INVESTIGATION OF AG AND PD FISSION PRODUCTS PENETRATION DEPTH IN ZRC LAYER OF HTGR TRISO FUEL PARTICLE USING SRIM/TRIM MONTE CARLO SIMULATION

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Abstract *High-temperature gas-cooled Reactors (HTGRs) is one type of Generation IV reactor that uses TRISO (tri-structural isotropic) coated-fuel particles (CFP) for containment of radioactive fission products, which is produced from the fission reaction of UO2 fuel. ZrC has been proposed to be the main barrier for containing fission products either as a replacement of the SiC layer or as an additional layer of the TRISO fuel particle to overcome the corrosion issue of SiC because of interaction with the fission product of silver (Ag) and palladium (Pd). ZrC is an excellent material because it has good physical and nuclear properties, i.e., high corrosion-resistant, excellent thermal shock resistance and a small cross-section for neutron capture. ZrC is expected to provide a better barrier against Ag and Pd diffusion attacks than SiC. However, ZrC is very challenging to manufacture, so it depends on factors such as microstructure, chemical composition and interactions, morphology and impurities. Many attempts have been made to study the interaction phenomena of Ag and Pd with ZrC that cause corrosion. Here, the penetration depth of those two fission products was studied using SRIM (Stopping and Range of Ions in Matter) /TRIM (TRansport of Ions in Matter) for simulation with 0.1-10 MeV of kinetic energies. The results provide detailed information about the Ag/ZrC and Pd/ZrC Ion Ranges and Doses. In addition, Ag and Pd's products of the depth and concentration within ZrC were observed as important first steps in understanding the corrosion phenomena of her ZrC layers in TRISO particles.*

INTRODUCTION

Energy is the most important human need to enhance society's living standards. The global energy demand is growing very fast, even faster than the population growth. From the recent data, energy demand could be around 50 TWh (1); about 80% comes from fossil fuels (2) and about 14% comes from nuclear energy (3). By using carbon dioxide that absorbs infrared rays from the earth's surface, the heat energy can not be released into space. It is called the greenhouse effect and causes global warming or increases the earth's temperature. Nuclear power could be one option for reducing carbon dioxide emissions.

Since the occurrence of large-scale nuclear power plant accidents such as the SL-1 accident (1961), the Three Mile Island accident (1979), the Chernobyl accident (1986), and the Fukushima Daiichi nuclear power plant accident (2011), engineers and scientists are working hard to find better concepts for future new nuclear power. At least six new nuclear power plants were proposed by the Generation IV International Forum (GIF) in 2002. One of the six proposed designs is a High-Temperature Gas-cooled Reactor (HTGR) that uses a prismatic block type and spherical pebble-type fuel form (4).

For those two types of fuel, they use TRISO-coated particles. The coated particles should be designed and fabricated to remain intact and retained when the reactor is operated in normal conditions or even under accident conditions. To meet these requirements, the TRISO particles consist of microspherical fuel coated with several layers.

When a reactor is under operation, there will be many fission products such as isotopes, dislocation of lattice due to fission product recoil, swelling of the kernel and oxygen gas production from the UO_2 fission process. High energy neutrons are also released during the UO_2 fission reaction and come out from the fuel to enter its surroundings like pyrocarbon and zirconium carbide layers. Through inelastic scattering collision, some of the fast neutron energy is given

to those layers so that the layers can shrinkage and/or expand. Noble gas is also released from the nuclear fission reaction as the fission product. Studying the fission products is not simple work because they are generated in large numbers with the distribution. The generated fission products depend on many parameters, such as the neuron energy, the fissile nuclide, the temperature, the burnups, etc. The fission products are categorized into four groups, namely fission gases and other volatile fission products, fission products forming metallic precipitates, fission products forming oxide precipitates, and fission products dissolved as oxides in the fuel matrix based on the elemental analyses on irradiated fuels of LWR, FBR and HTR power reactors (5,6). The chemical state of fission products depends on several factors above, making this issue complicated due to the variation in fission products generated by fuel reactions during nuclear power generation. An independent study must be carried out on the effects of fission products. Particularly for HTGR fuels, TRISO. The fuel is irradiated at high temperatures, higher than in the case of the light water reactors LWR and the fast breeder reactors FBRI.

The behaviour of the reaction between fission product and palladium has been studied. The fission product palladium reacting with the SiC layer and the reaction behavior of the fission product with silicon migrating from the carbon monoxide-corroded SiC layer and the interaction of the fission products other than palladium with SiC. Another report said that the fission product was found at dislocation cores and grain boundaries, with local corrosion and dissociation of C from Pd containing grain boundary agglomerations (7).

