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ORIGINAL ARTICLE

Simulation of Ag and Pd Fission Product Implantation in SiC layer of TRISO Fuel Particle of HTGR using SRIM/TRIM Monte Carlo Computer

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ABSTRACT – Silicon Carbide (SiC) has excellent characteristics such as wide band gap, high electron mobility, high thermal conductivity, and radiation effects resistance. Therefore, SiC is widely used for various applications, including nuclear fuel systems. SiC is used in TRISO (Tri-Structural Isotropic) coated fuel particle in HTGR (High Temperature Gas cooled Reactor). TRISO, which consists of Inner Pyrolitic Carbon, SiC, and Outer Pyrolitic Carbon, is one of the safety systems features of the reactor. However, one of the issues of the system is corrosion of SiC caused by silver (Ag) and palladium (Pd). Nevertheless, the detailed mechanism of this corrosion phenomenon, such as the existence of Ag and Pd and how deep those two fission products penetrate the SiC layer, are still unknown. This study aims to investigate the physical interaction of Ag and Pd with the SiC coating layer of TRISO nuclear fuel particles. For this purpose, the physical effect of the penetration of the energetic Pd and Ag fission products into the SiC layer has been simulated using SRIM (Stopping and Range of Ions in Matter) /TRIM (TRansport of Ions in Matter) computer code with Monte Carlo method. Various Ag and Pd ion kinetic energies have been employed in this simulation. The results showed the Ag/SiC and Pd/SiC Ion Ranges, Doses, and Damage as the first-step evaluation to understand the corrosion phenomenon of the SiC-layer in the TRISO particles of HTGR.

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INTRODUCTION

The twentieth century is the time when the use of fossil fuels increases rapidly, and it is about a twenty-fold increase. By the end of this century, the energy demand may reach 50 TWh [1]. It is estimated that fossil fuels contribute approximately 80% of total energy demand [2], while nuclear energy contributes about 14% of total energy demand [3]. The utilization of fossil energy produces carbon dioxide (CO_2) gas which absorbs infra-red radiation from the earth's surface so that the heat energy cannot be released into the atmosphere. It causes an increase in the earth's temperature, called global warming [3].

Nuclear power is one of the ways to cut carbon dioxide emissions. More than 40 countries (including France, Argentina, Brazil, Canada, Japan, the Republics of Korea, South Africa, the United States, the United Kingdom, Russia, and China) have approached UN officials to express interest in launching nuclear power programs.

They set up the Generation IV International Forum (GIF) to oversee the development of simpler technology reactors with improved safety and physical protection, as well as reactors that are more affordable to design, operate, and maintain, and that contribute to a viable energy source with more long-term commercialization [4].

In 2002, GIF selected six innovative nuclear systems after evaluating more than 100 different designs by more than 100 experts from 12 countries worldwide. These six systems include the following [5]:

- sodium-cooled fast reactor (SFR) system
- lead-cooled fast reactor (LFR) system
- gas-cooled fast reactor (GFR) system
- molten salt reactor (MSR) system
- very high temperature reactor (VHTR) system
- supercritical water-cooled reactor (SCWR) system

Among those six nuclear power reactors selected by GIF, HTGR (High Temperature Gas-cooled Reactor), similar to the very high temperature reactor (VHTR), has become popular globally. Indonesia will also adopt this power reactor through its experimental power reactor (RDE) program, introduced-at the beginning of 2014 [6]. The thermal power of RDE is 10 MW.

HTGR has good features such as high thermal efficiency, economic competitiveness, and more reliable safety and proliferation resistance. There are two types of fuel form: prismatic block type and spherical pebble type fuel (Figure 1). The first type of fuel has been used in the design of HTGRs in the United States and is also being used in the prototype of Japan's High Temperature Test Reactor (HTTR) [7], [8].

The second spherical pebble type fuel was used for the AVR reactor built in Juelich, Germany, and operated during 1967- 1988 [9]. Both types of fuel form are composed of Tri-structural-isotopic (TRISO) fuel particles.

