Measurement of displacement cross-sections of Nb irradiated by protons with kinetic energy range between 0.4 and 3 GeV

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Abstract.
In high-intensity proton accelerator facilities, it is crucial to evaluate the damage of beam-interception materials and accelerator components, such as a superconducting magnet coil and cavity. The displacement per atom (dpa) is used as a damage index derived by integrating the particle flux and the displacement cross section. Although the dpa is employed as the standard, the experimental data of displacement cross section are scarce for a proton in the energy region above 20 MeV. To obtain the data for superconducting materials for high-intensity accelerators and magnets, we measured the displacement cross section of Nb for proton irradiation with a kinetic energy range between 0.4 and 3 GeV at J-PARC. The present experimental results were compared with the calculation of PHITS code and Karlsruhe Institute of Technology (KIT) evaluation using both Norgertt-Robinson-Torrens (NRT) and the athermal recombination corrected dpa (arc-dpa) models. The experiment showed that the widely utilized NRT model overestimates the cross section by 50%. It is also found that the arc-dpa model shows remarkably good agreement with the present data.

1 Introduction
To reduce the hazard of radioactive waste generated in nuclear reactors [1], Japan Atomic Energy Agency (JAEA) has proposed an Accelerator Driven System (ADS) consisting of a very high power accelerator (30 MW) with a proton kinetic energy of 1.5 GeV. Lead-bismuth eutectic (LBE) is used as both the target material and the coolant. In the design of ADS, damage to the beam-intercepting material is one of the critical issues. The beam interceptor also plays an essential role in other high-intensity accelerator facilities, such as neutron source facilities. In the Materials and Life Science Experimental Facility (MLF) [2, 3] of the Japan Proton Accelerator Research Complex (J-PARC) [4], an aluminum alloy beam window (A5083) is employed [5]. The T2K collaboration [6] at J-PARC also uses a titanium alloy [7] for the beam window and uses niobium (Nb) for superconducting magnets with a high-magnetic field to transport the proton beam to the target. The fixed tungsten target is also one of the candidates for the COMET [8] experiment, which will be used to produce muons in plans of J-PARC. The Nb materials play an essential role in the superconducting (SC) magnet, which was used at T2K, and will be applied as the cavity of the SC accelerator. In order to operate the high-power accelerator confidently, it is essential to evaluate the damage to the Nb materials. In order to quantitatively identify the damage to the target materials, a measure called displacement per atom (dpa) is commonly employed.

The unit of dpa is widely used to quantify damage in nuclear and fusion reactors. The dpa is estimated using the particle fluence multiplied by the displacement cross section, which is usually obtained using the Norgertt-Robinson-Torrens (NRT) model [9]. In the low energy region below 20 MeV, the proton dislocation is mainly done by the Coulomb force so that the dislocation cross section can be reliably predicted. However, for protons in the high energy region above 20 MeV, there are few experimental data on the displacement cross section. Therefore, displacement cross sections have not been well studied overall. Since many reaction channels open above 20 MeV, we use a calculation code based on the nuclear cascade model to obtain the cross sections. Although the Nb materials play an important role in the accelerator components for the ADS, the spallation neutron source, and high-energy physics devices, there were no experimental data of displacement cross sections in the energy region above 20 MeV. Recent experiments [10] were performed in a beam transport system called 3NBT at J-PARC to obtain the displacement cross section of iron and copper. In this study, the same techniques were applied to measure the displacement cross sections of Nb.

2 Experiment at J-PARC
A vacuum chamber with a cryo-cooler was installed in front of the beam dump of the 3-GeV synchrotron (RCS). Because of insufficient space in the beamline to introduce the sample into the beam dump, the experimental setup

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was installed in the beamline where the high-intensity proton beam could be fed to the spallation neutron source in the MLF. Although, to maintain the cryogenic temperature of the sample, a low-intensity proton beam was used, an additional interlock of the accelerator was required for the experiment. In the RCS, the kinetic energy of the extracted protons varies in the range of 0.4 to 3 GeV, which changes the timing of the extraction kicker electromagnets. As the stiffness of the subsequent electromagnet changes, the beam is introduced into the sample.

