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Measurement of displacement cross sections of aluminum and copper at 5 K by using 200 MeV protons

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ABSTRACT

To validate the Monte Carlo codes for prediction of radiation damage in metals irradiated by > 100 MeV protons, we developed a proton irradiation device with a Gifford–McMahon (GM) cryocooler to cryogenically cool two 0.25-mm-diameter wire samples of aluminum and copper. By using this device, the defect-induced electrical resistivity changes related to the displacement cross section of copper and aluminum were measured under irradiation with 200-MeV protons at 5 K at the beamline of the cyclotron facility at RCNP, Osaka University. After irradiation to a 3.89×10^{18} proton/m^2 flux, the damage rate of the aluminum sample was 1.30×10^{-31} Ωm^3/proton at 185 MeV and that of copper was 3.60×10^{-31} Ωm^3/proton at 196 MeV. Based on measurements of recovery of the accumulated defects in aluminum and copper through isochronal annealing, which is related to the defect concentration in the sample, about 50% of the damage remained at 40 K, with the same tendency observed in other experimental results for reactor neutron, fusion neutron, and 125-MeV proton irradiations. A comparison of the measured displacement cross sections with the calculated results of the NRT-dpa and the athermal-recombination-corrected displacement damage (arc-dpa) cross sections indicates that arc-dpa with the defect production efficiencies provided by Almazouzi for aluminum and Nordlund for copper provide better quantitative descriptions of the displacement cross section than NRT-dpa.

1. Introduction

To predict the operating lifetime of materials in high-energy (>100 MeV) radiation environments at accelerator facilities, such as spallation neutron sources and accelerator-driven systems for transmutation [1,2], Monte Carlo codes such as PHITS [3,4], FLUKA [5,6], MARS [7], and MCNPX [8] are used to calculate the transport of particles, nuclear reactions between particles and materials, distribution of primary knock-on atoms, and the number of displacements per atom related to the number of Frenkel pairs. A Frenkel pair is defined as a vacancy and a self-interstitial atom in the irradiated material. The Norgertt–Robinson–Torrens (NRT) model [9] has been widely used to predict the number of "initial" Frenkel pairs generated in the cascades of atom–atom collisions without the recombination of atoms (NRT-dpa). For more accurate estimation of the actual damage production, athermal-recombination-corrected displacement damage (arc-dpa) [10,11], which includes the results of the Molecular Dynamics simulation method (MD), is used as well. A coordinated research project in International Atomic Energy Agency titled "Primary Radiation Damage Cross Sections" will provide numerical databases of NRT- and arc-displacement cross sections [12].

For validation of the calculated displacement cross sections one can measure changes in electrical resistivity of samples at cryogenic temperatures (around 4 K) [13,14]. The number of surviving defects is then related to defect-induced changes in the electrical resistivity of metals at around 4 K, where the recombination of Frenkel pairs by thermal motion is well suppressed. The increase in electrical resistivity due to high-energy protons can be used to derive the displacement cross section. In addition, it provides other useful

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information such as degradation of the superconductor stabilizer, which compromises quench protection and increases magneto-thermal instability [15], in case of the superconducting magnets of the Muon-to-electron-conversion (Mu2e) [16] and the COherent Muon to Electron Transition (COMET) [17] experiments as well as of the High Luminosity Large Hadron Collider (LHC) [18] and the Future Circular Collider (FCC) [19].

In high-energy region, experimental displacement cross sections in the case of 125-MeV proton irradiation of copper were obtained at the Fixed-Field Alternating Gradient (FFAG) accelerator facility in Kyoto University Research Reactor Institute (KURRI) [14] and those in the case of 1.1- and 1.94-GeV proton irradiation of copper and tungsten were obtained at the Brookhaven National Laboratory (BNL) [13]. In the BNL experiment, the cryostat assembly for sample irradiation consisted of a complicated cryogenics system to deliver a metered flow of liquid cryogen (mixture of liquid nitrogen and liquid helium) for controlling the sample temperature. In the FFAG experiment [14], we developed a cryogen-free cooling system by using a Gifford–McMahon (GM) cryocooler. The sample was maintained at 12 K by using thermal conduction plates of the oxygen-free high-conductivity copper (OFHC) and electrical insulation sheets of aluminum nitride ceramic (AlN). Then, we measured changes in the electrical resistivity of the copper sample in relation to the displacement cross-section obtained under 125-MeV proton irradiation. However, in this experiment, the minimum temperature of the sample did not reach 4 K owing to heat penetration through the AlN plate. Because of its structure, the cryogenic device could only cool one sample at a time, and the time required to change the sample was long (e.g. 3 h for cooling to cryogenic temperature and 24 h for heating the sample to room temperature), it was almost impossible to get data for more than two samples within the limited time of beam operation.