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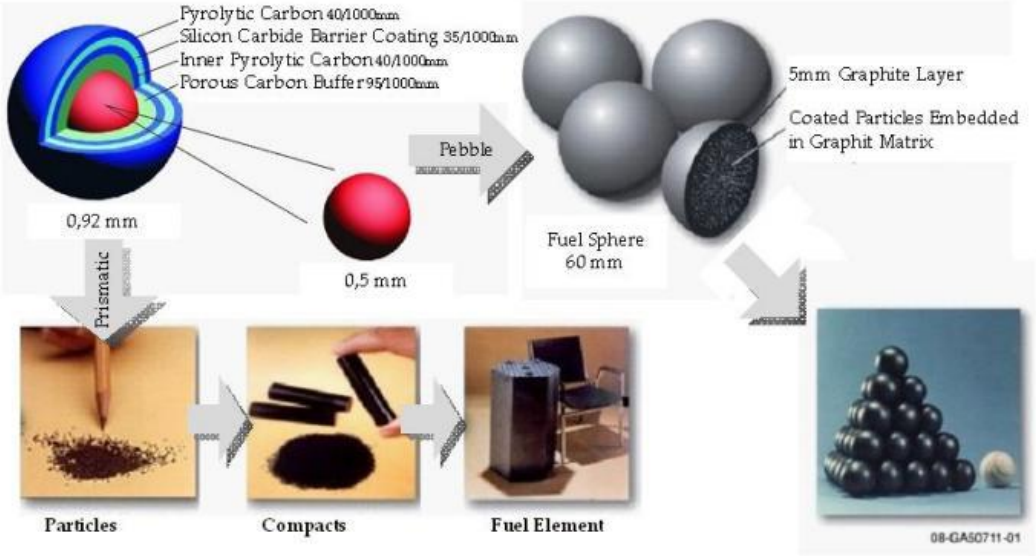


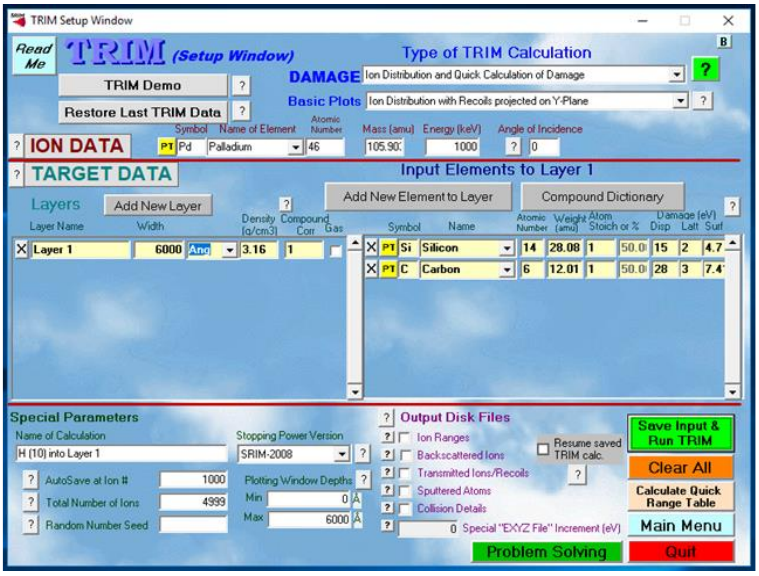
Figure 1. TRISO fuel particles for prismatic block-type and pebble bed-type cores [9]

TRISO Particles are designed for high temperature reactors so that they are expected to have good performance during the reactor being operated. The damage to TRISO particles can be caused by external and internal factors or either one-dimensional (1D) or three-dimensional (3D) effects. Pressure vessel failure causes the 1D effect, while shrinkage fractures within the IPyC layer, IPyC/SiC debonding, particle asphericity, kernel migration (the "amoeba effect"), and SiC coating thinning cause the 3D effect [10]. In this paper, Ag and Pd Fission Product Implantation on SiC layer in TRISO Fuel Particle of HTGR will be studied using SRIM/TRIM 2012 Monte Carlo Computer Code.

EXPERIMENTAL METHOD

SRIM is a program that simulates the interaction of ions in the matter based on Monte Carlo Method, and it contains an additional program called TRIM (the Transport of Ions in Matter). TRIM is a Monte-Carlo program that calculates the interaction of ions within a target. It establishes the collision details between ions and target atoms by inputting a random number for the impact parameter and distance to the next colliding atom to create a probability distribution dependent on the target density. There are three steps required in the simulation process: sampling on random input variables X, evaluating model output Y, and statistical analysis on model output.

