1. Summary of the head-end treatment procedures

	Thermochemical	Chemical				Mechanical		Electrical
Process	Thermal shock[2]	Electrolytical Disintegration[3]	Pure chemical disintegration[4]	Halogen gas oxidation [5]	Decomposition in molten salt[6]	Jet milling[7]	Crushing/Grinding and burning[8,9]	Electrical disintegration[6,10]
Description of the process	Treating whole fuel elements at 2500~3000 ℃ and a succeeding chemical separation treatment	Forming graphite intercalation compounds with anions of the electrolyte.	Applying vapors of potassium, cesium, bromine and FeCl₃	Decomposing the coating layers by Cl₂ or F₂ at a high temperature	Decomposing SiC into SiCl₄ and Silicate in a molten chloride and carbonate	Jet grinding of burned TRISO particles and a leaching after a second burning	Throughly grinding the fuel element including the broken-open fuel particles or Separating the fuel particles from the element and coring a kernel	Passing high voltage electrical pulses through a compact in water
Advantages	Simple process	Low temperature process	Efficient process by K, Ce, Br, FeCl₃ gas	Selective volatilization, chemical extraction can be adopted	· Fast reaction rate · Dust free · Collectable silicon component	Easy scaling-up	· Reducing the number of processing steps · Low temperature, minimized 2nd contamination	Easy separation of the coated particles from the graphite fines
Disadvantages	U, FPs migration to PyC, SiC layers (2nd contamination) and a reaction with the graphite part of the furnace	U, FPs co-dissolution(2nd contamination), vessel corrosion, low reaction rate	Corrosive, sophisticated process	Corrosive, sophisticated process	Corrosive, difficulties in handling molten salt	Contamination of FPs to crushed fines due to possible Kernel breakage	· Kernel breakage and Off-gas treatment including ¹⁴C, CO₂ · Difficulties in maintenance works of roll-gap clearance	Difficulties in a technological optimization due to the lack of phenomenological understandings

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 Cl_2 and F_2 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 CO_2 gases are not produced, whereas the difficulties in the handling of corrosive gases and a molten salt should be considered. A dry mechanical crushing and grinding cycle is regarded as a promising method for a practical head-end processing of the HTGR fuels. Mechanical process consists of two large directions, thoroughly grinding the fuel elements[7] and coring a kernel[8,9]. Former has potential drawbacks due to the formation of a mellithic acid ($C_{12}O_{12}H_6$) 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 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 ±0.15mm 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 other elements such as CO_2, CF_4 and CCl_4 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 as shown in Fig. 1. The high temperature urability 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

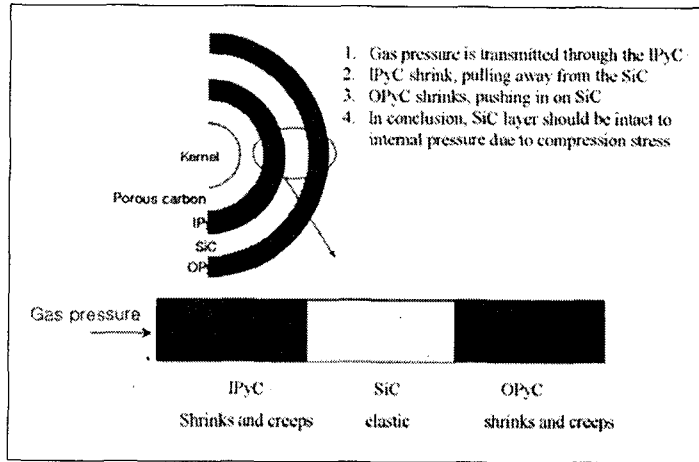irradiation at about 1400 ℃ for less than two days. Even with the intact outer PyC, it

Fig. 1. Structural stability of TRISO[11].

decomposes so rapidly above 2300 ℃ that the coated fuel particles lose their diffusion barrier

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

Temp. ℃	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	–

for the metallic FPs. Furthermore a pressure failure starts only after 2450 ℃ 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 ℃ which is the onset temperature of a migration of the FPs through the diffusion barrier.

2. Treatment Concepts 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.

$$\sigma_{max} \cong \frac{E\alpha\Delta T}{1-\nu} f(\beta)$$
(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,
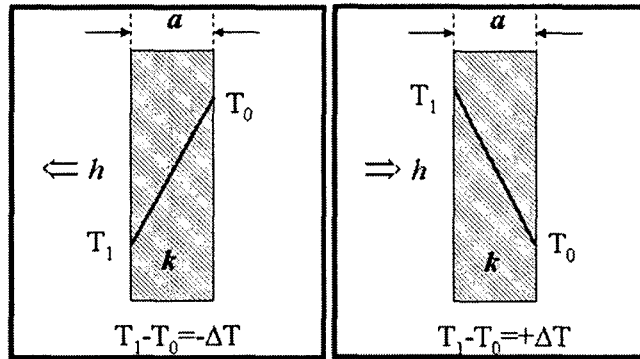
k : thermal conductivity of sample

Fig. 2. Temperature gradient on cooling(left) and 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.

$$\Delta T_c = \frac{\sigma_i (1-\nu)}{E\alpha f(\beta)}$$
(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 ℃, approximately in TRISO size[13].

