Acta Materialia 136 (2017) 390–401

Contents lists available at ScienceDirect

Acta Materialia

journal homepage: www.elsevier.com/locate/actamat

Full length article

Dislocation loop evolution during in-situ ion irradiation of model FeCrAl alloys☆

Jack C. Haley [a,*], Samuel A. Briggs [b], Philip D. Edmondson [c], Kumar Sridharan [b], Steve G. Roberts [a], Sergio Lozano-Perez [a], Kevin G. Field [c]

[a] University of Oxford, Oxford OX1 3PH, United Kingdom
[b] University of Wisconsin–Madison, Madison, WI 53706, USA
[c] Oak Ridge National Laboratory, 1 Bethel Valley Rd, Oak Ridge, TN 37831, USA

ARTICLE INFO

Article history:
Received 11 March 2017
Received in revised form
5 May 2017
Accepted 5 July 2017
Available online 6 July 2017

Keywords:
FeCrAl
Dislocation loops
Radiation damage
In-situ
Accident tolerant

ABSTRACT

Model FeCrAl alloys of Fe-10%Cr-5%Al, Fe-12%Cr-4.5%Al, Fe-15%Cr-4%Al, and Fe-18%Cr-3%Al (in wt %) were irradiated with 1 MeV Kr^{++} ions in-situ with transmission electron microscopy to a dose of 2.5 displacements per atom (dpa) at 320 °C. In all cases, the microstructural damage consisted of dislocation loops with ½⟨111⟩ and ⟨100⟩ Burgers vectors. The proportion of ½⟨111⟩ dislocation loops varied from ~50% in the Fe-10%Cr-5%Al model alloy and the Fe-18Cr%-3%Al model alloy to a peak of ~80% in the model Fe-15%Cr-4.5%Al alloy. The dislocation loop volume density increased with dose for all alloys and showed signs of approaching an upper limit. The total loop populations at 2.5 dpa had a slight (and possibly insignificant) decline as the chromium content was increased from 10 to 15 wt %, but the Fe-18%Cr-3%Al alloy had a dislocation loop population ~50% smaller than the other model alloys. The largest dislocation loops in each alloy had image sizes of close to 20 nm in the micrographs, and the median diameters for all alloys ranged from 6 to 8 nm. Nature analysis by the inside-outside method indicated most dislocation loops were interstitial type.

1. Introduction

Iron-chromium-aluminium (FeCrAl) alloys are an emerging class of Fe-based, high-Cr ferritic alloys for use in nuclear power generation. In particular, FeCrAl alloys are promising candidates as a fuel cladding in commercial light water reactors (LWRs) that can exhibit enhanced accident tolerance compared with the zirconium-based alloys in use today [1,2]. The perceived accident tolerance of FeCrAl alloys is predicated on FeCrAl alloys exhibiting oxidation kinetics more than 100 × slower than those of zirconium-based alloys in high-temperature steam environments (>1000 °C) [2,3]. The slower oxidation kinetics lead to lower rates of heat and hydrogen production in the reactor core under accident scenarios, resulting in additional coping time for active mitigation strategies [2]. Although FeCrAl alloys' oxidation resistance lends itself to high performance during specific accident scenarios, such as loss-of-coolant accidents, FeCrAl alloys could be susceptible to in-core degradation during normal operation. Degradation could be the result of the harsh environment of commercial LWRs, including elevated pressures/stresses, high-temperature water (288–320 °C, nominally), and neutron radiation. The combination of elevated temperature and neutron radiation is a concern, as it may lead to significant radiation-induced hardening and embrittlement effects. Significant embrittlement could ultimately limit the lifetime of FeCrAl alloys in LWR cladding applications.

Initial work on neutron-irradiated model FeCrAl alloys in a materials test reactor investigated the magnitude of radiation-induced hardening and embrittlement [4]. These studies, through

☆ This manuscript has been authored by UT-Battelle, LLC under Contract No. DE-AC05-00OR22725 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes. The Department of Energy will provide public access to these results of federally sponsored research in accordance with the DOE Public Access Plan (http://energy.gov/downloads/doe-public-access-plan).