To observe the damage in the specimen, we cooled the irradiated specimen to cryogenic temperature (< 20 K), where the recombination of Frenkel pairs is sufficiently suppressed by thermal motion. The displacement cross section can be measured by following Matthiessen’s rule. For a material above the superconducting critical temperature, the displacement cross section can be measured by following Matthiessen’s rule. The Nb sample is heated to 8 K by a heater placed on the sample holder, slightly above of critical temperature of superconducting (Tc). The experimental displacement cross section σ_exp(E) can be obtained by observing the electrical resistance change Δρ due to irradiation at cryogenic temperature, as given follows,

\[
\sigma_{\text{exp}}(E) = \Delta \rho / (\rho(E) \rho_{FP}),
\]

where ρ(E) is the average proton fluence in the sample and ρ_{FP} is the electrical resistivity change per Frenkel pair of the sample. The intensity of the proton beam introduced to the sample was observed using a well-calibrated current transformer. Results from another study [11] showed that a simplified two-dimensional Gaussian well characterized the beam shape. The width of the beam was observed by a beam profile monitor [12] placed near the sample. The beam widths at the sample protons were determined with the fitting of the beam optics. For 3-GeV protons, the beam widths were 4.0 mm and 5.0 mm at 1σ in the horizontal and vertical directions, respectively.

The vacuum chamber with a Gifford–McMahon (GM) cryogenic refrigerator (Sumitomo Heavy Industries RDK-408D2), having a cooling capacity of 1 W at 4 K, was installed at 3-GeV beam transport line (3NBT). The GM cryocooler cooled the samples using oxygen-free, highly conductive copper rods and an aluminum sample holder placed at the end of the rods, as shown in Fig. 1. The GM cooler consists of two stages: the first and the second. The temperature reaches 40 K in the first and 4 K in the second. To measure the thermal recovery rate of the sample, we attached a heater to a copper rod. The GM cryocooler and sample wire assembly were placed on a movable stage to facilitate irradiation control; the RCS kicks the beam out horizontally, so the beam position is more stable in the vertical direction than in the horizontal direction. To minimize the effect of beam position instability, we strung the sample wire horizontally.

The Nb wire with a diameter of 0.25 mm and purity of 99.9% was employed in the experiment. Before installation of the wire, to eliminate lattice defects, the wire was annealed to ~ 2273 K, slightly below the melting point (2744 K). Each sample was sandwiched between electrically insulating aluminum nitride ceramic (AlN) sheets and held in an aluminum holder. In order to minimize beam interaction on the holder, a 40 mm diameter aperture was provided in the AlN and aluminum holder. The resistance of the sample was measured using a voltmeter and a current source. For compensation of the cable resistance, the resistance of the sample wire was read using the current source and the voltmeter via 4 terminals of 1, Γ, V’, and V”, as shown in Fig. 1. To improve the accuracy, the delta mode supplying a current of ±0.1 with exchanged polarity was adopted, and the frequency was set to 10 Hz. The voltage on the wire was read at intervals of 1 minute. The accuracy of this resistance measurement was ±0.01 µΩ, which corresponds to a resistivity of ±3 Ωm.

The resistance thermometer attached to the sample holder was calibrated over the temperature range of 4 - 100 K. The silicon thermometer attached to the copper column was calibrated between 4 K and room temperature. As a result, it was confirmed that the resistance decreased with the temperature. It was also confirmed that the resistance saturated near the cryogenic temperature (~ 8 K).