In the current TRISO design, the kernel is surrounded by a buffer and SiC layer between inner pyrolytic carbon (IPyC) and outer pyrolytic carbon (OPy) in ∼35 μm dimension (8). This layer should be high density as a barrier for fission products. Study about the ion range of the fission product is important to make sure the fission product cannot be released from the barrier.

ZrC is considered as an alternative to replace the SiC layer due to excellent physical and nuclear properties such as high corrosion-resistant, excellent thermal shock resistance and low neutron capture cross-section (9). It is expected that ZrC will be a better barrier than SiC against the diffusion attack of Ag and Pd. ZrC has been suggested to be the main barrier for the containment of fission products by replacing the SiC layer or as an alternative or as an additional layer of the TRISO fuel particle to overcome the corrosion issue of SiC because of interaction with the fission product of silver (Ag) and palladium (Pd).

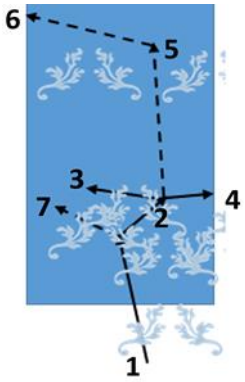
In this study, the penetration depth from the ion range of those two fission products, Ag and Pd in the ZrC layer was studied and simulated with various kinetic energies using SRIM (Stopping and Range of Ions in Matter) /TRIM (Transport of Ions in Matter) computer code with Monte Carlo method. By applying various kinetic energies of Ag and Pd that will bombard the ZrC layer with 1000A thickness, the ion range and depth penetration will be calculated

EXPERIMENTAL SECTION

SRIM is used as a program to simulate the interactions of ions that relate to Coulomb contact with target atoms and the exchange and correlation between electron shells which overlap. Based on these phenomena, the stopping and distance of ions to matter is determined. Unlike SRIM, TRIM based on the Monte Carlo method, calculates the ion's interactions in the target. Monte Carlo is a method for simulating via computer codes using deterministic methods, which the theoretical process of complex problems cannot model. Each probabilistic event that forms a process shall be simulated in sequential order. To describe the real phenomenon, the probability distribution of the event is statistically sampled.

Random number selection shall be taken into account in the statistical sampling process. It follows each of many particles from a source throughout its life to its death, whether absorbed or escapes from a system, as shown in Figure 1. The figure explains the probability of neutrons entering the material and the life of the neutron in the material is traced. For the purpose of determining the result in each stage of life, probability distribution shall be randomized by means of transport data.

In this research, the stopping and range of silver and palladium ions with several different energies into the ZrC layer are calculated using statistical algorithms and averaging results of the ion-atom collisions. The thickness of the ZrC layer of a TRISO is 41.1 micrometres. The energy of ions varies from 0.1 to 150 MeV as fission products have a maximum kinetic power of about 165 MeV (11). The ZrC density is 6.73 g/cm^3 (12).



1. Incoming neutron
2. Fission, photon production
3. Neutron capture
4. Neutron leakage
5. Photon scatter
6. Photon leakage
7. Photon capture

Figure 1. History of a neutron entering a material (10).

Table 1. Penetration Depth of Pd ion through ZrC layer

No.	Palladium (Pd) Ion kinetic energy (MeV)	Penetration Depth through ZrC (A)
1.	0.10	286
2.	0.15	395
3.	0.25	618
4.	0.50	1,194
5.	1.00	2,341
6.	2.50	5847
7.	5.00	11,200
8.	10.00	19,500

The table shows the TRIM simulation input. We put ZrC as layer with 1000A, atomic stoichiometry 1:1 with displacement damage 28 eV for Zr and 28 eV for C. ZrC density is 6.73 g/cm^3. Ag and Pd are put as elements with the maximum number of ions 4999, which will bombard the layer with kinetic energy 0.1, 0.15, 0.25, 0.5, 1, 2,5 and 5 MeV.

RESULTS AND DISCUSSION

TRIM simulations were done with several Pd ion kinetic energies, namely 10; 5;2.5;1; 0.5;0.25; 0.15 and 0.1 MeV. Through the ZrC layer, the depth of the Pd ion is penetrated with various kinetic energy, as shown in Table 1.