For these simulations, two elements, Si and C were used with a ratio of 1:1. The simulation SRIM is used to predict the movement of the fission products Ag or Pd phenomena together with TRIM Calculation of Full Damage Cascade. This simulation can demonstrate the entire penetration process, recoil cascades, and target atom mixing [10]. The calculations were started one ion at a time to make accurate analyses of the physics of each and every interactio between



DAMAGE: Ion Distribution and Quick Calculation of Damage
Basic Plots: Ion Distribution with Recoils projected on Y-Plane

Symbol	Name of Element	Atomic Number	Mass (amu)	Energy (keV)	Angle of Incidence
Pd	Palladium	46	105.90	1000	0

Layer Name	Width		Density (g/cm3)	Compound Corr	Gas
Layer 1	6000	Ang	3.16	1	☐

Symbol	Name	Atomic Number	Weight (amu)	Atom Stoich	or %	Damage (eV) Disp	Latt	Surf
Si	Silicon	14	28.08	1	50.0	15	2	4.7
C	Carbon	6	12.01	1	50.0	28	3	7.4

Name of Calculation: H (10) into Layer 1
Stopping Power Version: SRIM-2008
AutoSave at Ion #: 1000
Total Number of Ions: 4999
Random Number Seed:
Plotting Window Depths: Min 0 Å, Max 6000 Å
☐ Ion Ranges
☐ Backscattered Ions
☐ Transmitted Ions/Recoils
☐ Sputtered Atoms
☐ Collision Details
0 Special "EXYZ File" Increment (eV)
☐ Resume saved TRIM calc.

Figure 2. SRIM/TRIM input

the ion and the target atom. The computation process can continue even if it is halted for a short time, and the output results can be saved and used later. The running time varies depending on the type of ion, which might range from a few seconds to several minutes. Averaging across a large number of simulated particle trajectories yielded the results [3]. In this study, the energies for the incident ions are varied from 100 keV up to about 5 MeV, and the angle of incidence of the ions is 00. Plots showing ion trajectories, depth vs. Y-Axis, transverse view, ionization, phonons, collision events, atom distributions, and energy to recoil may be generated by simulating comprehensive calculations with full damage cascades [11]. The TRIM simulation input for a 1 MeV fission product (Ag or Pd) through the SiC target is shown in Figure 2.

RESULT AND DISCUSSION

TRISO fuel is coated by four layers: buffer, IPyC, SiC, and OPyC layer, as shown in Figure 1 and its cross-section is illustrated in Figure 3. Very high temperature reactor (VHTR)/High Temperature Gas Cooled Reactor (HTGR) is designed to have an outlet temperature between 800–1000ºC and operated by using TRISO fuel in the form whether prismatic block type or spherical pebble type fuel [7], [11], [12].

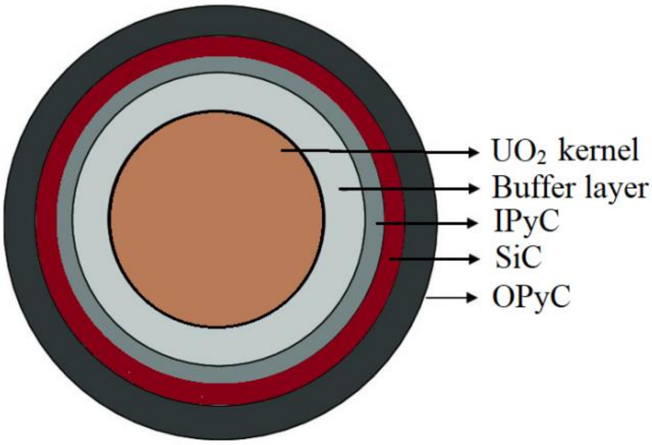


Figure 3. TRISO fuel cross-section

When VHTR/HTGR reactor is in operation, it results in more than 20 fission products [4], and among them are Pd and Ag. ^{110m}Ag is a radionuclide that emits gamma-rays with a half-life of 250.4 days so that it must be retained so as not to come out of TRISO both the reactor in regular operation or an emergency. SiC confinement layer is responsible for confining Ag so that it does not come out of TRISO. Fission products have kinetic energy, so they will move from the point of fission reaction where the resulting fission strives to penetrate the barrier layer. The schematic transport mechanism of both Pd and Ag is illustrated in Figure 4.

