EXPERIENCE WITH MOVING FROM DPA TO CHANGES IN MATERIAL PROPERTIES*

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Abstract

Atomic displacements by high energy particles induce formation of point defects and defect clusters of vacancies and interstitial atoms in a crystalline solid. The damaged microstructure results in significant changes in materials physical and mechanical properties. Besides displacement damage, nuclear transmutation reactions occur, producing He and H gas atoms that can have pronounced effect on materials performance. Radiation effects in materials have been studied using various irradiation sources, e.g. fission, fusion and spallation neutron sources, high-energy ions and electron beams, etc. With different types of bombarding particles, radiation damage correlation is essential so that radiation effects produced by different irradiation sources can be compared and data can be transferred or extrapolated. The parameter commonly used to correlate displacement damage is the total number of displacements per atom (dpa). Irradiation-induced changes of material properties are measured as a function of dpa. Considering that several aspects of radiation exposure can give rise to property changes, the extent of radiation damage cannot be fully characterized by a single parameter. This paper will discuss damage correlation under various irradiation environments, key irradiation parameters and their effects on irradiation-induced property changes.

INTRODUCTION

Radiation damage is produced by energetic particles, such as neutrons, ions, protons, or electrons, interacting with a crystalline solid. An energetic particle transfers recoil energy to a lattice atom, so-called primary knock-on atom (PKA), and the PKA displaces neighbouring atoms, resulting in an atomic displacement cascade. The displacement threshold is typically about a few tens of electron volts [1]. Atomic displacements by high energy particles induce the formation of point defects and defect clusters of vacancies and interstitial atoms. The displacement cascade event occurs within picoseconds. With time, diffusion processes take place and irradiation-induced defects recombine or cluster to form more stable damage structures, e.g. dislocation loops, dislocation networks, voids, helium bubbles, precipitates, etc. The damaged microstructure results in significant changes in physical and mechanical properties of a material. In addition to the displacement damage, nuclear transmutation reactions occur, producing helium and hydrogen gas atoms and solid impurities. The production of helium and hydrogen can have pronounced effect on materials performance even at low concentrations [2,3].

Radiation damage has been studied using various irradiation sources, e.g. fission neutrons in nuclear reactors (e.g. liquid metal fast reactors, gas-cooled and water-cooled mixed-spectrum reactors), fusion neutrons in a D-T fusion neutron source, spallation neutron sources, ion irradiation with accelerators, and high-energy electron beams, etc. Nuclear fission reactors are by far the most commonly-used irradiation facilities. A number of simulation irradiation techniques have been developed for materials research, particularly when there is lack of prototypic irradiation facilities. For instance, material development for fusion reactors, which currently are still in the development stage, has been made primarily in thermal or fast fission reactors. Fusion reactors have significantly higher neutron energy (14.1 MeV) than fission reactors (< 2 MeV). Radiation effects expected to be produced by intense 14.1 MeV neutrons from a fusion reactor have been simulated with low-energy fission neutrons in existing reactors [4]. Another way to obtain radiation effect information in materials is through the use of accelerators. High energy proton accelerators have been used for irradiation studies of fusion reactor materials [5]. Energetic ions are used to simulate neutron irradiation damage for various other reasons, such as minimization of high residual radioactivity, low-cost, better-controlled irradiation conditions, and declined availability of neutron irradiation sources.

High energy protons produce spallation reactions in the target, leading to high-level radiation damage, a large amount of deposited energy, and production of H and He and other transmutation products. This extremely aggressive irradiation environment poses a significant challenge for the target design of high-energy accelerators. Graphite is a candidate material in a number of target designs [6]. The structural behaviour of graphite, e.g. strength and ductility, dimensional stability, susceptibility to cracking, is a complex function of the source material, manufacturing process, chemical environment, temperature, and irradiation conditions. Although extensive knowledge exists on the irradiation effects in graphite, the assessment of the radiation resistance of the high energy proton beam target (e.g. the Neutrinos at the Main Injector (NuMI) target) is however, difficult, as most of the information available on radiation effects in materials is based on nuclear fission reactor irradiations, while the irradiation conditions in the NuMI facility is considerably different from nuclear reactor irradiations. The potential impact of radiation damage on

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the target material by the high energy proton beam must be properly assessed in the target design, and extreme caution must be taken in transferring nuclear reactor irradiation data to the accelerator target irradiation conditions.

