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Irradiation damage from low-dose high-energy protons on mechanical properties and positron annihilation lifetimes of Fe–9Cr alloy

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HIGHLIGHTS

- Nuclear reactions in accelerator-driven systems (ADS) result in the generation of helium within the ADS materials.
- The amount of helium produced in this way is approximately one order of magnitude higher than that generated by nuclear fusion.
- As helium is well-known to induce degradation in the mechanical properties of metals, its effect on ADS materials is an important factor to assess.
- The results obtained in this study show that low-dose proton irradiation (11 MeV at 573 K to 9.0×10^{-4} dpa and 150 MeV at room temperature to 2.6×10^{-6} dpa) leads to a decrease in yield stress and ultimate tensile strength in a Fe–9Cr alloy.
- Moreover, interstitial helium and hydrogen atoms, as well as the annihilation of dislocation jogs, were identified as key factors that determine the observed softening of the alloy.

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ABSTRACT

Nuclear reactions in accelerator-driven systems (ADS) result in the generation of helium within the ADS materials. The amount of helium produced in this way is approximately one order of magnitude higher than that generated by nuclear fusion. As helium is well-known to induce degradation in the mechanical properties of metals, its effect on ADS materials is an important factor to assess. The results obtained in this study show that low-dose proton irradiation (11 MeV at 573 K to 9.0×10^{-4} dpa and 150 MeV at room temperature to 2.6×10^{-6} dpa) leads to a decrease in yield stress and ultimate tensile strength in a Fe–9Cr alloy. Moreover, interstitial helium and hydrogen atoms, as well as the annihilation of dislocation jogs, were identified as key factors that determine the observed softening of the alloy.

1. Introduction

Accelerator-driven systems (ADS) are devices coupling a subcritical nuclear reactor with a proton accelerator. These devices have received considerable attention in the field of nuclear energy [1,2]. The beam window and the solid target in ADS are subjected to heavy irradiation damage by source protons, spallation products, and fission neutrons generated in the reactor. An important feature of the damage process in ADS is the formation of large displacement cascades [3], in which primary knock-on atoms and secondary particles formed by nuclear reactions produce a considerable number of defects in localized areas of the material [4]. Also important is the formation of spallation products, which act as impurities and cause the material properties to change during irradiation. In particular, the generation of insoluble helium impurities induces substantial bubble formation at sufficiently high temperatures, because the ratio of helium generated (measured in appm) to displacement per atom produced (measured in dpa) is more than 10 times higher in ADS than that in fusion reactors [5,6]. Proton beam irradiation tests are essential for the development of structural materials for ADS. Several previous irradiation studies have been performed using strong spallation neutron sources such as SINQ [7–9] and LANSCE [10]. Currently, however, there are no materials that enable the beam window to be operational for the desired period of time, such as 600 effective full-power days (about

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110 dpa, for the case of 600 MeV incident proton energy) without deteriorating the mechanical properties [1,6].

High-Cr ferritic steels are excellent candidates for fusion and advanced fission reactors because they are low-activation alloys [11] with superior heat and swelling resistance [12–14]. These steels also hold significant promise as ADS materials. Previous studies on these alloys have mainly involved experiments with high irradiation doses [7–9]. However, the irradiation damage processes are multiscale phenomena, whose fundamental understanding requires experiments performed with low irradiation doses [15].

In this study, in order to investigate the changes in mechanical properties of the alloys following irradiation, high-energy low-dose proton irradiation tests of a ferritic steel were performed using an 11 MeV linear accelerator (linac) and a 150 MeV fixed-field alternating-gradient (FFAG) synchrotron accelerator at the Research Reactor Institute of Kyoto University [16].

2. Experimental procedures

Fe–9Cr (wt% composition Fe–8.5Cr–0.88Mo–0.44Mn–0.28Si–0.2V–0.096C-0.076Nb–0.05Ni–0.05N–0.005Al–0.002S) ferritic steel was used in the present study. The chemical composition was determined by Nippon Steel & Sumikin Technology Co. Ltd. using inductively coupled plasma-mass spectrometry (ICP-MS) and X-ray fluorescence analysis (XRF) to measure the levels of impurities and main elements, respectively. A block of Fe–9Cr alloy was sliced by a wire saw to 0.3 mm in thickness and further polished mechanically and chemically to a final thickness of 0.25 mm. Miniature tensile specimens (gauge: 0.25 mm × 1.2 mm × 5 mm; see Fig. 1) were punched from the Fe–9Cr sheets and annealed at 923 K in a 5 × 10^{-3} Pa vacuum. The tensile testing samples were irradiated vertically with 150 MeV protons at room temperature using the FFAG synchrotron, and by 11 MeV protons at 573 K using a linac accelerator.