It is shown in Table 1 that the larger the Pd ion kinetic energy, the deeper the Pd ion is through the ZrC layer. It is illustrated in Figure 4.a and b, the output of SRIM/TRIM ion ranges of Pd and Ag after entering the ZrC layer with 1 MeV kinetic. From their atomic radius, Pd has a bigger atomic radius (2.6132 A) than Ag (2.5199 A). However, Pd ion has a bigger ion range (2,341 A) than Ag ion (2,183 A). SRIM/TRIM theoretically duplicates a statistical process using the Monte Carlo method. For complicated problems, which cannot be modelled by computer codes using determinate methods, it is especially helpful. It consists of following each of many particles from a source throughout its life to its death (Figure 1), whether the particles are absorbed or escape from a system. To determine the result at every stage of its life, probability distributions shall be randomly sampled using transport data. Here is any quantity C in equation (1):

$$C = \int \psi(\mathrm{E})\mathrm{f}(\mathrm{E})\mathrm{dEC} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (1)$$

where ψ(E) is the energy-dependent fluence, and f(E) is any product or summation of the quantities in the cross-section libraries or a response function provided by the user (13). Based on equation (1), it can be drawn the understanding that the distance travelled by ions in a material depends on its kinetic energy and the cross-section of the interaction between ions and atoms in ZrC material, and it is not based on the size of the atomic radius.

Figure 5 illustrates the cross-section of ZrC TRISO-coated fuel. Pd and Ag fission products are formed in the kernel and move out through the porous carbon buffer layer and inner pyrolitic carbon and attempt to penetrate the ZrC confinement layer. The interaction of Pd fission products with ZrC's confinement layer has two possibilities: crystal damage and chemical reaction.

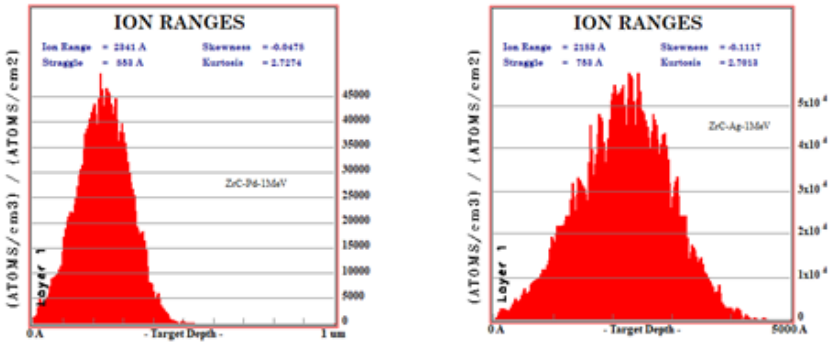


Figure 4. Ion range of 1 MeV kinetic energy (a). Pd (b). Ag in ZrC.

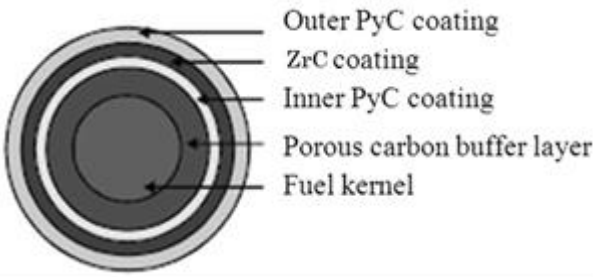


Figure 5. TRISO fuel cross-section.

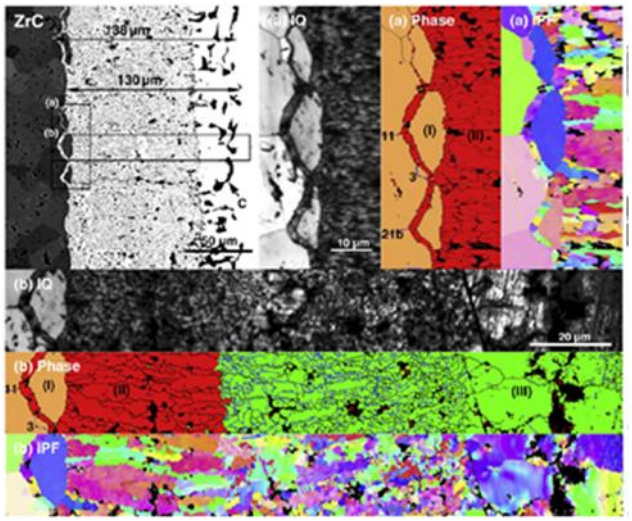


Figure 6. ZrC, Pd_3Zr and Pd(Zr) phase zones (14).

Crystal damage occurs due to collisions of atoms and transfers energy so that the Zr or C atoms move from their positions and form void crystal defects. The chemical reaction between Pd and ZrC can be illustrated in chemical reaction equation (2) :

$$3Pd + ZrC \rightarrow Pd_3Zr + C \quad \text{.....(2)}$$

The interaction between Pd fission products and the ZrC layer during their transport from the inner side of the ZrC layer results in ZrC damage. From the result of Tan et al. experiments, there are three phase zones: Pd (Zr), Pd_3Zr and ZrC (10). These three phase zones are shown in Figure 6.