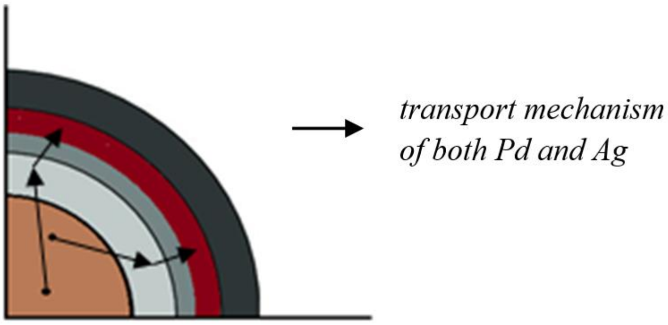


Figure 4. Pd and Ag transport in SiC TRISO coated fuel

When an atom collides with other atoms, it can result in the loss of an electron from the atom and it is called atom ionization. Metallic fission products such as Pd and Ag move from the point of fission reaction and they are ionized by their neighboring atoms. The Pd and Ag ions then proceed to the buffer layer, the inner pyrolytic carbon and the SiC confinement layer.

The transport mechanism of Pd and Ag ions is simulated using SRIM/TRIM code. SRIM/TRIM ion implantation simulation running window when the starting kinetic energy of Pd fission product is 1 MeV is illustrated in Figure 5.

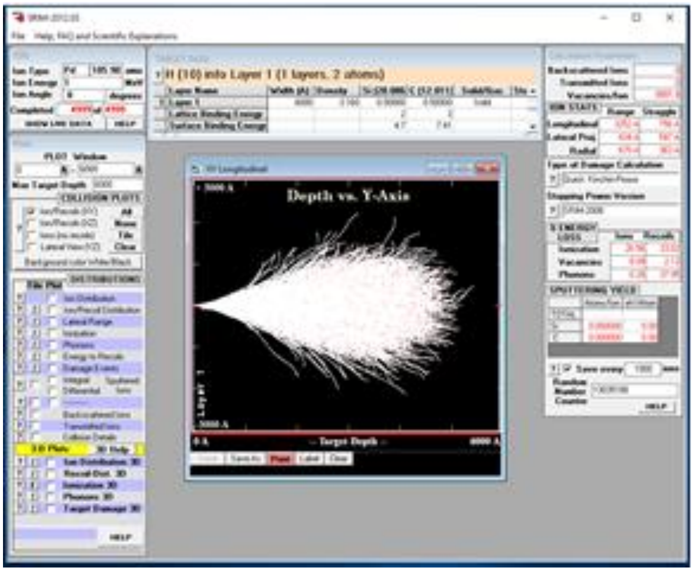


Figure 5. SRIM/TRIm running window

The simulation results can be obtained by clicking the running window file plots. There are at least eight output file plots. As the illustration, the output file plots are depicted in Figure 6, namely ion depth penetration, ion ranges, energy to recoils, target ionization, target phonons in 2D, damage/collision events, total displacement in 3D, and target phonons in 3D.

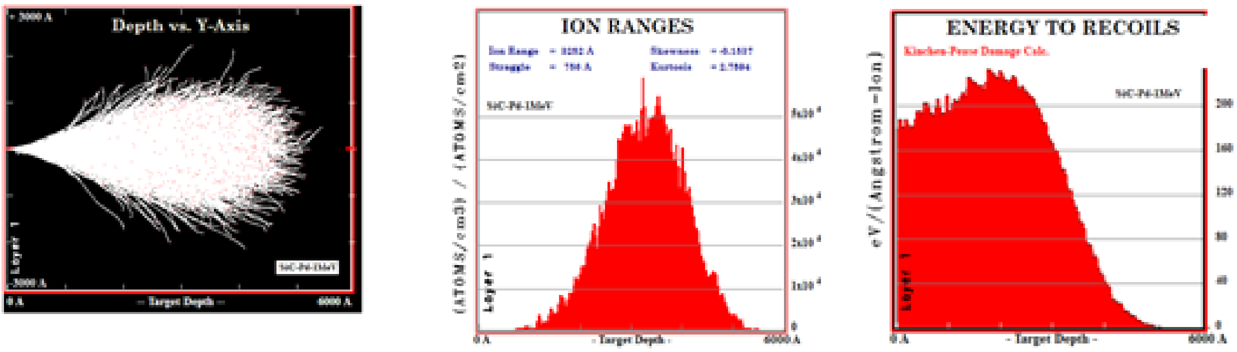
From Figure 6, many phenomena explain what happens when Pd fission product enters SiC layer. Figure 6 (a) and (b) provide information about the path of each ion during its journey until it stops after losing all of its kinetic energy and the average trajectory length or the penetration depth in the SiC layer. For Pd ion with the starting kinetic energy of 1 MeV, the ion range is 3252 A. The penetration depth of Pd ion with different starting kinetic energies is shown in Table 1. This plot and file tabulate the direct energy loss by the ion to the various target atoms. This energy loss, plus the direct energy loss of the ion to the target electrons, sum to the energy loss of the ion into the target. From Table 1, it is shown that the bigger the starting kinetic energy of Pd ion, the longer its trajectory length.

