Measurement of defect-induced electrical resistivity change of tungsten wire at cryogenic temperature using high-energy proton irradiation

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For validation of the number of displacements per atom (DPA) of tungsten, we measured the defect-induced electrical resistivity change of the wire sample of tungsten related to the displacement cross section under irradiation with 389-MeV protons at 10 K. The Gifford–McMahon (GM) cryocooler cooled the sample by means of a conduction coolant via the aluminum plate and the oxygen-free high-conductivity copper (OFHC) block. After irradiation with $1.36 \times 10^{19}$ protons/m², the damage rate of the wire sample was $4.35 \times 10^{-30} \Omega \text{m}^3$/proton. In comparison with experimental data under 1.1 and 1.9 GeV proton irradiation, we found that the damage rate of tungsten increases with proton energy due to an increase in the number of secondary particles produced by nuclear reactions.

KEYWORDS: Proton Irradiation, Damage Rate, Tungsten

1. Introduction

To predict the operating lifetime of target materials in high-energy (>100 MeV) radiation environments at spallation neutron sources, radiation transport codes such as PHITS [1,2] are used to calculate the number of displacements per atom (DPA) related to the number of Frenkel pairs that is defined as a vacancy and a self-interstitial atom in the irradiated material. For validation of the displacement cross sections in the high-energy region, it is necessary to measure displacement cross sections in relation to changes in electrical resistivity at cryogenic temperature (below 10 K). The number of surviving defects is related experimentally to defect-induced changes in the electrical resistivity of metals at cryogenic temperature, where the recombination of Frenkel pairs by thermal...
motion is well suppressed.

The electrical resistivity changes of copper and tungsten at cryogenic temperature in the case of 1.1- and 1.94-GeV proton irradiation were obtained at the Brookhaven National Laboratory (BNL) [3]. In the BNL experiment, the cryostat assembly for sample irradiation consisted of a complicated cryogenics system to deliver a metered flow of liquid cryogen for controlling the sample temperature.

In order to measure the data without liquid cryogen, we developed a cryogen-free cooling system by using a Gifford–McMahon (GM) cryocooler and measured the data in the case of 125-MeV and 200-MeV proton irradiation of copper and aluminum at the Fixed-Field Alternating Gradient (FFAG) accelerator facility in Kyoto University Research Reactor Institute (KURRI) [4] and the RCNP cyclotron facility [5], Osaka University. The sample was maintained at cryogenic temperature by using thermal conduction plates of oxygen-free high-conductivity copper (OFHC) and electrical insulation sheets of aluminum nitride ceramic (AlN).

In the present work, we measured the electrical resistivity change of tungsten by using 389-MeV protons at the RCNP cyclotron facility with same device in our previous measurements [5]. The damage rate of tungsten was compared with other experimental data under proton irradiations.

2. Experiments

The experiments were carried out at the N0 beam line in the RCNP cyclotron facility with same device in our previous experiments as shown in [5]. Protons with energies of 389 MeV and beam current of 10 nA were directed to be incident on the irradiation chamber.

Figure 1 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 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. 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. A pre-calibrated electrical resistance thermometer (Cernox thermometer, Lake Shore Cryotronics, Inc.) was attached in the OFHC block and the AlN plate to measured temperature using a temperature controller (model 335, Lake Shore Cryotronics, Inc.). To measure the change in electrical resistivity of the sample under proton irradiation, the aluminum plate with the tungsten wire sample was connected to the OFHC block.

![Fig. 1. Schematic of the cryogenic irradiation chamber.](image-url)
Table I lists the characteristics of the wire sample. The tungsten wire was set on the AlN plate in a serpentine-shaped line within a 20 mm diameter circle. Before irradiation, the tungsten wire was annealed in vacuum for 0.5 hours at 1973 K. The length between the two potential points was 144 mm. 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 2182A, Keithley Instrument, Inc.). This apparatus is based on the current-reversal method (four-probe technique) in the delta mode, which works by sourcing pulses with opposite polarity and taking one measurement during each pulse. The electrical resistivity of the sample \( \rho \) is expressed as follows: \( \rho = RA/L \), where \( R \) is the measured electrical resistance, \( L \) is the length between two potential points, and \( A \) is the area of the sample.