DAMAGE CORRELATION

Displacements Per Atom (DPA)

With various types of bombarding particles, radiation damage correlation is essential so that radiation effects produced by different irradiation sources can be compared and irradiation data can be transferred or extrapolated. The parameter commonly used to correlate the displacement damage in a material is the total number of displacements per atom (dpa). Irradiation-induced changes of material properties are measured as a function of dpa. Dpa is a calculated irradiation exposure unit. The calculation of the dpa values takes into account the irradiation particle type, energy spectrum, irradiation time, etc. It is also a function of the irradiated material. Dpa as a damage-based exposure unit represents the number of primary and second atoms displaced from their normal lattice sites as a result of energetic particle bombardment. It can be calculated using the following equation [7,8]:

$$dpa = \int_0^{t_r} \phi_{tot}(t) \int_0^{\infty} \sigma_d(E) \varphi(E,t) dE dt \quad (1)$$

where $\sigma_d(E)$ is the displacement cross section for an incident particle at an energy E, t is the irradiation time, $\varphi(E,t)$ is the fluence rate spectrum, and $\phi_{tot}(t)$ is the time-dependent fluence rate intensity. In calculating the displacement damage in a material, the primary recoil energy spectrum must be determined. Different bombarding particles result in significantly different recoil spectra. If the primary recoil spectra in two irradiation environments are substantially different, the effects of radiation per dpa can vary significantly. For instance, low-energy recoils are more efficient at producing point defects (e.g. electron irradiation), while high-energy recoils produce cascade damage and defect clusters (e.g. heavy ion irradiation, neutron irradiation). These differences in primary damage states have significant implications in long-term microstructural evolution and radiation-induced property changes [9]. Though dpa neglects the cascade structure of damage, it gives equivalent dose values for different types of irradiation, and it is regarded as the most appropriate correlation parameter for atomic displacement-induced property changes under irradiation [7,8].

Considering that several aspects of radiation exposure can give rise to the property changes, e.g. atomic displacement, nuclear transmutation, ionization, or their combined effects, the extent of the radiation damage cannot be fully characterized by a single parameter. To correlate radiation damage under various irradiation environments, several irradiation parameters must be considered, including (1) type and energy of irradiation particles and thus recoil spectrum. (2) fluence or dose, e.g. dpa (3) flux or dose rate, e.g. dpa/s, (4) irradiation temperature history, and (5) transmutation rates such as helium and hydrogen (He/dpa, H/dpa) and other solid impurities by nuclear reactions [10]. With regard to the high energy proton beam target design, focus will be on the effects of displacement dose rate, transmutation production rates, and pulsed irradiation.

Effect of Displacement Rate

The dose rate for high energy proton irradiations can be 2-3 orders of magnitude higher than neutron irradiation. In typical thermal (or mixed spectrum) neutron reactors, the dose rate is about 10^{-7} dpa/s, and in a fast fission reactor the dose rate is in the order of 10^{-6} dpa/s. Irradiations in accelerators and electron facilities can provide a wide range of dose rates with differences of several orders in magnitudes up to 10^{-3} dpa/s (see Fig. 1). While the wide range of dose rates obtained in different irradiation facilities provide an excellent tool for the accelerated irradiation experiments, the effect of dose rate on microstructural evolution and physical and mechanical properties is a significant issue. It is well recognized that the dose rate plays a critical role in irradiation-induced swelling, irradiation creep, and solute segregation [11]. As shown in Fig. 2, the peak swelling temperature in nickel and 18Cr10NiTi steel shifted to a higher temperature as the dose rate increased [12,13].

Effect of Transmutation Rate

Helium and hydrogen gas atoms are produced as transmutation products in materials during particle irradiations. Transmutation production rate (e.g. He/dpa, H/dpa) under various types of irradiation environments can be calculated using theoretical models and computer codes and verified by experiments [14,15]. The production rates of helium and hydrogen can be exceptionally high under high energy proton irradiations compared to those under fission neutron irradiations. For instance, the He/dpa ratios for stainless steel are 0.5, 15, and 200 ppm/dpa for fast fission, fusion and spallation neutron irradiations, respectively; the production rate for hydrogen is even higher: the H/dpa ratio for stainless steel for spallation is about 3000 ppm/dpa [16]. Figure 3 compares the helium production as a function of dpa in different irradiation sources in iron, except one case for W (high energy proton beams) [15-17].

