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Characterization of neutron induced damage effect in several types of metallic multilayer nanocomposites based on Monte Carlo simulation

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ABSTRACT

Metallic multilayer nanocomposites are known to have excellent interface self-healing performance when it comes to repairing irradiation damages, thus showing promise as structural materials for advanced nuclear power systems. The present study investigated the neutron irradiation displacement damage rate, spectra of the primary knocked-on atoms (PKAs) produced in the cascade collision, and the H/He ratio in four kinds of metallic multilayer nanocomposites (Cu/Nb, Ag/V, Fe/W, and Ti/Ta) versus neutrons' energy. Results suggest that the three neutron induced damage effects in all multilayer systems increased with the increasing of incident neutrons' energy. For fission reactor environment (1 MeV), multilayer's displacement damage rate is 5–10 × 10^{22} dpa/(n/cm^2) and the mean PKAs energy is about 16 keV, without any noteworthy H/He produced. Fe/W multilayer seems very suitable among these four systems. For fusion reactor environment (14 MeV), the dominant damage effect varies in different multilayer systems. Fe/W multilayer has the lowest displacement damage under the same neutron flux but its gaseous transmutation production is the highest. Considering the displacement damage and transmutation, the irradiation resistance of Ag/V and Ti/Ta systems seems much greater than those of the other two.

1. Introduction

Metallic multilayer nanocomposites have received significant attention in recent years for their excellent self-healing properties in trapping and releasing defects [1–6]. The microstructure of multilayers with small individual layer thickness of nanometer order size is proved to be much more stable than bulk materials after ion implantation [7]. Molecular dynamics (MD) simulations demonstrate that interfaces between the neighboring heterogeneous layers of the multilayer nanocomposites act as efficient sinks for the interstitial atoms and can immediately emit them to nearby vacancies. Through the "absorb and re-emit" mechanism, the velocities of defect recombination in multilayers become more rapid [8]. Recently, Misra et al. developed a novel approach to process such nanocomposites in bulk form by fabricating a 4 mm-thick Cu/Nb multilayer nanocomposite with crystallographically stable interfaces via accumulative roll bonding technique [9,10]. As a result of this remarkable breakthrough in nanocomposite manufacturing techniques, bulk nanocomposites have become promising candidates as structural materials for advanced nuclear power systems.

Since most investigation focus on the interface self-healing performance of multilayer nanocomposites, however, its radiation induced damage effects in reactor radiation environment are seldom concerned. Just as the traditional block materials, metallic multilayers' radiation damage effect varies depend on the material system and the radiation condition. For instance, nickel alloy has a strong anti-displacement damage feature but its gaseous transmutation production problem in fusion reactor environment is serious. Hence, knowledge of the radiation-induced displacement damage rate, energy transfer between the incident neutron and primary knock on atoms (PKAs), and the gaseous transmutation production under neutron irradiation with different energy is of primary importance in achieving the goal of designing radiation-tolerant materials. The current paper presents the neutron irradiation damage effects on four kinds of metallic multilayer nanocomposites (Cu/Nb, Ag/V, Fe/W, Ti/Ta), which have been tested using the Monte Carlo method toolkit MCNP and Geant4. These four kinds of multilayers have good interface self-healing properties [11–14].

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2. Methodology

2.1. Molding of metallic multilayers

In our work, we set the multilayer nanocomposites as a 200 μm × 200 μm × 100 μm box. The box is composed of 10,000 single layers (10 nm thick) and the material in each layer varies alternately between two metallic elements, such as Cu/Nb, Ag/V, Fe/W and Ti/Ta. The mono-energetic neutron source is set to locate on the central axis of the model and its incidence direction is parallel to the depth direction of the multilayer. Because we set the material outside the multilayer as vacuum, neutrons don't lose any energy until they are implanted into the multilayer (Fig. 1).