The electrical resistivity of a sample ρ is expressed as

\[
\rho = RA/L = RD^2/4L,
\]

where R[Ω] is the measured electrical resistance, A is the area of the sample (4.9×10^{-8} m^2) with diameter, D (0.25 mm), and L is the length between two potential points (40 mm). In order to avoid the heat introduction due to radiation, a double-walled radiant aluminum heat shield was employed around the sample. The outer and inner shields were 2 mm and 1 mm thick, respectively, and were directly connected to the first and second stages of the GM cooler. To minimize the scattering of the proton beam at the shields, thin aluminum foils with a thickness of 5 µm were placed in the beam entrance and exit holes (40 mm diameter) of both shields. After 6 hours of cooling, the temperature of the sample holder was less than 4 K. At the beginning of the beam irradiation experiment, the temperature was not 4 K, but ~ 20 K. Several experiments were conducted with different conditions, but the heat was introduced from the resistance and temperature measurement cables. A heater anchor was
installed for cables inside the vacuum chamber at the second stage of the GM cooler to reduce the introduction of heat from the outside. As a result, it was confirmed that the heat from the outside was significantly reduced. In addition, thin wires were used for the measurement cables installed in the chamber, reducing the heat to the sample.

### 3 Experimental results of displacement cross-section

In order to maintain the damage to the specimen due to irradiation, a low-intensity proton beam is preferable to avoid excessive heat generation. However, with a low-intensity beam, it is difficult to observe the distribution by the beam profile monitor. Therefore, we observed the beam shape by varying the intensity per pulse. Here, we adopted a proton intensity of $2.7 \times 10^{12}$ per pulse, which is considered the minimum intensity to obtain a precise beam shape. For each kinetic energy, 400 pulse shots of the proton beam were irradiated to the sample.

As the first step of the experiment, the sample wire was irradiated with 3 GeV protons. In order to obtain the precise cross-section, the vertical beam position was aligned to the sample wire because it was stung horizontally. Before starting irradiation, the vertical beam position was scanned within 3 mm using the upstream steering magnet of the sample. By observing the resistance change during the scan, the beam position was aligned to the sample wire. After continuous beam irradiation, the sample resistance was found to be increased by about 0.2 $\mu$Ω from the start of beam irradiation caused by displacement. From the resistance value when the specimen is cooled without irradiation, the average temperature is expected to be less than 20 K, which is low enough to maintain the damage of the specimen during irradiation.

Using Eq.(1), the displacement cross-sections of Nb are obtained and shown in Table 1. The resistivity change per Frenkel pair ($\rho_{FP}$) was determined to be 14 ± 3 $\mu$Ωm for Nb, which was obtained in a different study [13]. It should be pointed out that the error dominates the error in the present cross-sectional data in the resistivity change per the Frenkel pair. The uncertainty in the resistivity change due to Frenkel pair creation needs to be improved to obtain highly accurate results. For this purpose, the further experiment use of electron beams is expected to improve the uncertainty.

**Table 1. Result of displacement cross sections obtained by the present study**

<table>
<thead>
<tr>
<th>Proton kinetic energy [GeV]</th>
<th>Nb [b]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.4</td>
<td>1920 ± 455</td>
</tr>
<tr>
<td>0.8</td>
<td>2140 ± 508</td>
</tr>
<tr>
<td>1.3</td>
<td>2390 ± 568</td>
</tr>
<tr>
<td>2.2</td>
<td>2250 ± 534</td>
</tr>
<tr>
<td>3.0</td>
<td>2550 ± 606</td>
</tr>
</tbody>
</table>

### 4 Calculation of displacement cross section

The displacement cross section is defined by the following equation, which is based on the calculation using the intra-nuclear cascade model.

$$\sigma_{\text{disp-cal}}(E) = \sum \int_{E_d}^{T_i^{\text{max}}} N_d(T_i) \frac{d\sigma}{dT_i}dT_i,$$

where $E$ is the kinetic energy of the projectile, $d\sigma/dT_i$ is the recoi atom kinetic energy distribution, $T_i$ is the kinetic energy of the recoil particle $i$ effective up to a maximum of $T_i^{\text{max}}$, and $E_d$ is the effective threshold displacement energy, which is 78 eV for Nb. In addition, $N_d(T_d)$ is the number of Frenkel pairs representing the efficiency of defect formation defined by vacancies and inter-self atoms in the irradiated material, which is widely used in the NRT model [9]. The number of atomic displacements as a function of cascade energy and damage function $(N_d)$ is given by the NRT model given as the following equation:

