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Use of double and triple-ion irradiation to study the influence of high levels of helium and hydrogen on void swelling of 8–12% Cr ferritic-martensitic steels

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A B S T R A C T

In accelerator-driven spallation (ADS) devices, some of the structural materials will be exposed to intense fluxes of very high energy protons and neutrons, producing not only displacement damage, but very high levels of helium and hydrogen. Unlike fission flux-spectra where most helium and hydrogen are generated by transmutation in nickel and only secondarily in iron or chromium, gas production in ADS flux-spectra are rather insensitive to alloy composition, such that Fe–Cr base ferritic alloys also generate very large gas levels. While ferritic alloys are known to swell less than austenitic alloys in fission spectra, there is a concern that high gas levels in fusion and especially ADS facilities may strongly accelerate void swelling in ferritic alloys. In this study of void swelling in response to helium and hydrogen generation, irradiation was conducted on three ferritic-martensitic steels using the Electrostatic Accelerator with External Injector (ESUVI) facility that can easily produce any combination of helium to dpa and/or hydrogen to dpa ratios. Irradiation was conducted under single, dual and triple beam modes using 1.8 MeV Cr^{+3}, 40 keV He^{+}, and 20 keV H^{+}. In the first part of this study we investigated the response of dual-phase EP-450 to variations in He/dpa and H/dpa ratio, focusing first on dual ion studies and then triple ion studies, showing that there is a diminishing influence on swelling with increasing total gas content. In the second part we investigated the relative response of three alloys spanning a range of starting microstructure and composition. In addition to observing various synergisms between He and H, the most important conclusion was that the tempered martensite phase, known to lag behind the ferrite phase in swelling in the absence of gases, loses much of its resistance to void nucleation when irradiated at large gas/dpa levels.

1. Introduction

Various austenitic, ferritic or ferritic-martensitic steels are potential alloys for structural materials not only in fission and fusion environments, but also in accelerator-driven spallation (ADS) devices used for transmutation of long-lived fission products, especially actinides. In ADS systems especially, the target and window components are particularly susceptible to strong direct radiation damage, as well as thermal shock, erosion and corrosion problems [1–4].

Similar issues arise in fission and fusion devices, but production of transmutant gases is generally lower compared to those of ADS systems [5].

The response of structural materials to displacive irradiation is more complicated when transmutation or spallation reactions produce significant levels of helium and hydrogen, gases known to affect not only the mechanical properties, but especially to accelerate void swelling. Gas production in fission and fusion spectra are not only sensitive to details of the neutron spectra, with higher production rates in fusion spectra, but are also sensitive to the alloy composition, with nickel being the major source of both gases in Fe–Ni–Cr alloys [6]. In iron-base alloys not containing nickel, gas production rates per dpa are lowest for fast reactors, increasing

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with neutron mean energy, followed by mixed-spectrum water-cooled reactors, and then by fusion reactors.

In spallation spectra, however, the incident high energy protons and the neutrons produced during spallation yield large gas production rates that are relatively insensitive to composition, with iron and chromium producing these gases at roughly the same rates as nickel [7–11]. In general, while both particles participate in displacement and gas production, the neutrons produce the majority of the displacement damage and the protons produce the majority of the transmutant gases. The production rates in ADS facilities, while much higher than in fusion devices, are somewhat device-specific, however, varying with the incident proton energy, target composition, target geometry and the coolant used, thereby producing different spectra and ratios of neutrons to protons, which generate different amounts of helium and hydrogen per dpa. The generation rates can range from ~40 to ~150 appm He per dpa, usually with hydrogen generation rates that are an order of magnitude higher.

Due to their lower void swelling compared to that of austenitic steels, ferritic and ferritic-martensitic steels are candidates for fusion and ADS environments to reach higher damage levels than can be attained by austenitics [12]. Not only are ferritic steels inherently lower swelling in fission and fusion spectra, but less helium and hydrogen are generated due to the low levels or absence of nickel in these steels. However, in ADS environments, these gases are generated at high rates even in the absence of nickel, leading to the concern that the inherent swelling resistance of ferritic alloys will be overwhelmed. Helium is well known to assist void nucleation and thereby accelerate the onset of void swelling during the incubation period. Until recently, hydrogen was thought to be too mobile to accumulate in the alloy matrix and thereby was unable to strongly influence void nucleation. However, hydrogen is now known to be strongly captured and stored in helium-nucleated or argon-nucleated voids or bubbles, thereby contributing to cavity stabilization [13,14]. Furthermore, in some alloys co-injection of helium and hydrogen appears to interact synergistically to strongly promote swelling [15,16].