Helium and hydrogen gas atoms have a negative impact on mechanical properties. Helium is essentially immobile in structural metals at typical temperatures of nuclear interest, while hydrogen has limited temperature-dependent mobility. Helium is known to assist void nucleation. It also causes high-temperature embrittlement in irradiated materials by forming helium bubbles at grain boundaries. Low-temperature embrittlement caused by helium has also been observed in some irradiated metallic materials [18]. The effect of hydrogen occurs at relatively low temperatures when its diffusivity is limited. In water-

cooled spallation components operating at low temperatures, high concentration of hydrogen can be accumulated in materials causing ductility loss by hydrogen bubble formation, or formation of hydrides [19].
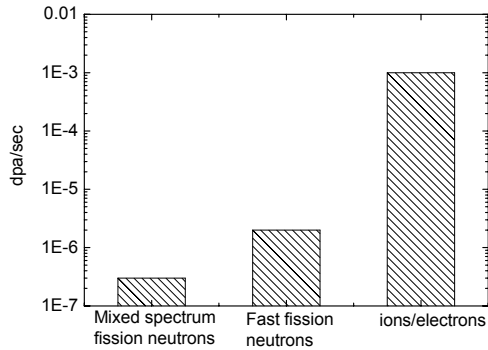

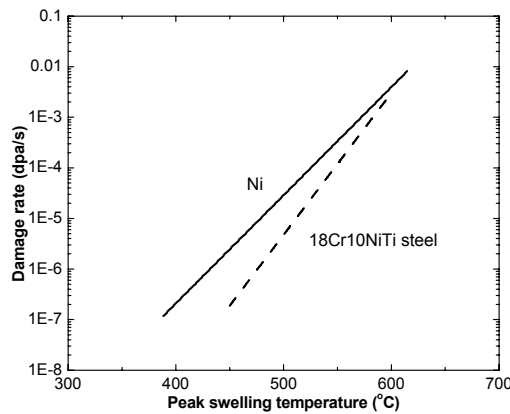Figure 1: Damage rates of mixed spectrum fission neutrons, fast fission neutrons and charged particles.

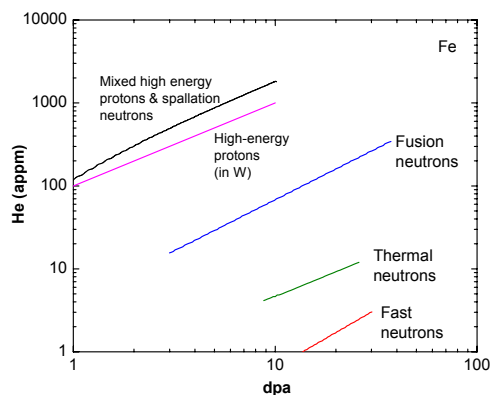Figure 2: Temperature shift of swelling as a function of dose rate in Ni and 18Cr10NiTi.

Figure 3: Helium production as a function of dpa in different irradiation sources.

Pulsed Irradiation vs. Continuous Irradiation

Under most irradiation conditions of fission neutrons and charged particles, irradiation occurs in a continuous manner at a given dose rate and temperature. In the proposed NuMI facility, however, irradiation occurs by a pulsed proton beam. Due to the pulsed nature of irradiation, the interplay of irradiation flux, temperature and pulse frequency can change the kinetics of irradiation damage accumulation compared to a steady-state continuous irradiation. The instantaneous displacement and transmutation rates can be extremely high in pulsed high energy proton irradiations. Very high dose rate in a pulse may severely limit the recombination of defects, resulting in much greater damage accumulation. On the other hand, annealing between pulses may significantly reduce the damage accumulation rate if the pulse frequency is sufficiently low and defect immigration is significant at the temperature.