Based on the kinetic energy of the ions, the physical interactions that occur are divided into three groups: moderate low energy (below 100 keV), moderate energy (above 100 keV–about 1 MeV), and high energy above 1 MeV. The interaction mainly occurs in the low-energy range is the absorption of ion by the target surface. Therefore, diffusion and desorption occur. As a result, the ion's kinetic energy is transferred to the lattice atoms of the target and induces oscillations of the atoms that can be interpreted as thermal energy. The part volume of the target can be heated by the thermal energy locally up to several thousand Kelvin in a short time, and it can lead to local lattice destruction or local sample melting. The ions are fully stopped when all of their kinetic energy is transferred to the lattice atoms during their interaction. This process is called as the nuclear stopping ions due to the deceleration of the ion process by Coulomb interaction and the process of elastic collision with lattice atoms. The elastic scattering of atoms by an energetic ion results in collision cascades that can affect or damage the material's structure [13].

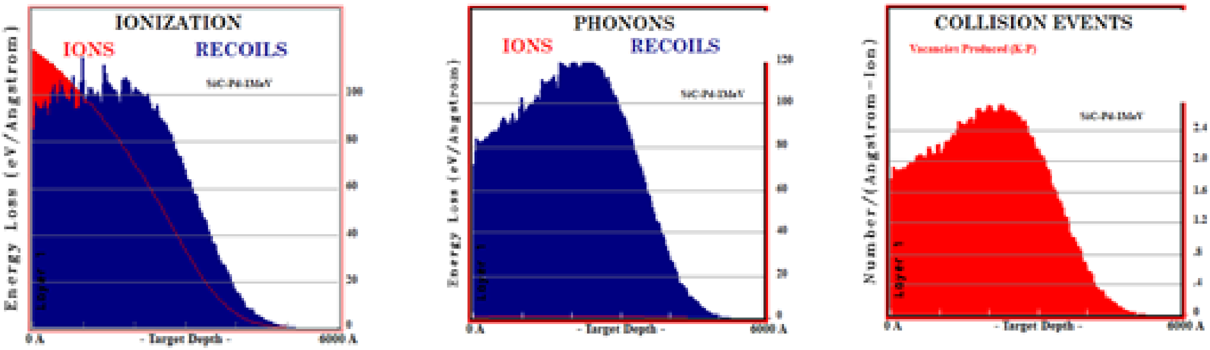
If the ion kinetic energy is in the moderate energy range (100 keV <E_k < 1 MeV), the interaction occurs between the ion and the electron of the target atoms. This process is dominant when the kinetic energy of the ions increases to the order of MeV, and the process is called electronic stopping. High electronic stopping is found to recrystallize irradiation damaged samples and observed in SiC [13], [14]. In insulators, high electronic energy transfer frequently plays a dominant role, leading to structural defects, radical formation, bond breakage, and the formation of novel bonds [15].

Energy loss by radiation emission occurs when the energy of ion is above several MeV. The energy loss from these ions is in radiation emission and is called radiation stopping or bremsstrahlung. Another effect is the emission of Čerenkov radiation which occurs for faster-charged particles than the phase velocity of electromagnetic waves. Moreover, at very high energies nuclear reactions or decays can appear.

