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Opportunities and limitations for ion beams in radiation effects studies: Bridging critical gaps between charged particle and neutron irradiations

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ABSTRACT

Ion beams are indispensible for understanding radiation effects in materials, enabling access to conditions challenging to perform with neutrons. However, ion irradiations can produce potential artifacts from beam rastering, high displacement damage rate effective "temperature shifts", subthreshold collisions, high ionization rates, and non-monoenergetic primary knock-on atom energies. The influence of near-surface denuded zones and implanted ion effects is analyzed, including diffusional broadening effects. At high ion irradiation energies, "swift heavy ion" effects can lead to enhanced defect production or recovery. Ion energies of 10–50 MeV typically offer a good compromise for minimizing near-surface, implanted ion, and swift heavy ion effects.

1. Advantages of ion beams for radiation effects research

Charged particle irradiation studies have been utilized for understanding irradiation effects on the microstructure and microchemistry in materials for many decades [1–3]. It is well established that ion irradiations can reproduce essentially all of the standard microstructural features observed in neutron-irradiated materials (dislocation loops, cavities, radiation-induced solute segregation, radiation-induced precipitation, etc.). Charged particle irradiations are a valuable complementary tool to bulk neutron irradiation studies, and in many cases provide unique capabilities to explore radiation effects phenomena. In particular, they allow the possibility to carefully and systematically control irradiation conditions over a wide range of doses in order to explore important test conditions such as exposure temperature, dose rate (including pulsed irradiation effects) and primary knock-on atom (PKA) energy. High displacement damage levels of ~100 displacements per atom (dpa) or higher can be achieved rapidly at relatively low cost with average PKA energies comparable to that of neutron or fission fragment irradiations, with little or no induced radioactivity [4,5]. Ion beams provide opportunities for improved experimental control that are difficult or impossible to perform during reactor neutron irradiations such as separate effects testing. For example, simultaneous dual or triple ion beam studies can quantify the roles of helium and hydrogen transmutation products on microstructural evolution during irradiation [6]. The thermal control region is located immediately adjacent to the irradiated region in the same sample, enabling quantitative differentiation of thermal- vs. radiation-induced microstructural changes.

The capability to produce controlled amounts of displacement damage under well-defined experimental conditions enables ion irradiations to be the preferred approach over neutron irradiations for many fundamental radiation effects studies. The length scale of the irradiation damage region for ion beams with MeV energies (~1 μm) is sufficiently large to extract microstructural information relevant for bulk irradiation conditions. By utilizing cross-section analysis techniques, the depth dependence of the damage rate and cumulative damage level can in principle be used to explore multiple dose rates and damage levels within a single fluence specimen. The near-atomic-scale resolution now achievable in modern electron microscopes and atom probe tomography allows chemical and defect cluster information to be obtained on scales comparable to that from modeling and simulation studies such as molecular dynamics. Valuable depth-dependent structural and chemical information can also be obtained from a variety of Rutherford spectrometry, nuclear reaction analysis [7], and/or glancing incidence x-ray diffraction measurements. In addition, technology improvements in time-dependent "4-dimensional" transmission electron microscopy [8] and synchrotron x-ray sources [9] enable valuable kinetic information to be obtained from in-situ ion irradiation studies in specialized facilities where the ion beams are linked to an electron microscope [10] or synchrotron beam line. Important defect production information can be obtained from analysis of bulk vs. thin foil ion irradiated specimens [11, 12]. Similarly, ion beam irradiations can enable important experimental

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validation of fundamental radiation effects processes, such as confirmation of molecular dynamics simulations of glissile defect clusters that may exhibit one-dimensional migration [13,14].

2. Limitations of ion beams for investigating neutron radiation effects

These important experimental benefits can be contrasted with a number of well-known limitations associated with ion beam irradiations that require careful data interpretation. Due to the limited penetration depth of ion beams in materials it is difficult to extract bulk property information from the irradiated region beyond microstructural and microchemical information, although promising improvements in nanoscale thermal conductivity [15–18] and mechanical property [19,20] techniques are under development.

A general limitation of ion beam irradiations is that the PKA recoils are distributed over a relatively wide range of energies compared to neutron irradiations, with most of the collisions producing low energy recoils due to long-range Coulombic interactions. For example, over 80% of the PKAs have energies <1 keV for 1 MeV self ion irradiation of medium mass targets such as Cu or Fe whereas the maximum PKA energy extends up to 1 MeV; in contrast, about 80% of the PKAs from 1 MeV neutron irradiation have energies within a factor of two of the maximum PKA energy of ~60 keV [21]. Therefore, near-monoenergetic PKA irradiations generally cannot be achieved using ion beams. Fig. 1 summarizes molecular dynamics simulations of the calculated surviving defect fraction (normalized to the NRT dpa) versus PKA energy for displacement events in Fe at 100 K [22]. The inset figure summarizes the calculated weighted defect recoil spectra W(T) versus PKA energy for 1 MeV neutrons and several ions in Cu, where W(T) is defined as the fraction of displacement damage produced in recoils of energy T [21]. The displacement events for ion irradiation occur over a wider energy range compared to neutrons, with a significantly higher fraction of low-energy and high-energy recoil events for the same average value of W(T). For example, ~20% and ~40% of the calculated displacement damage in Cu is associated with PKA energies below 10 keV for 1 MeV Kr and Ne ions, respectively, compared to <2% for 1 MeV neutrons. Whereas the surviving defect fraction is approximately constant at high PKA energies due to onset of subcascade formation [22] (>10 keV for medium-mass targets such as Fe, cf. Fig. 1), it increases rapidly at low PKA energies. Overall, the larger fraction of low energy PKAs for ion irradiation compared to neutron irradiation produces smaller-sized defect clusters created directly within displacement cascade events (with potentially different stability and mobility) and a higher surviving defect fraction per dpa compared to neutron irradiations.