Table 2. Penetration Depth of Ag ion with various kinetic energy through ZrC layer

No.	Silver (Ag) Ion kinetic energy (MeV)	Penetration Depth through ZrC (A)
1.	0.10	269
2.	0.15	381
3.	0.25	585
4.	0.50	1,115
5.	1.00	2,183
6.	2.50	5,663
7.	5.00	11,500

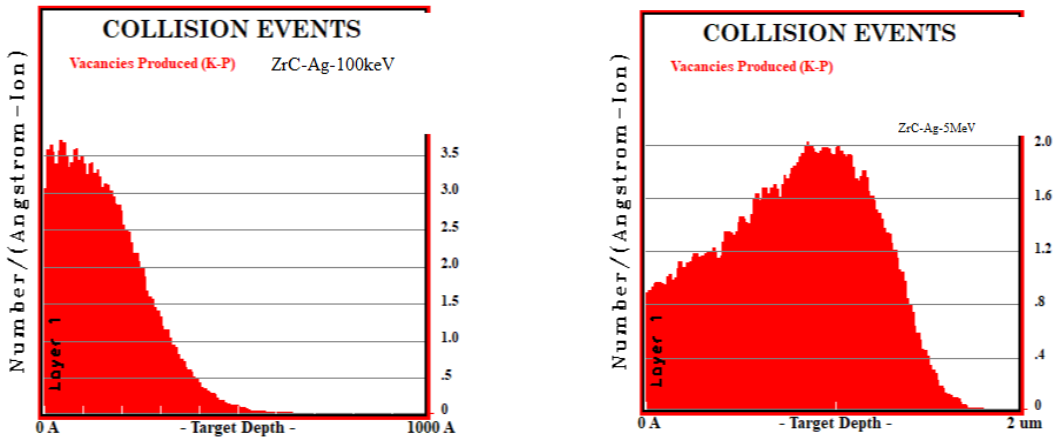


Figure 7. Damage/collision events when 100 keV and 5 MeV Ag enter into ZrC.

Similar to Pd fission products, Ag fission products move from the inner side of TRISO fuel to the outer ZrC confinement layer. Based on the implantation simulation, the Ag moves into the ZrC layer with different depths, as illustrated in Table 2. The results show that the bigger the Ag ion kinetic energy, the deeper the Ag ion moves into the ZrC layer.

It has been reported (14-17) that the most releasable radionuclides from SiC-TRISO fuel coating is radionuclide. Some publications on palladium (Pd) transport in ZrC have been made (18). However, there are still very few publications about Ag released from ZrC TRISO fuel coating, even though it is still challenging to find. The results of Ag ion implantation on ZrC with various kinetic energies are shown in Table 2.

As we know, the fission products have kinetic energy that leaves out the fission reaction point from the kernel, passes through the buffer, inner pyrolytic carbon (IPyC) and enters the ZrC layer. The energetic silver ion interacts with the atom of ZrC molecules through the elastic and inelastic scattering. The movement of the Ag ion stops after all of its kinetic energy has been given to the atoms of the ZrC molecule or has reached equilibrium with the energy of its neighbouring atoms. The interaction between Ag ions with Zr and C atoms in the ZrC molecule will cause damage or not depending on the Ag ions kinetic energy. Figure 7 shows two examples of damage/collision events when specific energy enters into ZrC layer. It is shown that the probability of ZrC layer damage becomes greater depending on the Ag ion's kinetic energy.

The atoms in bulk ZrC get the kinetic energy from Pd or Ag ions when they enter the ZrC layer. Regarding on the quantity of energy ions present, an energy transfer process can take place as follows: Nuclear, electrical or radiation stops. The ZrC layer in the path of ion movements will be damaged until it has stopped (19). The 3 main factors which lead to the severe damage in ZrCs are total displacement, total vacancies created and replacement collisions.

CONCLUSION

The penetration depth of Pd and Ag on the ZrC layer has been investigated using SRIM/TRIM simulation method. From the simulation in 0.10-5 MeV kinetic energy, the bigger energy kinetic of Pd and Ag, the deeper ion of these penetrated the ZrC layer. Ion ranges of Pd and Ag after

entering the ZrC layer with the 1 MeV kinetic energy show that Pd ion has a bigger ion range (2,341 A) than Ag ion (2,183 A), even though Pd has a bigger atomic radius (2.6132 A) than Ag (2.5199 A). Basically, the Pd ions will generate kinetic energy by binding to bulk ZrC atoms when they penetrate a layer of ZrC. Depending on the amount of energy consumed by ions, an energy transfer process may be nuclear, electronic or radiation stop. Damage occurs within the ZrC layer as well as on the ion's movement until it stops. Three main factors that cause a large amount of damage in ZrC are total displacement, total vacancies induced and replacement collisions.

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