The proton flux and the total fluence for the 150 MeV proton-irradiation were 6.3 × 10^9 p/cm^2/s and 4.5 × 10^{15} p/cm^2, respectively. The beam current was monitored by a Faraday cup. The stability of the current was within ±20%. The damage and the transmutation were calculated using the Particle and Heavy Ion Transport code System (PHITS-2.52) [17,18]. The damage was mainly due to the secondary particles formed by nuclear reactions of 150 MeV protons with elements in the alloy. Figs. 2 and 3 display the generation and the energy spectra of secondary particles such as neutrons, hydrogen, and helium formed by nuclear reactions between protons and the Fe–9Cr sample. The secondary particles formed include light atoms such as hydrogen (Z = 1) and helium (Z = 2), as well as heavy atoms such as iron (Z = 26), nickel (Z = 28), and molybdenum (Z = 42). The total damage was 2.6 × 10^{-6} dpa, whereas the appm/dpa ratios for hydrogen and helium were 1100 and 120, respectively, according to the simulation. The temperature

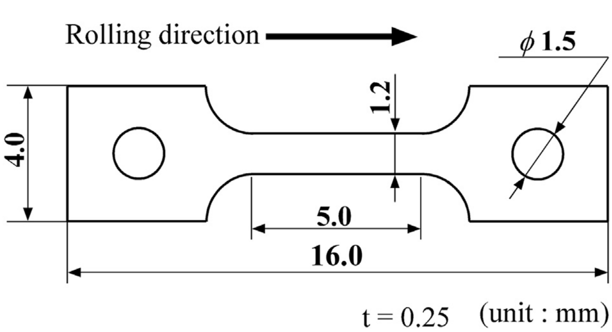

Fig. 1. Configuration and dimensions of the tensile testing specimens.

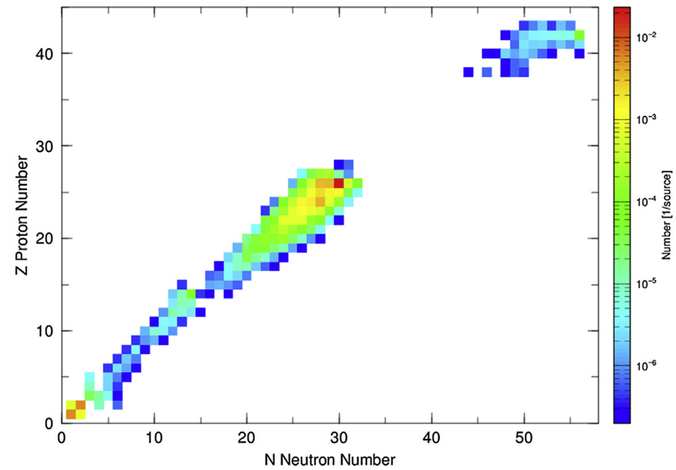

Fig. 2. Total production of secondary particles with *Z* protons and *N* neutrons in the 150 MeV proton-irradiated Fe–Cr alloy.

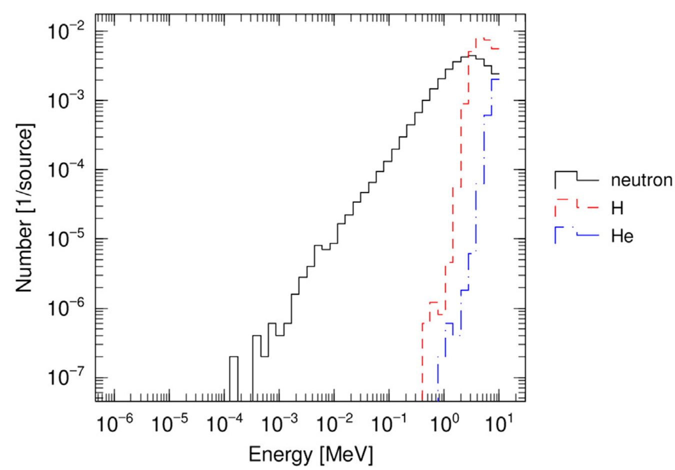

Fig. 3. Energy spectra of generated neutron, hydrogen and helium in the 150 MeV proton-irradiated Fe–Cr alloy.

during irradiation was monitored by a thermocouple mounted on an aluminum block, where samples were fixed using a 0.2 mm-thick aluminum cover.

In the case of irradiation with 11 MeV protons, the proton flux and the total fluence were 7.3 × 10^{11} p/cm^2/s and 6.0 × 10^{16} p/cm^2, respectively. The beam current was monitored by a current transformer. The stability of the current was within ±3%. As the hydrogen and damage distributions were not homogeneous along the beam incident direction, the sample was covered with a Fe–9Cr dummy plate of 0.125 mm thickness, and the damage and hydrogen peaks were set in the center of the 0.25 mm-thick sample. The total damage and the hydrogen concentration averaged in the sample, calculated through the Stopping and Range of Ions in Matter (SRIM) code [19], were 9.0 × 10^{-4} dpa and 2.8 × 10^{-5}, respectively. The temperature during the irradiation was monitored by a thermocouple mounted on a copper block, to which samples were mounted as described above.

Positron annihilation spectroscopy (PAS) was performed to identify the defects in the samples after proton irradiation. ^{22}Na (0.7 MBq) sealed with a Kapton film (7 μm in thickness) was used as the positron source, set between two experimental samples. The system employed for the positron annihilation lifetime measurements was a conventional fast–fast circuit with two BaF_2