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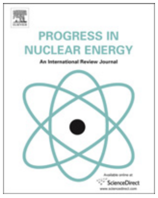

# Modeling spent TRISO fuel for geological disposal: corrosion and failure under oxidizing conditions in the presence of water

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ABSTRACT

High temperature gas reactors (HTGRs) are being considered for near term deployment in the United States under the GNEP program and farther term deployment under the Gen IV reactor design (U.S. DOE Nuclear Energy Research Advisory Committee, 2002). A common factor among current HTGR (prismatic or pebble) designs is the use of TRISO coated particle fuel. TRISO refers to the three types of coating layers (pyrolytic carbon, porous carbon, and silicon carbide) around the fuel kernel, which is both protected and contained by the layers. While there have been a number of reactors operated with coated particle fuel, and extensive amount of research has gone into designing new HTGRs, little work has been done on modeling and analysing the degradation rates of spent TRISO fuel for permanent geological disposal. An integral part of developing a spent fuel degradation modeling was to analyze the waste form without taking any consideration for engineering barriers. A basic model was developed to simulate the time to failure of spent TRISO fuel in a repository environment. Preliminary verification of the model was performed with comparison to output from a proprietary model called GARGOYLE that was also used to model degradation rates of TRISO fuel. A sensitivity study was performed to determine which fuel and repository parameters had the most significant effect on the predicted time to fuel particle failure. Results of the analysis indicate corrosion rates and thicknesses of the outer pyrolytic carbon and silicon carbide layers, along with the time dependent temperature of the spent fuel in the repository environment, have a significant effect on the time to particle failure. The thicknesses of the kernel, buffer, and IPyC layers along with the strength of the SiC layer and the pressure in the TRISO particle did not significantly alter the results from the model. It can be concluded that a better understanding of the corrosion rates of the OPyC and SiC layers, along with increasing the quality control of the OPyC and SiC layer thicknesses, can significantly reduce uncertainty in estimates of the time to failure of spent TRISO fuel in a repository environment.

## 1. Introduction

Currently in the United States, high temperature gas reactors (HTGRs) such as the Gas Turbine-Modular Helium Reactor (GT-MHR) and the Very High Temperature Reactor (VHTR) are being designed to use TRISO coated particle fuel (U.S. DOE Nuclear Energy Research Advisory Committee, 2002). TRISO fuel is a spherical fuel form that is about 1.5 mm in diameter. The fuel is composed of a 0.35-mm diameter fuel kernel of uranium ($UO_2$, UCO, or $UC_2$) surrounded by four coating layers: porous carbon buffer, inner pyrolytic carbon (IPyC), silicon carbide (SiC), and outer pyrolytic carbon (OPyC), respectively. A magnified image of the coating layers of TRISO fuel can be seen in Fig. 1. These TRISO fuel particles are typically placed into cylindrical graphite compacts or into coated graphite pebbles.

One of the obstacles for nuclear power from the public perception is the radioactive waste generated by nuclear power. One way to better understand the degradation of spent nuclear fuel in a geological disposal site is to model the waste form and not just the engineering barrier system. A sensitivity analysis was performed on spent TRISO fuel to determine which parameters would have the most impact on the overall performance of the fuel. The focus of this paper is the failure of spent TRISO fuel under oxidization conductions in the presence of water. Only the individual TRISO fuel particles were analyzed for degradation rates even though some disposal plans include disposing the TRISO fuel surrounded by

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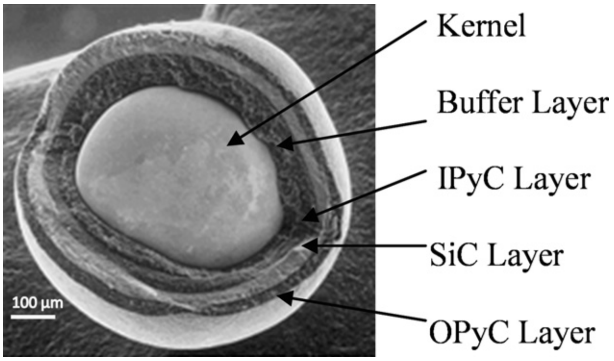

**Fig. 1.** Cross-section of TRISO fuel coating layers (DOE, 2002).

graphic in either the prismatic or pebble form (Grambow et al., 2006). This approach is very conservative and the degradation of the surrounding graphic along with the transportation of the radionuclides through the geological system should be taken into consideration for a more complete repository study.