2.2. Calculation method of displacement damage (dpa) using MCNP

Displacement per atom (dpa) is often used to evaluate materials' displacement damage. It can be calculated from the following equations:

$$\text{dpa} = \left(\int \sigma_{\text{dis}}(E)\cdot\varphi(E)dE\right)\cdot t \tag{1}$$

$$\sigma_{\text{dis}}(E) = \frac{\beta}{2E_{\text{d}}}\sigma_{\text{damage}}(E) \tag{2}$$

where $\sigma_{\text{dis}}(E)$ is the atomic displacement cross section for neutron at an energy E, $\varphi(E)$ is the incident neutron flux, and t is the irradiation time. $\sigma_{\text{dis}}(E)$ can be calculated from the damage energy cross section $\sigma_{\text{damage}}(E)$. The factor β is a normalized constant (displacement efficiency) with the value of 0.8. E_{d} is a constant, which refers to the displacement threshold energy. Values of E_{d} in different metal materials used in the simulation are listed in Table 1 [15].

In this work, we performed the dpa calculations using the Monte Carlo neutron-transport code MCNP [16]. Unlike the first principle and the molecular dynamics calculation, the effect of materials' crystallinity is not taken into account in the simulation.

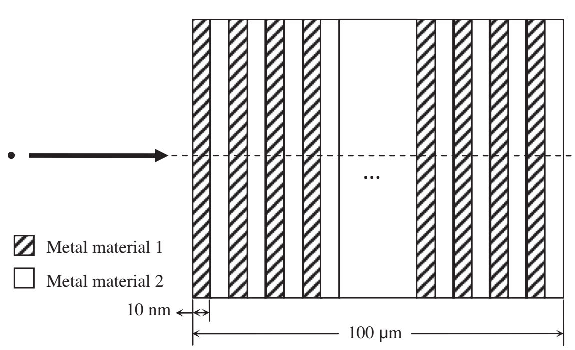

Fig. 1. Geometry of the metallic multilayer nanocomposites model in the Monte Carlo simulations.

Table 1
Displacement threshold energies of elemental metals.

Metal	E_{d}/eV
Cu	18.3
Nb	28.2
Ag	26.0
V	28.0
Fe	17.4
W	44.0
Ti	20.8
Ta	26.7

The neutron flux $\varphi(E)$ was scored by F4 card. $\sigma_{\text{damage}}(E)$ is the cross section of 444 reaction channel.

2.3. PKA and gaseous transmutation production score using Geant4

Geant4 package is another Monte Carlo toolkit for simulating the transport of particles through matter [17]. In implementing customized complex scoring, we had to handle the information of each step around the particle track, which was unavailable through MCNP. In the Geant4 simulation, we used the physics list QGSP_BERT_HP to define the physical process without any modifications. As reflected in its naming scheme, QGSP_BERT_HP is based on Bertini cascade models and also includes high-precision methods for transporting neutrons with energies below 20 MeV [18]. All particles (neutron, ion, gamma ray, electron, etc.) used in the simulation and their processes are included in it. 1×10^7 events were performed to ensure the accuracy of the calculation results.

3. Results and discussion

3.1. Neutron's energy influence on dpa

Most Gen IV fission reactor and fusion reactor neutrons' energy is in the range of 0.1–14 MeV. At this scale, not only do the neutron flux and irradiation time determine materials' radiation damage, the neutrons' energy also affects it markedly. From the perspective of neutron interaction probability in materials, the interaction cross section of neutron reduces with increasing neutron's energy,

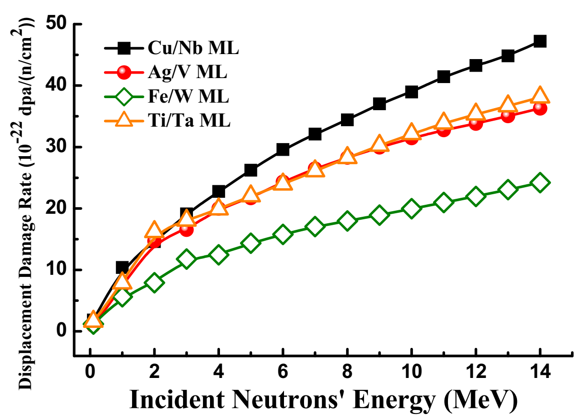

Fig. 2. Neutron induced displacement damage rate in four multilayer systems versus incident neutron's energy.