* Corresponding author. Department of Materials, University of Oxford, 16 Parks Road, Oxford, OX1 3PH, United Kingdom.

E-mail addresses: jack.haley@trinity.ox.ac.uk (J.C. Haley), sabriggs2@wisc.edu (S.A. Briggs), edmondsonpd@ornl.gov (P.D. Edmondson), kumar@engr.wisc.edu (K. Sridharan), steve.roberts@materials.ox.ac.uk (S.G. Roberts), sergio.lozano-perez@materials.ox.ac.uk (S. Lozano-Perez), fieldkg@ornl.gov (K.G. Field).

http://dx.doi.org/10.1016/j.actamat.2017.07.011
1359-6454/

combined ex-situ structure-property evaluations, have determined the hardening and loss of ductility in this alloy class to be in part due to precipitation and dislocation loop formation. The precipitation and dislocation loop formation can be directly linked to the radiation damage processes, including radiation-enhanced diffusion and defect agglomeration in these alloys.

The development and eventual optimization of FeCrAl alloys for nuclear applications would require detailed insight into these fundamental radiation damage processes and any subsequent defect production and evolution. Unfortunately, materials test reactor irradiations do not lend themselves to detailed studies of these fundamental processes owing to their limited capability for active control of experimental parameters. For example, the same region of interest cannot be studied under increasing dose or irradiation temperature; and it is well known that microstructural variables such as grain size, prior existence of dislocations, or other local defect sinks can locally alter the precipitation and dislocation loop formation in metals. Furthermore, in-reactor temperature control is inherently difficult; this was especially the case in previous studies of FeCrAl alloys [4] in which temperature control was passively determined by the sample holder–capsule diametrical gap. Finally, radiation dose and dose rate can vary greatly depending on the axial and/or radial location within the material test reactor core.

Ion irradiations have proven to show significantly better control of experimental variables, compared with materials test reactor irradiations, including irradiation dose, dose rate, and temperature. Additionally, in-situ ion irradiation performed within a transmission electron microscope (TEM) on thin foils enables the determination of dynamical processes within a single region of interest, eliminating (or characterising) grain-to-grain variations, among many other variables typical of ex-situ studies. This results in the capability to design experiments in which parameters can be selected and closely controlled, with only one varying during an experiment (e.g., total dose); an ideal situation for examining the fundamental aspects of radiation damage in a specific material system, including defect nucleation and growth. Additionally, the correlation of single-parameter (or at least reduced-parameter) in-situ ion irradiations with modern modelling and simulation techniques provides an opportunity to extend the value of such experiments and modelling.

Preliminary work has already been performed on correlative studies of fundamental radiation damage aspects in Fe and FeCr alloys; for example, the impact of microstructure [5], chemistry [6–12], irradiation temperature [13,14] and dose [8,9] on defect formation. In particular, the dislocation loop production and evolution kinetics have been evaluated. It has been shown that dislocation loops formed in Fe and FeCr during irradiation have Burgers vectors of ⟨100⟩ or ½⟨111⟩, in contrast to other body-centred-cubic (BCC) metals such as molybdenum, tungsten, and vanadium, which contain almost exclusively ½⟨111⟩ dislocation loops. Understanding the mechanisms behind the formation of ⟨100⟩ dislocation loops is important, as they are a more resistant barrier to dislocation slip than ½⟨111⟩ dislocation loops [4,15] and therefore have a greater potential for hardening. It has been shown experimentally [13] that the fraction of ⟨100⟩ dislocation loops is higher at higher temperatures; Dudarev et al. [14] showed theoretically how the stability of ⟨100⟩ and ½⟨111⟩ dislocation loops depends greatly on temperature, which helps to explain experimental observations of a decreasing fractional population of ½⟨111⟩ loops with increasing temperature in Fe [13]. Monte Carlo–based simulations by Xu et al. [16] of coalescing ½⟨111⟩ dislocation loops have shown that new ⟨100⟩ dislocation loops can emerge from such an interaction. Such a formation process would be heavily dependent on the frequency of loop coalescence, which in turn is highly dependent on dose, dose rate, temperature, composition, and impurity content. From a combination of thin foil [7,8] and bulk-material irradiations [17–19] of FeCr, it has been shown that the proportion of ⟨100⟩ dislocation loops after irradiation declines with increasing Cr content (corresponding to decreasing ½⟨111⟩ loop mobility [16]).