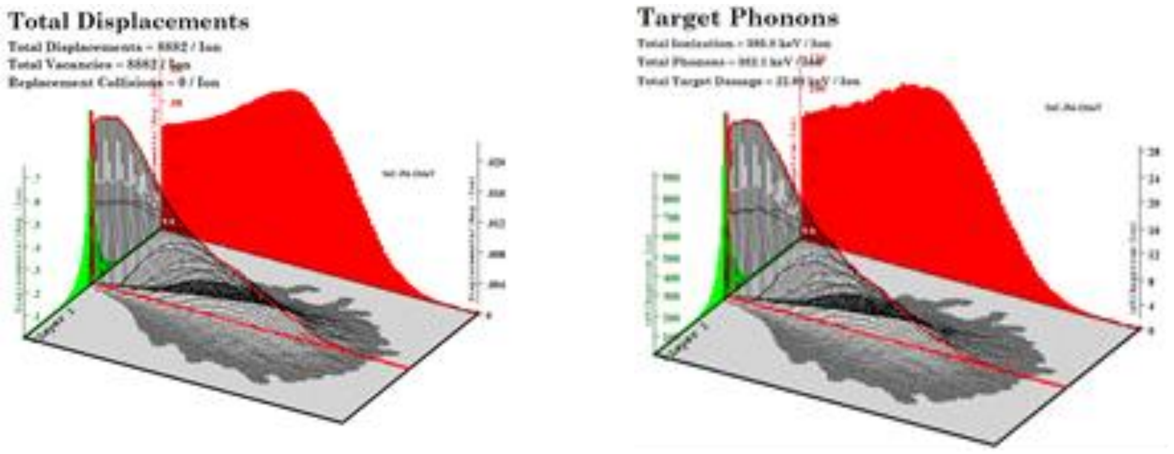
In short, when Pd ions enter the SiC layer, they will give their kinetic energy to the atoms in bulk SiC. The energy transfer process can be nuclear stopping, electronic stopping, or radiation stop depending on the amount of energy possessed by ions. Damage occurs in the SiC layer along the path of the ion movement until the ion stops.



(a) SiC-Pd-1MeV-Depth trajectory (b) SiC-Pd-1MeV-Ion Ranges (c) Energy to Recoils



(d) Ionization (e) 2-D Target phonon (f) Damage/collision events



(g) Total displacement (h) Target phonons

Figure 6. Output file plots from running window at 1 MeV of Pd fission product

Table 1. Penetration Depth of Pd ion with various kinetic energy through ZrC layer

No.	Palladium (Pd) Ion kinetic energy (MeV)	Penetration Depth through SiC (A)
1.	0.10	400
2.	0.15	555
3.	0.25	868
4.	0.50	1,658
5.	1.00	3,252
6.	2.50	8,104
7.	5.00	15,200

Similarly, the three energy transfer processes described above also occur by using the same SRIM/TRIM when Ag ions move into the SiC layer. The depth range varies depending on the initial energy of the Ag ion. The degree of damage from the SiC layer depends on the amount of kinetic energy from the Ag ion. In general, damage occurs along the path from the Ag ion until the ion stops. The simulation results of the calculation of the depth range of Ag ions in the SiC layer as a function of the kinetic energy of the Ag ions can be shown in Table 2. It appears that the higher the kinetic energy of the Ag ions, the longer the ion's depth range.

Figure 6(c) shows information about energy given by ions to the target atom to make recoil cascades. An ion that hits an atom of the target gives some of its energy called recoil energy~~.~~ The recoil energy is divided into displacement collisions, vacancy production, replacement collisions and interstitial atoms. Figure 6(d) illustrates both energy ions and recoils of target atoms as the results of the process of collision between the implant ions and target atoms. Some energy of the implant ions that is given to the target atoms results in the phonon production as shown in Figure 6(e). Another result of TRIM simulation using Ion Distribution and Quick Calculation of Damage menu is damage/collision events. The damage calculated with this option will be the quick statistical estimates based on the Kinchin-Pease formalism. The damage event results due to the entering of Pd ions with the kinetic energy of 1MeV are shown in Figure 6(f). Figure 6(g) and (h) show the total displacement of the target atoms and phonons production in 3D.

Table 2. Penetration Depth of Ag ion with various kinetic energies through SiC layer

No.	Silver (Ag) Ion kinetic energy (MeV)	Penetration Depth through SiC (A)
1.	0.10	379
2.	0.15	519
3.	0.25	804
4.	0.50	1,511
5.	1.00	2,958
6.	2.50	7,746
7.	5.00	15,500

Table 2 shows the penetration depth of Ag ion with various energies through SiC layer. The bigger the starting kinetic energy of Ag ion is, the longer the penetration depth becomes and it means that more damage in SiC layer.

CONCLUSION

Implantation of Pd and Ag in SiC layer has been simulated using SRIM/TRIM software. The kinetic energy of Pd and Ag used in this simulation was in the range of 0.1 to 5.0 MeV. The depth trajectories of Ag and Pd fission products, energy to recoils, ionization recoils, phonon recoils, total atom displacements were obtained from the simulation. From these data, the SiC defects due to the implantation of Ag and Pd fission product can be predicted.

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