$$N_d(T_d) = \begin{cases} 
0, & (T_d < E_d) \\
1, & (E_d < T_d < 2E_d/0.8) \\
\xi(T_d) \cdot 0.8T_d/2E_d, & (2E_d/0.8 < T_d) 
\end{cases}$$

where $T_d$ is the damage energy, i.e., the kinetic energy available to produce the atomic displacement. The NRT model gives the defect efficiency of $\xi$ as 1, regardless of $T_d$, which applies an approximation with a simple linear collision cascade in the lattice. The PHITS code [14] implemented the NRT calculation model [15].

Recently the damage model has progressed based on the molecular dynamics (MD) simulations, which suggested to use athermal-recombination-corrected dpa (arc-dpa) model [16, 17] for accurate estimation of the displacement. When the atoms are highly excited due to receiving recoi energy, called the primary knock-on atom (PKA), many of them are displaced from their initial lattice. When the cascade equilibrates with its surroundings thermally, the MD results show that almost all atoms return to their full lattice positions. The final number of defects is much smaller, and the number of atoms replaced by other atoms (atom mixing) is much larger than that predicted by the NRT model. In the arc-dpa model, when $T_d$ is greater than $2E_d/0.8$, the defect production efficiency $\xi$ is expressed by the following:

$$\xi(T_d) = (1 - c)(2E_d/0.8)^{-b}T_d^b + c,$$

where the parameters $b$ and $c$ are derived from the MD [17], which were determined to be -1 and 0.63, respectively. The arc-dpa model is also implemented in PHITS.

### 5 Results and discussion of displacement cross-section

Figure 2 compares the present results of the displacement cross section of Nb with the previous experimental data [18]. Since the cross sections of Jung [18] are not explicitly given, the experimental cross sections were calculated
by dividing the damage rate [18] given in the literature by the value of $\rho_{FP}$ [13], which is the same as the present experimental data. The experimental data shows that the displacement cross section is dominated by Coulomb force in the low-energy region and that the cross section decreases rapidly as the proton energy increases. On the contrary, in the high-energy region, nuclear reactions give the cross sections almost exclusively.

Figure 2 was calculated using PHITS [15] with the INCL-4.6 [19] cascade model. The present data showed an overestimation of the NRT model using PHITS by about 50%, whereas a factor of 2 ~ 3.5 overestimated the data for Fe and Cu [10]. Also, in Fig. 2, the evaluation data of Karlsruhe Institute of Technology (KIT) are shown, which were evaluated using the NRT model and the arc-dpa model. It was found that the KIT evaluation by the NRT model overestimated the actual results by about 50%. Applying the arc-dpa model, the PHITS calculation and KIT evaluation show a good agreement with the experimental data, except for a slight underestimation at 3 GeV of the KIT evaluation.

![Figure 2. Comparison of the present data on Nb with previous experimental data [18], calculated data by PHITS [15], and evaluated data by KIT [20] as a function of kinetic energy.](image)

6 Conclusion

Displacement cross-section experiments were carried out at J-PARC to evaluate the accelerator components used in the high-intensity proton accelerator. We successfully obtained displacement cross sections of Nb irradiated with protons having kinetic energies of 0.4 – 3 GeV. The data were obtained by observing the change in the electrical resistance of the irradiated specimens at low temperatures (8 K). The present data are the first experimental data of Nb in the energy region above 400 MeV, which is essential for the evaluation of the displacement of the superconducting accelerator components.

The present experimental data of displacement cross sections were compared with the calculations using PHITS and the evaluation data by KIT for applying both NRT and arc-dpa models. The widely adopted NRT model overestimated the present experimental data by 50%, which showed agreement with the data for low-energy proton projectiles below 10 MeV. In contrast, the arc-dpa model agrees remarkably well with the present experimental results all over the energy range. Therefore, it can be concluded that the arc-dpa model should be adopted for the dpa calculation for the damage evaluation of Nb.

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References