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TEM characterization of irradiated microstructure of Fe-9%Cr ODS and ferritic-martensitic alloys

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ABSTRACT

The objective of this study is to evaluate the effects of irradiation dose and dose rate on defect cluster (i.e. dislocation loops and voids) evolution in a model Fe-9%Cr oxide dispersion strengthened steel and commercial ferritic-martensitic steels HCM12A and HT9. Complimentary irradiations using Fe^{2+} ions, protons, or neutrons to doses ranging from 1 to 100 displacements per atom (dpa) at 500 °C are conducted on each alloy. The irradiated microstructures are characterized using transmission electron microscopy (TEM). Dislocation loops exhibit limited growth after 1 dpa upon Fe^{2+} and proton irradiation, while any voids observed are small and sparse. The average size and number density of loops are statistically invariant between Fe^{2+}, proton, and neutron irradiated specimens at otherwise fixed irradiation conditions of ~3 dpa, 500 °C. Therefore, we conclude that higher dose rate charged particle irradiations can reproduce the neutron irradiated loop microstructure with temperature shift governed by the invariance theory; this temperature shift is ~0 °C for the high sink strength alloys studied herein.

1. Introduction & background

Ferritic-martensitic (F/M) alloys [1–9] and oxide dispersion strengthened (ODS) steels [4,10–18] are considered leading candidates as structural and fuel cladding components in advanced nuclear reactor designs, primarily due to their strength and dimensional stability under irradiation. However, these new reactor concepts will demand such components to withstand extreme conditions of irradiation damage up to several hundred displacements per atom (dpa) at temperatures as high as 700 °C [19–21]. To validate materials for these environments, the long-term irradiation resistance of these alloys needs to be well understood. Unfortunately, neutron irradiation experiments are time-consuming (10 + years in a fast neutron spectrum to accumulate up to 100 dpa), extremely costly, and specimens become highly activated and thus difficult to handle and characterize. In order to accelerate the evaluation process for F/M and ODS alloys, charged particles are increasingly being used to emulate neutron irradiations [22]. Charged particle irradiations allow one to conduct irradiation experiments within a shorter time period (i.e. over a few hours or days) and typically cause little to no activation of the material, enabling lower cost and faster turnaround in post irradiation examination and analysis.

Historically, ion irradiation conditions have been selected to emulate a defined neutron irradiation condition based on the invariance theory [23], which purports that the accelerated irradiation damage rate from ions can be offset with temperature adjustments, to produce equivalent void microstructures. Recent studies using modern computational techniques [24,25], have corroborated the invariance theory. However, experimental evidence for the role of temperature and damage rate specifically in F/M and ODS alloy microstructure evolution is somewhat limited because of the challenge of comparing neutron to ion irradiations on identical heats of archival alloys. One of the seminal studies on this topic has been conducted by Was et al. [26], who characterized the F/M alloy HT9 under neutron and ion irradiation conditions. Was' irradiation temperatures are selected based on the invariance theory; 5 MeV Fe^{2+} self-ion irradiations are carried out to 188 dpa at 460 °C to attempt to emulate 155 dpa, 443 °C neutron irradiation. While Was et al. [26] is able to obtain a relatively consistent void morphology between neutron and ion irradiations, the G-phase and dislocation loop morphologies are markedly different between

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neutron and ion irradiations. Ions produce smaller G-phases at a higher number density than do neutrons. The opposite is observed with dislocation loops: ions produce larger loops at a lower number density, although the resultant total loop line length is relatively constant between the irradiation types. Thus, there remains a need to understand the implications of the invariance theory – and more broadly, dose rate effects – on microstructure evolution.

Irradiated microstructure features can generally be classified as either below or above transmission electron microscopic (TEM) resolution. The former are features such as nanoclusters, including oxide nanoparticles in ODS and G-phase and Cu-rich nanoclusters in F/M alloys. These features are more effectively characterized by approaches such as atom probe tomography, than by TEM. In a recent paper [27], we explore the concepts of dose rate and temperature shift with respect to these nanoclusters. In this paper, we now focus on those features that are above TEM resolution, namely defect clusters such as voids and dislocation loops.

Considerable research efforts have focused on void evolution, especially at very high doses (i.e. >100 dpa), in F/M alloys; average void sizes from these studies are summarized in Fig. 1a [6,28,29]. The most comprehensive study of void dose dependence has been conducted by Getto et al. [28] on F/M alloys HT9, T91, and T92 irradiated with 5 MeV Fe^{2+} self-ions (at 460 °C). In this study, voids appear in HT9, T91, and T92 between 75 and 130 dpa, then exhibit typical exponential growth with increasing irradiation dose up to 650 dpa. Of the three alloys, voids tend to be smallest in T92, while voids in T91 and HT9 are nearly the same size. A similar study by Getto et al. [29] on HT9 irradiated with 5 MeV Fe^{2+} ions at 440 °C and 480 °C exhibited similar trends. In comparing these two studies on HT9, void size exhibits a bell-shaped dependence, with maximum void size observed at 460 °C compared to smaller voids at both 440 °C and 480 °C. Anderoglu et al. [8] and Sencer et al. [6] also study neutron irradiated HT9 at doses as high as 155 dpa at temperatures ranging 380–505 °C; they find voids only after irradiation to 155 dpa at 443 °C (28 nm average).