To address this concern we have conducted ion irradiations over a wide range of gas/dpa ratios to assess the possibility that helium and hydrogen, separately or perhaps synergistically, influence the development of void swelling in ferritic and ferritic-martensitic steels. Steels with 8–12% Cr were chosen for investigation because of their excellent response to neutron irradiation compared with conventional Cr–Mo steels [17–20]. As an alloy class, significant amounts of data have been generated for these alloys under fission neutron irradiation, but much less for ADS conditions [21–23].

In the current study three alloys were chosen to span a range of structural starting states: martensite (F82H), tempered martensite (EK-181) and dual phase ferrite and tempered martensite (EP-450). Russian alloy EK-181 (Rusfer) and Japanese alloy F82H are developmental low-activation steels. EP-450 is a well-established Russian alloy with radiation experience in a number of Russian or Kazakh fast reactors, and is routinely used as fuel assembly wrappers in the BN-600 fast reactor.

2. Experimental details

2.1. Preparation of specimens

The chemical compositions and heat treatment conditions of the three alloys are listed in Table 1. The specimens were standard 3 mm diameter microscopy disks of 0.2 mm thickness. Full details of the preparation procedure has been described previously [24], but briefly the procedure involves electrochemically polishing the ion-incident surface to insure no mechanical damage was present. Following irradiation a pulse electropolishing procedure was used to remove 100 ± 5 nm from the ion-incident surface. While protecting this surface, the back surface was electropolished until perforation occurred at the front surface to yield electron-transparent areas for transmission microscopy of the 100–200 nm depth.

2.2. Irradiation of specimens

Irradiation was carried out on the ESUVI electrostatic heavy-ion accelerator located at Kharkov Institute of Physics and Technology. The design and main parameters of the facility have been presented previously [24–27]. Irradiations were performed using 1.8 MeV Cr^{3+} at the temperature of maximum swelling as determined for each material during earlier studies not involving gas implantation (450 and 480 °C).

A unique feature of ESUV1 is that it uses a hollow guide source located toward the end of the beam tube to inject and accelerate helium and/or hydrogen gas, producing a single-beam, three-ion source with fully adjustable gas/dpa ratios. Accelerating He^{+} and H_2^{+} to 40 keV deposits He and H at the depth region chosen for microscopy. The H_2^{+} dissociates when it crosses the specimen surface, producing two 20 keV H^{+} ions that have essentially the same range as the He ion. Note that in this paper we focus on only two dose levels, 50 and 200 dpa, which were chosen to represent the incubation stage of swelling under ADS conditions and the steady-state stage of swelling under fission and some fusion conditions.

The primary emphasis of this study is on the 50 dpa level, however, reflecting the recognition that swelling-resistant alloys primarily are very resistant to void nucleation, much more so than void growth, and that gas co-implantation is one of the most effective ways to overcome nucleation resistance. Additionally, stronger emphasis was placed on gas effects in dual-phase EP-450, reflecting its wider utilization and greater radiation data base.

Table 1
Chemical compositions of steels (wt%).

Steel (structure)	Type of precipitates	Chemical composition, wt%									
		C	Cr	W	Si	Mn	Mo	V	Nb	Ni	Other
EP-450 (ferrite + tempered martensite)	$M_{23}C_6$, MC	0.10–0.15	11.0–13.5	–	0.6	0.6	1.2–1.8	0.1–0.3	0.25–0.55	0.30	B-0.004
F82H (martensite)	$M_{23}C_6$, MC	0.10	7.44	2.0	0.14	0.5	–	0.20	–	–	Ta-0.04, N-0.002, P-0.001, S-0.001, Al-0.019
EK-181 (tempered martensite)	$M_{23}C_6$, M_6C, M_3C, TaC, VC	0.14	11.2	1.17	0.37	0.94	0.04	0.29	0.01	0.03	Ta-0.17, N-0.04, P-0.01, B-0.006, Co-0.01

EP-450: 1050 °C/0.5 h + 720 °C/1 h.
F82H: 1040 °C/0.5 h + 740 °C/2 h.
EK-181: 1100 °C/1 h + 720 °C/0.5 h.