Kmetyk et al. [20] studied the radiation effects in Al and Mo under cyclic pulsed irradiation. It was found that the pulse nature of irradiation is important when the characteristic pulse times are comparable to or greater than the vacancy and interstitial reaction times. If the pulsing is much more rapid than the reaction time, the system will not see any new effects. Radiation-induced temperature pulses increases void growth rate when the ambient temperature is below the peak swelling temperature, and conversely decreases void growth rate if the ambient temperature is above the peak swelling temperature. When radiation-produced temperature pulses are suppressed and the material remains at a constant ambient temperature, the void growth rate is not affected by pulse structure of radiation. However, these conclusions are not applicable for high ambient temperatures and very short intense radiation pulses.

Caturla et al [21] studied damage accumulation in Cu and Fe under continuous neutron irradiation and pulsed 14 MeV neutron irradiation of frequencies of 1, 10, and 100 Hz by combined molecular dynamics (MD) and kinetic Monte Carlo (kMC) simulations. The dose rate in the pulse was about 1.4 dpa/s in Fe and 2.2 dpa/s in Cu, and the pulse length was 1 μs. The simulation temperature was 300-340 K. The simulation data showed that damage accumulation in Cu under high-frequency pulsed irradiation ($\geq$10 Hz) was similar to that under continuous irradiation at a dose rate of 10^{-4} dpa/s. The evolution of vacancy cluster density in Cu was not affected by pulse frequency at high frequencies, while the damage accumulation rate was significantly reduced at 1 Hz, which implies that defect annealing between pulses at low frequencies can be significant in Cu at this temperature. The damage accumulation behavior in bcc Fe was different from Cu due to different crystalline structure and high interstitial impurity concentrations in Fe. In contrast to fcc Cu, irradiation damage under pulsed irradiation at 1 Hz in Fe was similar to the damage under continuous irradiation at a dose rate of 10^{-4} dpa/s. As frequencies increased, the damage difference between pulsed

irradiation and continuous irradiation increased, with higher damage accumulation at higher-frequencies.

RADIATION EFFECTS IN GRAPHITE

Experimental data of graphite irradiated under the NuMI-relevant irradiation conditions are extremely limited. The expected property changes in graphite must be inferred from nuclear fission and fusion materials research. It is therefore necessary to calculate the radiation damage not only in dpa, but also the dose rate (dpa/s) and helium and hydrogen production rate under the NuMI irradiation conditions. With the quantitative differences in displacement damage and He and H concentrations, property changes caused by high energy protons may be inferred from neutron irradiation data by incorporating the effects of damage correlation parameters such as dpa, dpa/s, He/dpa, H/dpa, temperature history, pulse character, etc. Depending on the property of interest, different damage correlation methods may be required.

It should be mentioned that the radiation effect studies in graphite have primarily focused on displacement damage rather than helium generation. Due to low production rates of helium and hydrogen in nuclear fission reactors, the effects of helium and hydrogen have received limited attention in fission reactor materials research. Only limited information on helium effects in irradiated graphite can be found in the literature [22-24]. Data on the dose rate effect and under pulsed irradiations are scarce. A thorough analysis on other types of nuclear reactor materials must be made to develop a better understanding of the effects of damage correlation parameters in graphite. The prediction of the production of helium and hydrogen in the target material and their effects on target performance also requires a well-developed understanding of microstructural evolution and their correlation with materials physical and mechanical properties.

SUMMARY

High energy protons cause displacement cascades and transmutation production of helium and hydrogen and solid impurities. Both displacement damage and production of helium and hydrogen must be considered in correlating damage between neutron and high energy proton irradiations and in evaluating the lifetime of the target. The pulsed nature of high energy proton irradiations in the NuMI environment should also be considered. High energy proton irradiations have higher recoil energies, and the production rates of helium and hydrogen and other foreign elements are also much more significant than under fission reactor irradiations. The pulsed proton beam generates extremely high instantaneous displacement rates that can potentially affect the kinetics of irradiation-induced defect production and accumulation behavior. These unique features of pulsed, high-energy proton irradiations may shift the temperature ranges of various radiation effects, change the incubation dose thresholds and rates of

irradiation-induced swelling and creep rates. In particular, the effects of transmutation products such as helium and hydrogen can have profound effects on materials performance not expected under typical fission neutron irradiations.

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