For imaging with proton beams, a 1-mm-thick ZnS fluorescent screen was attached onto the thermal radiation shield. When protons interact with the fluorescent screen, a light detectable using a camera is emitted. The camera was set near the glass view port of the cryogenic proton irradiation chamber to capture the beam shape with data for the red, green, and blue (RGB) lights. The beam size was adjusted to cover the wire sample by controlling the magnetic fields of the quadrupole and bending magnets upstream of the irradiation chamber. The irradiation length of the wire was 113 mm. The number of protons during irradiation was measured by a Faraday cup at the beam dump. About 70% protons through the sample were bent to the beam dump correctly.

### Table I. The characteristics of the wire sample.

| Material | Tungsten |
|----------|----------|
| Diameter (mm) | 0.25 |
| Length (mm) | 144 |
| Purity (%) | 99.95 |
| Annealing | 1973 K for 0.5 hours |

3. Experimental results

Figure 2 shows the relationship between the electrical resistance and the temperature of the AlN plate during proton irradiation with an energy of 389 MeV with average currents of 10 nA. The temperature was maintained below 10 K during beam irradiation. The electrical resistances of the tungsten increased during beam irradiation owing to the production of defects in the wires. The jump in resistance at the beam-on and beam-off times resulted mainly from the temperature increase and decrease due to beam heating. The total beam fluence on the sample was \( 1.36 \times 10^{19} \) protons/m\(^2\), the total increase in resistance was 0.136 m\( \Omega \) and the damage rate of the tungsten, which is the ratio of the change in resistivity of metal \( \Delta \rho_{\text{metal}} \) to the beam fluence \( \phi \), was \( 4.35 \times 10^{-30} \) m\( \Omega \) m\(^3\)/particle.

Table II lists the damage rate of tungsten, copper and aluminum at cryogenic...
temperature in high energy region. For the comparison with experimental data using 1.1 and 1.9 GeV protons, damage rate of tungsten increases with proton energy. According to the PHITS calculation, the number of secondary charged particles produced by nuclear reaction increases with proton energy in the energy range from 389 MeV to 1.9 GeV because the nuclear reaction cross section related with the number of secondary particles increases with proton energy in this energy range. On the other hand, damage rates of copper are almost same because the nuclear reaction cross section for proton incidence on copper is almost same in this energy range.

In future, we will derive the displacement cross section of tungsten using the damage rate and compare the experimental data with results calculated by the PHITS code.

| Damage rate \(10^{31}\Omega m^3/\text{proton}\) | 185 MeV proton [5] | 196 MeV proton [5] | 389 MeV proton [3] | 1.1 GeV proton [3] | 1.9 GeV proton [3] |
|---------------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| Tungsten                        | -               | -               | 43.5            | 68.9            | 101             |
| Copper                          | -               | 3.6             | -               | 2.9             | 3.7             |
| Aluminum                        | 1.3             | -               | -               | -               | -               |

4. Summary

For validation of calculated DPA values of tungsten, we measured the defect-induced electrical resistivity change of the wire sample of tungsten related to the displacement cross section under irradiation with 389-MeV protons at 10 K in RCNP, Osaka University. The GM cryocooler cooled the sample by means of a conduction coolant via the aluminum plate and the OFHC block. After irradiation with \(1.36 \times 10^{19}\) protons/m\(^2\), the damage rate of the wire sample was \(4.35 \times 10^{-30}\) \(\Omega m^3/\text{proton}\). In comparison with experimental data using 1.1 and 1.9 GeV protons, damage rate of tungsten increases with proton energy in the energy range from 389 MeV to 1.9 GeV due to increase the number of secondary particles produced by nuclear reactions.

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