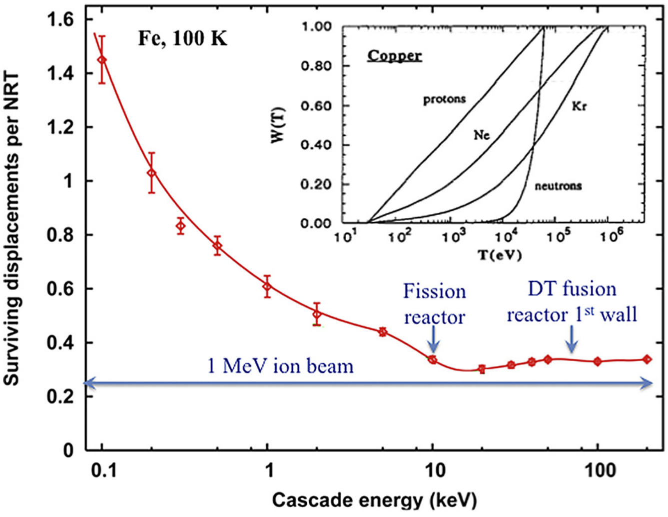

Fig. 1. Calculated surviving defect fraction (normalized to the NRT dpa) versus PKA energy for displacement events in Fe at 100 K [22]. The inset figure summarizes the calculated weighted defect recoil spectra W(T) versus PKA energy for several 1 MeV light ions and neutrons in Cu [21].

Energetic ion beams (>1 MeV) also produce high amounts of ionization per displaced atom along with an increased proportion of very low collisional energy transfers (below the threshold for stable displacement damage) compared to neutrons. These high ionization and subthreshold collision energy processes may cause pronounced increases in defect diffusion and point defect recombination relative to neutron irradiation conditions [23–29], particularly for energetic light ions. Since the energy transfer associated with subthreshold collisions (i.e., up to ~25–40 eV) is much higher than typical defect migration energies (~1 eV), subthreshold collisions can stimulate enhanced point defect migration and recombination in all irradiated materials [28,29]. However, further research is needed to quantify the importance of subthreshold collisions on defect evolution under a wide variety of irradiation conditions. The high amount of ionization per dpa for ion compared to neutron irradiations can also produce potentially dramatic reductions in surviving defect production in nonmetals such as semiconductors and ceramic insulators [23–27]. <10% of the energy loss of ~1 MeV medium-mass ions is associated with displacement damage collisions (with the majority energy loss associated with electron/ionization interactions), vs. approximately equal magnitudes of electronic and nuclear stopping powers for PKAs associated with neutron collisions. The ratio of ionization to nuclear stopping power increases with decreasing mass and increasing ion energy above ~1 MeV [23]. Ionizing radiation environments up to electronic stopping powers of ~10 keV/nm are typically of no consequence in metals due to their high concentration of conduction electrons, but can produce dramatic microstructural differences such as pronounced defect cluster coarsening or inhibition of amorphization in semiconductors and insulators compared to low ionization per dpa irradiation conditions [23–27].

An important advance in accelerator technology over the past few decades is the capability to raster a finely focused ion beam in order to achieve uniform irradiation fluences over the exposed area. However, the use of rastered or pulsed beams (frequencies of 0.1–250 Hz) has been shown in several experimental studies to lead to suppressed void swelling at intermediate temperatures [30–33]. Modeling studies indicate that the irradiation pulsing associated with beam rastering can alter the microstructural evolution, with effects most pronounced at pulse frequencies near ~1–100 Hz at intermediate temperatures [32–34]; the beam pulsing effect is predicted to disappear at very high pulse frequencies. Although further experimental and modeling studies are needed to quantify the material-, dose rate- and temperature-dependent conditions where beam pulsing effects become significant, it is recommended that defocused beams should generally be used instead of rastered beams for irradiation effects studies at elevated temperatures. Some slight electromagnetic wobbling of a defocused beam (where the ion flux at a given position never falls below ~50% of the peak flux) may be an alternative to provide a more uniform fluence over broad irradiated regions.

In order to take advantage of the high dose rate (which varies with depth) for typical ion irradiations, careful experimental planning and interpretation is required to enable quantitative comparison with neutron irradiation (lower dose rate) conditions. For example, in order to achieve comparable defect arrival rates at features such as sinks or defect cluster nuclei, high dose rate irradiations must be performed at higher temperature compared to low dose rate irradiations [35,36]. Unfortunately, this quantitative "temperature shift" is predicted to vary for different microstructural features (e.g., voids, interstitial dislocation loops, precipitates), since they depend on rate-controlling processes such as vacancy or solute migration with different thermal activation energies. Furthermore, kinetic rate theory analyses [32] predict a different functional dependence when trying to achieve equivalent point defect arrival fluxes at sinks versus when trying to achieve equivalent dislocation loop concentrations; in general, these analyses indicate no

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