In the present work, we have modified the device to be able to cryogenically cool two samples at once by using a better cooling system. Then, we measured the changes in electrical conductivity of aluminum and copper samples by using 200-MeV protons at the RCNP cyclotron facility [20], Osaka University, which can provide proton beams with energies from 100 to 400 MeV and beam intensities of up to 1 μA. Recovery of the accumulated defects through isochronal annealing [21], which is related to the defect concentration in the sample, was measured after the cryogenic irradiation. Finally, the derived displacement cross-sections were compared to the calculated results of the NRT-dpa and arc-dpa models considering the defect production efficiencies.

2. Experiments

2.1. Cryogenic irradiation chamber and target assembly

The proton irradiation chamber developed in this work was installed at the N0 beam line in the RCNP cyclotron facility, Osaka University, as shown in Fig. 1. Protons with energies of 200 MeV and beam current between 1 and 3 nA were directed to the irradiation chamber with the quadrupole magnets, which control beam size at the sample position. The cryogenic proton irradiation chamber was maintained at a vacuum level of about 1×10^{-5} Pa by using a rotary pump and a turbo-molecular pump.

Fig. 2 shows a schematic of the cryogenic irradiation chamber with the GM cryocooler (RDK-408D2, Sumitomo Heavy Industries, Ltd.) with a cooling capacity of 1 W at 4 K and the twin sample assembly connected to the 2nd stage of the cold head. The GM cryocooler cooled the sample by means of a conduction coolant via the aluminum plate and the OFHC block, which have high thermal conductivities, 220 W/(m K) for aluminum at 300 K and 300 W/(m K) for OFHC at 300 K. The 1-mm-thick aluminum plates of the thermal radiation shield connected to the 40 K stage of the refrigerator covered the entire sample assembly to intercept any thermal radiation from the ambient irradiation chamber. For a comparison of the cryogenic irradiation chamber with the previous device used in the FFAG experiment, the thermal radiation shields attached to the 40 K stage of the refrigerator were strengthened by using bolts, and the 1 W cooling capacity of the GM refrigerator was double the 0.5 W cooling capacity of the refrigerator used in the FFAG experiment.

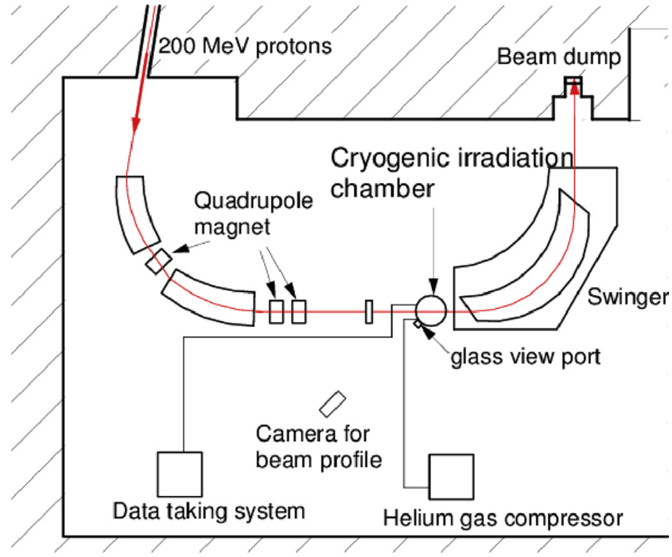

Fig. 1. Schematic of N0 beam line of cyclotron accelerator facility at RCNP, Osaka University.

For a subsequent annealing study of the accumulated damage products, a 40 Ω electrical resistance heater was attached to the OFHC block. A pre-calibrated electrical resistance thermometer (Cernox thermometer, Lake Shore Cryotronics, Inc.) was attached in the OFHC block and the AlN plate to confirm the cooling performance of the GM cryocooler. To simultaneously measure the changes in electrical resistivity of two samples under proton irradiation, two aluminum plates with the aluminum and copper wire samples were connected to the OFHC block by using bolts, as shown in Fig. 2.

Fig. 3 shows a detailed drawing of the AlN sample holder with the wire sample, and Table 1 lists the characteristics of wire samples. Each aluminum and copper wire with a 0.25-mm diameter, purchased from Nirako Corporation, was set on the AlN plate in a serpentine-shaped line. The AlN plate was used in the FFAG experiment as well because of its excellent electrical insulation and high thermal conductivity (21 W/m K at 4 K) [22]. Before the irradiation, the aluminum wire and the copper wire were preheated in vacuum for 1 h at 550 °C (823 K) and 1000 °C (1273 K), respectively. The wire and the CX1050-SD Cernox resistance thermometer were carefully sandwiched between 1-mm thick and 1.5-mm thick AlN plates, respectively. The sample folder was embedded into a 0.5-mm-deep hollow on the aluminum back plate and compressed by the 1-mm thick aluminum plate, providing good thermal contact between the sample and the AlN sheets.

The electrical resistance of the wire was measured using a combination of a current source (model 6221, Keithley Instruments, Inc.) and a nano-voltmeter (model 2182 A, Keithley Instrument, Inc.) in the same manner as that in the FFAG experiment [14]. This apparatus is based on the current-reversal method (four-prove