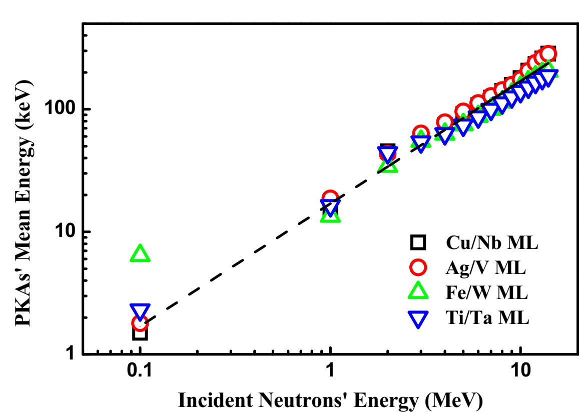

Fig. 3. The mean PKA energies produced by incident neutrons with different energies.

that is, more neutrons move through the material before them interact with any atoms when neutrons' energy rises up. On the other hand, however, the atomic displacement cross section of neutron shows an opposite tendency to the neutron's interaction probability with increasing neutron's energy. Since materials' radiation damage is influenced by the both two aspects, how the neutrons' energy will impact on it is indistinct yet. In the present work, we calculated the radiation damage of several metallic multilayers using the neutron-transport code MCNP.

With neutron's energy moving up, the neutron-induced displacement damage rates of the four multilayer systems are presented in Fig. 2. As seen from all the four curves, increasing