This paper discusses an investigation of the details of radiation-induced dislocation loop formation and evolution in model FeCrAl alloys by using ion irradiation coupled with in-situ and ex-situ TEM. This work provides a fundamental insight into how composition affects nucleation and growth of dislocation loops in these alloys, which are contributors to radiation-induced hardening [20]. Ultimately, design of a cladding alloy requires tailoring the composition and microstructure to balance performance with respect to radiation hardening/embrittlement (both from loops and precipitates), oxidation/corrosion, high temperature mechanical properties, component fabricability, and neutronics. As such, this study directly informs design with regards to one of the many performance metrics for a candidate alloy. However, it should be noted, neutron irradiation studies [20] have shown α' phases to be a dominant hardening mechanism in these alloys, and hence although this study provides insight on the loop formation, it does not provide a full picture of the features contributing to hardening in the alloys or other performance metrics for these alloys as cladding material.

Experimental conditions were specifically tailored to provide insight into the role of composition in dislocation loop formation and evaluation within the low-dose regime, thereby serving as a guide toward compositional control for radiation tolerance in FeCrAl alloys. A set of well-annealed model FeCrAl alloys with a yttrium addition, that reside near the kinetic boundary for passivation in 1200 °C steam environments [1], was used. The absence of significant dislocation densities, internal defect sinks, and minor alloying elements, such as molybdenum and silicon, reduced the complexities in this study. A site-specific focused ion beam (FIB) lift-out technique was used to produce large thin areas of relatively uniform thickness (on the order of 25–100 μm^2) orientated with a ⟨100⟩ pole aligned normal to the foil surface. These samples were irradiated at the Intermediate Voltage Electron Microscopy (IVEM)–Tandem Facility (Argonne National Laboratory) with 1 MeV Kr^{++} ions at 320 °C up to damage doses of 2.5 displacements per atom (dpa). The temperature was selected to represent commercial reactor conditions for fuel cladding in LWRs. Although a temperature shift such as the one proposed by Mansur [21] could have been used to correlate between irradiations of different dose rates, it was not used here because of the absence of a strictly established dose-temperature shift for dislocation loop formation in FeCrAl alloys. The end dose of 2.5 dpa represents early-life doses expected for a typical commercial fuel cladding and was found to reside well within the coarsening regime of the dislocation loop evolution.

2. Experiment

Four model FeCrAl alloys with nominal compositions of Fe-10%Cr-5%Al, Fe-12%Cr-4.5%Al, Fe-15%Cr-4%Al, and Fe-18%Cr-3%Al (in wt%) with designations of F1C5AY, B125Y, B154Y-2, and B183Y-2 in Refs. [4,22–25] were studied. The full compositions, including impurity contents, are provided in Table 1. The alloys span a wide composition range of model FeCrAl alloys shown to be protective in high-temperature steam at up to 1200 °C [1] and for which a wealth of base materials properties and radiation performance data exist [4,22–25]. The base feedstock for each alloy was fabricated by arc-melting and drop casting to prepare bar-shape ingots with a size of 13 × 25 × 125 mm. The as-cast ingots were homogenized at 1200 °C for 2 h in an argon gas atmosphere and then water-quenched. The ingots were hot-rolled at 700 °C with a 10%