In ODS alloys, voids are not frequently reported in the archival literature. Even the absence of voids is rarely noted; one such study, Ribis [30], reports no cavities in MA957 after 50–75 dpa neutron irradiation in PHENIX at 412 °C and 430 °C. Two studies observe voids forming at the oxide-matrix interface in Fe^{2+} irradiated MA956 at 450 °C [31] and neutron irradiated EUROFER-ODS at 350 °C [32]. Finally, the most comprehensive study of voids in ODS is that of Toloczko et al. [33], which evaluates the roles of both temperature and dose using 1.8 MeV Cr^{+} ion irradiations of MA957. Similar trends can be gleaned as from the commercial F/M alloys: bell-shaped temperature dependences for both size and number density are observed with a maxima at 450 °C, and void sizes increase with dose from 100 to 500 dpa.

Dislocation loop evolution has also been reported extensively in the archival literature; a visual summary of average loop sizes is provided in Fig. 1b for F/M alloys [6,28,29,34]. Overall, dislocation loops in the F/M alloys are consistently <25 nm in average diameter at doses at or below ~150 dpa, regardless of irradiation temperature. Once again, the most comprehensive study of dose dependence in F/M alloys is by Getto et al. [28] on HT9, T91, and T92 irradiated at 460 °C with 5 MeV Fe^{2+} ions. In Getto's study, dislocation loop sizes appear to reach a semi-steady-state after doses ranging 188–450 dpa (23–32 nm), then dramatically increase in size after 550 dpa (~78 nm). To the authors' knowledge, few studies have reported quantitative dislocation loop data in ODS alloys as most research has focused on the evolution of the oxide nanoclusters. There are quantitative reports of loops following 400 °C proton irradiation of Fe-9%Cr ODS [15], 400 °C Fe^{3+} irradiation of EUROFER-ODS [35], and 350 °C fast neutron irradiation of EUROFER-ODS [32]. However, there is insufficient evidence from which to draw conclusions about the effects of dose rate.

The archival literature sheds light on the effects of irradiation dose and temperature on void and loop evolution, but because most of these studies have utilized the invariance theory to define ion irradiation conditions, there remains a knowledge gap on the effects of dose rate. In this work, instead of utilizing the invariance theory, we conduct 10^{-7} dpa/sec (fast neutron), 10^{-5} dpa/sec (proton), and 10^{-4} dpa/sec (Fe^{2+} self-ion) irradiations at a fixed damage level of ~3 dpa and a fixed temperature of 500 °C. Such an experimental design enables us to isolate the influence of the irradiating particle and dose rate on defect cluster (i.e. dislocation loops and voids) evolution in a model Fe-9%Cr ODS steel and the commercial F/M alloys HCM12A and HT9. TEM and scanning transmission electron microscopy (STEM) techniques are used to characterize the irradiated microstructure. New results presented herein are aggregated with previous TEM/STEM results from the same ODS heat subject to different irradiation conditions, reported by Swenson et al. [36] and Yano et al. [37]. These collective results enable a more comprehensive understanding of dose rate dependence and application of the invariance theory.

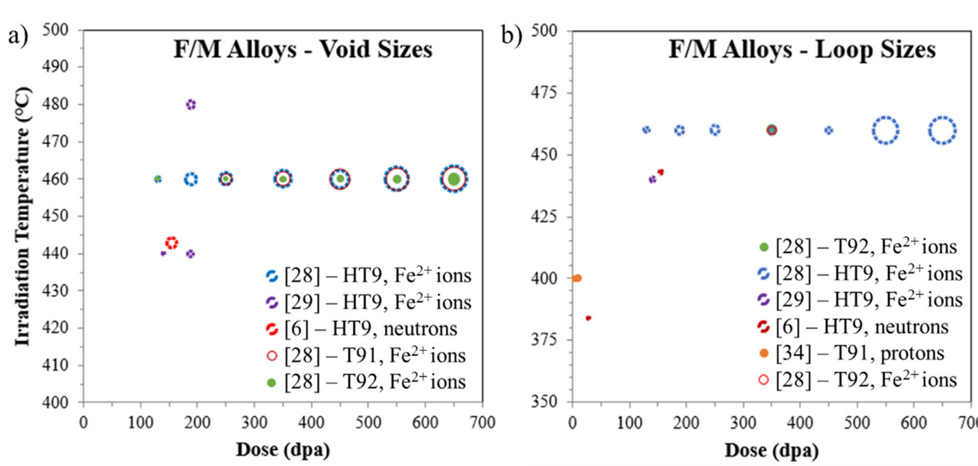

Fig. 1. Results from archival literature for F/M alloys showing a) average size of irradiation-induced voids, and b) average size of irradiation-induced dislocation loops. Size of bubbles represent relative size of features.