2-2. Rapid Heating and Quenching Schemes

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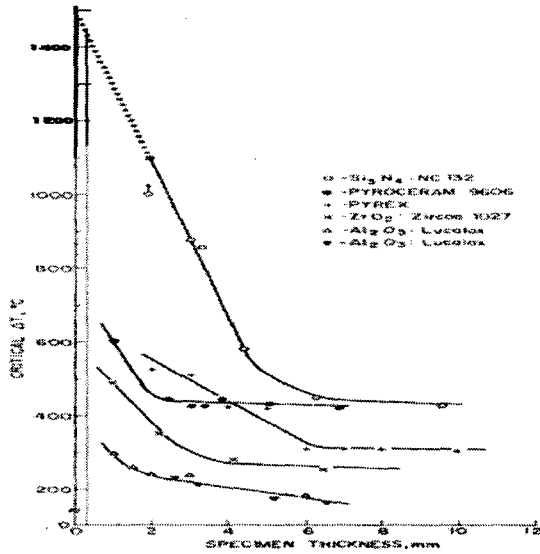
Fig. 3. Effect of a specimen size variation on the Δ Tc for various ceramics. Note:Biot number is proportional to the specimen size(dotted line is interpolated)[13].

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).

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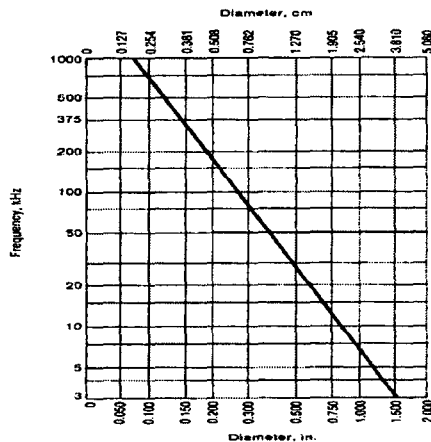

Fig. 4. Critical frequency as a function of diameter for round bars[14].

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.

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

The alternative heating processes considered in this study are summarized in Table 3. Each process has 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 ㎑ 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 as depicted in Table 3.

Table 3. Characteristics of various induction heating methods.

Heating methods	Frequency range	Characteristics
Induction heating	~10 ㎑	Applicable to large sample, suitable for the heating of fuel compact
Radio frequency heating	70㎑~0.3㎓	Wide Choosability of frequency according to sample size, suitable for the heating of TRISO particle
Microwave heating	0.3~300 ㎓	Surface heating, suitable for the heating of TRISO particle

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 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/m³)	Thermal conductivity(W/m°K)		Specific heat(W·sec/kg°K)		Heat transfer coefficient(W/m²K)
Kernel	300E-6(dia)	10000	300K	9.76	300	235	-
			1773K	2.52	1773	338.4	
			3120K	3.959	3120	792	
Buffer layer	150E-6	1000	300K	~200	300K	~540	4500
							Thermal contact
IPyC	35E-6	1850	300K	340	300K	711.72	Thermal contact
SiC	35E-6	3200	300K	250	300K	720	10000
OPyC	40E-6	1850	300K	340	300K	711.72	

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 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.

Application of a Quenching to Proposed Heating Methods

An example of the experimental apparatus utilizing a electromagnetic and a plasma 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.
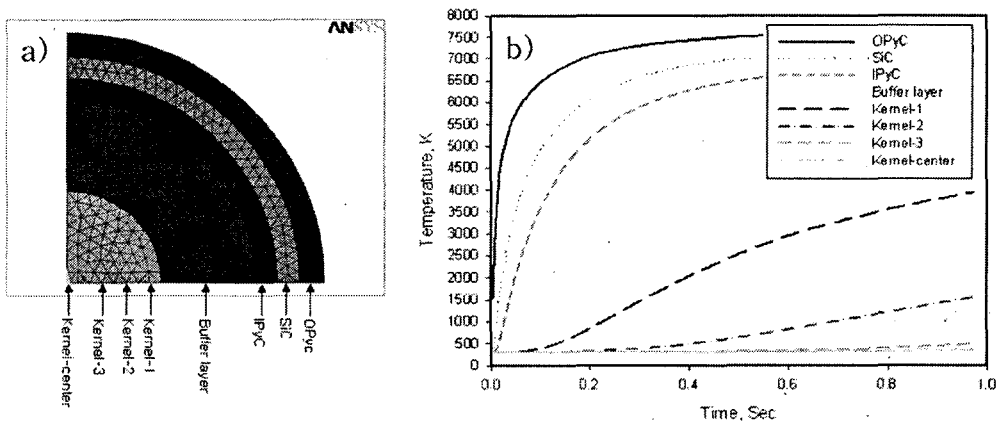
Fig. 5. FEM mesh indicating the temperature measurement points a) and their profiles b).

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.
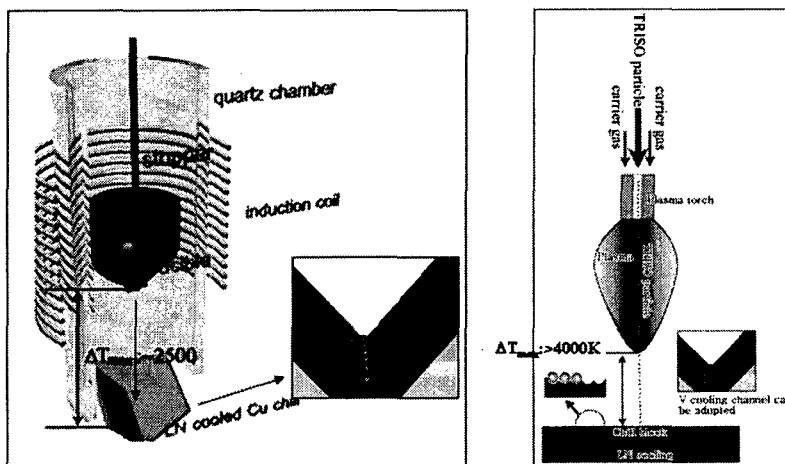

Fig. 6. Schematic diagram of the electromagnetic wave(a) and plasma(b) assisted quenching process of the spent TRISO particle

3. 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 proliferation resistance. Preliminary investigations have shown that the electromagnetic waves accelerate the 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.

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