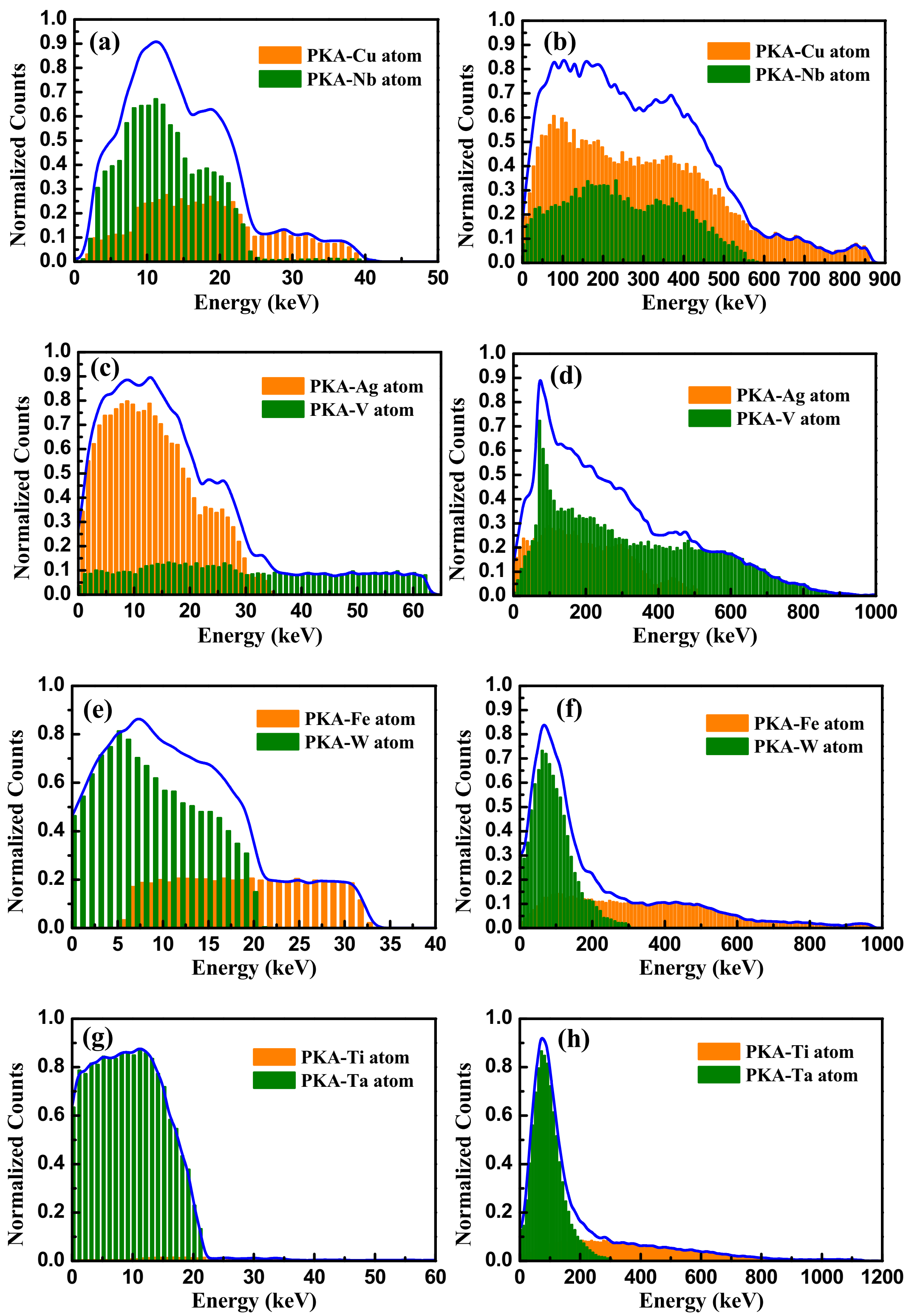

Fig. 4. PKA spectra in four multilayer nanocomposites produced by 1 MeV ((a), (c), (e) and (g)) and 14 MeV ((b), (d), (f) and (h)) neutrons: (a and b) Cu/Nb, (c and d) Ag/V, (e and f) Fe/W, (g and h) Ti/Ta.

neutron's energy increased the displacement damage rate markedly. For Cu/Nb multilayers, the displacement damage rates induced by 1 MeV and 14 MeV were 1.039×10^{-21} and 4.723×10^{-21} dpa/(n/cm^2), respectively. Suppose the neutron flux in Gen IV fission reactor is 10^{15} n/(cm^2 s), if the Cu/Nb multilayers had been used in it, the displacement damage rate can reach 32.7 dpa/y, not to mention the damage rate induced by fusion neutron (14 MeV) is nearly 5 times larger than fission neutron (1 MeV). Besides, comparing these four multilayers, their displacement damage rate was different. The damage rate of Cu/Nb multilayer was the maximum within the whole energy range of 0.1–14 MeV, while the Ag/V, Ta/Ti multilayers' damage rates were approximately the same and slightly less than Cu/Nb multilayer. Among the four, Fe/W multilayer showed the greatest resistance performance to the neutron radiation damage.

3.2. PKA spectra

Although the calculated results of multilayer's displacement damage rate informs materials modelers and experimentalists about the irradiation dose at the atomic level, however, it's not sufficient to characterize the damage effect yet. The majority of radiation damage effects on the materials' microstructure evolution involve a complex cascade collision processes, and these collisions are associated with the PKA spectra [19]. Fig. 3 shows the dependence of PKA's mean energy E_m on the incident neutron's energy using log–log coordinate. E_m increased with the increase of neutron's energy. For different metallic multilayer system, values of E_m didn't appear much difference in the whole energy range (0.1–14 MeV). It was about 16 keV at 1 MeV and 240 keV at 14 MeV. In previous MD studies [20,21], most of the cascade collision simulations of multilayers were performed using PKAs with a few keV energies. However, according to our result, the 1 MeV fission neutron and 14 MeV fusion neutron produce PKAs with 10–200 keV or even larger. Hence, studies on the structural stability of metallic multilayers after cascade collision of PKAs with dozens or hundreds of keV energies are also significant for application in fusion reactors.

Fig. 4 shows the PKA spectra induced by 1 MeV (Fig. 4(a), (c), (e) and (g)) and 14 MeV neutrons (Fig. 4(b), (d), (f) and (h)) in Cu/Nb, Ag/V, Fe/W and Ti/Ta multilayers, respectively. We separate the PKA energies for different elements, bin them and show both separately in these plots. The blue curves represent the total PKA spectra including the two different elements in multilayers.

From the plots of PKA spectra induced by 1 MeV, peaks of the total PKA energies were all around 10 keV. As shown in Fig. 4(a), the PKA yield of Nb was much more than the Cu PKA and all of them concentrated on the scale of 0–25 keV, while Cu PKAs had a wider energy distribution (0–40 keV). In Fig. 4(c) and (e), the same phenomenon that two elements of multilayers have different PKA energy distribution was also observed in Ag/V and Fe/W systems (In Ag/V system, Ag PKAs ranged from 0 to 35 keV, and V PKAs ranged from 0 to 60 keV. In Fe/W system, W PKAs ranged from 0 to 20 keV, and Fe ranged from 5 to 35 keV). For Ti/Ta systems, 1 MeV neutron can hardly produce any Ti PKAs.