## 2. Model development and implementation

A TRISO fuel particle can be modeled as a pressure vessel that contains fission products. As with any pressure vessel, when it corrodes significantly it will erupt. This eruption will release gaseous fission products and allow the other radioactive material to be available for transport out of the particle. The SiC layer acts like a pressure vessel within the TRISO fuel. Corrosion of this layer is caused by oxidation with air and water at the outer surface. The inner surface of the SiC layer is corroded in-reactor by chemical interactions with fission products. Gas volume within the TRISO particle increases over time with the production of stable fission gases, radiogenic helium, and carbon monoxide (CO). Carbon monoxide formation occurs at high fuel burn up when excess oxygen in the kernel chemically reacts with the surrounding carbide layers. One may expect that when the stress on the SiC layer exceeds the strength, the SiC layer will fail, allowing oxidation of the kernel to occur. Kernel oxidation exposes fission products and actinides, which can be dissolved and transported away from the kernel as dissolved ions or as colloids.

A model was developed to represent the degradation of spent TRISO fuel after emplacement into a geological repository environment. Degradation of the TRISO particle was modeled as the complete corrosion of the OPyC layer followed by the corrosion of the SiC layer. The TRISO fuel particle was assumed to fail as soon as the SiC layer failed from corrosion and pressure buildup. Corrosion rate is the amount of mass lost per surface area and time, which can be written as follows (Equation (1)):

$$k = \frac{dm_l}{A_s dt} = \frac{\rho A_s dx}{A_s dt} = \frac{\rho dx}{dt} \tag{1}$$

$k$ = corrosion rate [g/m$^2$/day]
$dm_l$ = mass loss [g]
$A_s$ = surface area [m$^2$]
$dt$ = time [day]
$\rho$ = density of material [g/m$^3$]
$dx$ = mass loss thickness [m]

Corrosion rates used in model calculations with Equation (1) came from Fachinger et al. as seen in Table 1 (Fachinger et al., 2006). It should be noted that all of the corrosion rate experiments performed by Fachniger et al. took place in a sealed glass tube containing approximately 110 ml of water at 90 °C. The temperature dependence of corrosion is described by the empirical Arrhenius expression as seen in Equation (2) (Talbot and Talbot, 1998).

**Table 1**
Corrosion rates used in the TRISO fuel failure model for the OPyC and SiC layers.

| Corrosion Rates | k (g/m$^2$/day) | T(°C) | Activation Energy (kj/mol) | Frequency Factor (g/m$^2$/day) | Reference |
|---|---|---|---|---|---|
| OPyC | 3.40E-07 | 90 | | | (Fachinger et al., 2006) |
| SiC | 9.64E-06 | 90 | 41.8 | 27.9 | (Fachinger et al., 2006) |

$$k = k_0 \exp\left[\frac{-Q}{RT}\right] \tag{2}$$

$k_o$ = frequency factor [g/m$^2$/day]
$Q$ = activation energy [kJ/mol]
$R$ = ideal gas constant [8.314 J/mol/K]
$T$ = absolute temperature [K]

Terms $-Q/R$ and $k_o$ of the Arrhenius expression can be determined numerically for known corrosion rates at different temperatures. This method is demonstrated in Fig. 2, in which the natural log of the corrosion rate value is plotted as a function of inverse temperature (1/T) and fit with a line. The corrosion data for this plot came from Fachinger et al. (Fachinger et al., 2006). Rearranging Equation (1) gives an expression for calculating time to complete degradation of the OPyC layer.

$$dt = \frac{\rho_{OPyC}(r_{OPyC} - r_{SiC})}{k_{OPyC}} \tag{3}$$

$dt$ = change in thickness of the OPyC layer
$\rho_{OPyC}$ = density of OPyC [g/m$^3$]
$r_{OPyC}$ = outside radius of the OPyC layer [m]
$r_{SiC}$ = outside radius of the SiC layer [m]
$k_{OPyC}$ = corrosion rate of OPyC [g/m$^2$/day]

The model assumes that SiC corrosion begins after the OPyC layer has completely corroded. As the SiC layer thins, the stress of the SiC layer increases. This phenomenon is described by Equation (4), which is used to represent thin pressure vessels (Ebert et al., 2000).

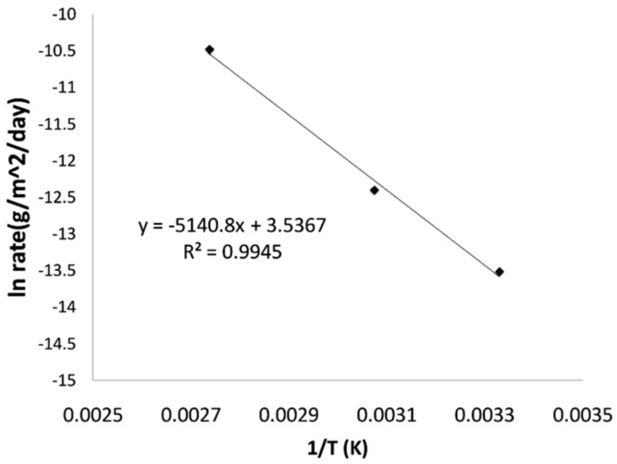

**Fig. 2.** Arrhenius plot of the corrosion of SiC at three different temperatures.