For 14 MeV neutron case (Fig. 4(b), (d), (f) and (h)), the Cu, V, Fe and Ti PKAs' energy distribution was much higher than their complementary elements in the multilayer systems, respectively. These comparisons of energy distribution between different elements in each multilayers are consistent with the results observed in plots of 1 MeV case. However, the most PKA yield place had shifted from the Nb, Ag layers to the Cu and V layers in Cu/Nb, Ag/V multilayers when neutron's energy increasing from 1 MeV to 14 MeV. Most notably, nearly half of PKAs' energy was upon 400 keV, the threshold energy that can produce subcascades in materials. Hence, materials' microstructure evolution in fusion reactor environment is supposed to make a big difference to the fission environment case.

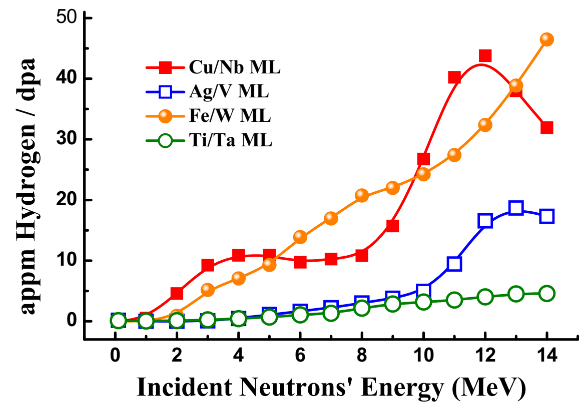

Fig. 5. Hydrogen concentration accumulate induced by different neutron energy.

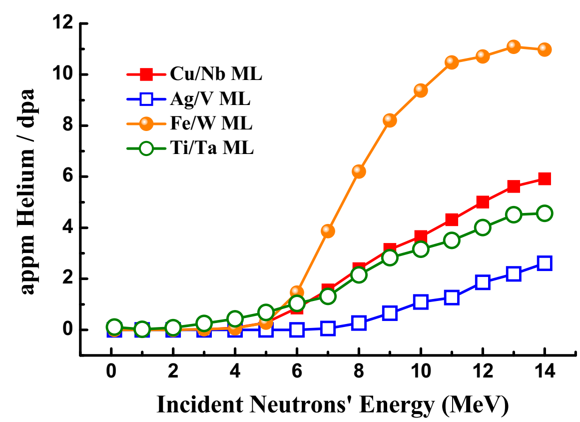

Fig. 6. Helium concentration accumulate induced by different neutron energy.

3.3. H/dpa and He/dpa ratio

Transmutation is another important neutron induced damage effect besides the displacement damage effect. The gaseous transmutation products include hydrogen and helium, which accelerate the materials' embrittlement dramatically. Figs. 5 and 6 presented the hydrogen and helium accumulation rate vs. neutron's energy. The gaseous accumulation rate is very low when the incident neutron's energy is less than 1 MeV. With the increasing of neutron's energy, the accumulation rate became higher and higher. In comparison, the hydrogen accumulation rate is especially significant to the helium. Among the four multilayer systems, the H/He ratio is also different. As shown in these plots, the H/He ratio of Ag/V and Ti/Ta systems was the lowest.

4. Conclusions

In this study, we have compared the displacement damage rate, PKA spectra and gaseous transmutation production of several types of metallic multilayers, induced by different energy neutron. As the incident neutron's energy increases, the three neutron induced damage effects in all multilayers get more excruciating. For fission reactor environment, the most crucial problem is the displacement damage effect. For fusion reactor environment, the dominant damage effect varies in different multilayer systems.

Fe/W multilayer has the lowest displacement damage under the same neutron flux but its gaseous transmutation production is the highest. Considering the displacement damage and transmutation, the irradiation resistance of Ag/V and Ti/Ta systems seem much greater than those of the other two.

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