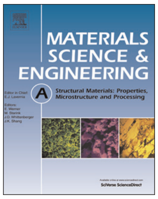
Materials Science & Engineering A 639 (2015) 585–596

Contents lists available at ScienceDirect

Materials Science & Engineering A

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



On the microstructure and strengthening mechanism in oxide dispersion-strengthened 316 steel: A coordinated electron microscopy, atom probe tomography and *in situ* synchrotron tensile investigation

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ARTICLE INFO

Article history:
Received 2 April 2015
Received in revised form
19 May 2015
Accepted 21 May 2015
Available online 27 May 2015

Keywords:
Oxide dispersion-strengthened steel
Atom probe tomography
Scanning transmission electron microscopy
Synchrotron X-ray scattering

ABSTRACT

An oxide dispersion-strengthened (ODS) 316 steel was developed to simultaneously provide the advantages of ODS steels in mechanical strength and radiation tolerance as well as the excellence of austenitic steels in creep performance and corrosion resistance. The precipitate phases within the austenite matrix were identified by the combined techniques of atom probe tomography (APT), scanning transmission electron microscopy equipped with electron dispersive X-ray spectroscopy (STEM-EDS), and synchrotron wide-angle and small-angle X-ray scattering (WAXS and SAXS). Coarse TiN, hexagonal $YAlO_3$ and orthorhombic $YAlO_3$ precipitates were found along with fine Y–Ti–O nanoparticles. *In situ* WAXS experiments were performed at room and elevated temperatures to examine the size effect on the load partitioning phenomenon for TiN, hexagonal $YAlO_3$ and $Y_2Ti_2O_7$ phases. In addition, the dislocation density evolution throughout the tensile tests was analyzed by the modified Williamson–Hall method and confirmed by transmission electron microscopy (TEM) observations, revealing the difference in plasticity at various temperatures.

1. Introduction

Materials challenges are among the most crucial factors that limit the advances of nuclear technology, since materials used in nuclear reactors are required to persistently maintain their performance in extreme environments including elevated temperature, aggressive coolant, high pressure, and intense neutron flux. The development of Generation IV nuclear fission reactors is facing significant difficulties in the selection of structural materials [1]. The novel designs of these nuclear reactors usually involve higher operating temperatures and advanced coolants, which demand excellent high temperature mechanical performance and corrosion resistance of the structural materials involved. The development of the supercritical water reactor, the only Generation IV design to employ water coolant, is facing challenges in the search for structural materials, especially in the fuel cladding material. A series of advanced materials, including austenite stainless steels, reduced activation ferritic/martensitic alloys (RAFM), ferritic/martensitic (F/M) oxide dispersion-strengthened (ODS) steels, nickel-based advanced alloys and refractory alloys, have been investigated as prospective candidates [2–5]. Recently, a number of F/M ODS steels have been developed and examined for several key properties necessary for the applications in nuclear reactors [6,7]. However, the F/M phase of Fe-based alloys, with its low Cr content of less than 9 wt%, has intrinsically poor corrosion resistance, which limits its applications in fuel cladding materials, especially in the supercritical water environment. A higher Cr content may improve the corrosion performance [8], but at the expense of introducing α–α' phase separation during thermal aging enhanced by neutron irradiation [9,10]. These Cr-enriched α' precipitates embrittle the material, degrading its ductility and toughness.

On the other hand, the advantages of austenitic stainless steels

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http://dx.doi.org/10.1016/j.msea.2015.05.064
0921-5093/

include their excellent creep performance and corrosion resistance, making them competitive candidates for fuel cladding materials. In addition, austenite is the stable phase at high temperatures, assuring reliability in accident conditions. Unfortunately, conventional austenitic steels suffer from severe radiation swelling [11], and are of relatively low mechanical strength when compared to ferritic steels. Both these disadvantages limit the application of austenitic steels to advanced reactors. Mechanical alloying followed by a series of heat treatment and thermal processing procedures introduces into the steel matrix a dense and dispersed distribution of ultra-fine oxide nanoparticles that are quite stable at high temperatures [12] and under high irradiation conditions [13,14]. Previous studies of the ferritic ODS steels clearly show that the ultra-fine oxide nanoparticles as well as the reduced grain size enhance the mechanical strength by pinning dislocations [15]. In addition, the extra interfaces created by the nanoparticles can act as sinks for point defects generated by irradiation and even attract helium atoms produced by (n, α) reactions [16–19]. Thus, both the irradiation-induced dislocation loop evolution and radiation swelling can be suppressed. Therefore, oxide nanoparticles coupled with a finer grain structure are expected to give austenitic steels excellent radiation tolerance and mechanical strength with marginal compromise in the intrinsic advantages of austenitic steels. A series of ODS austenitic steels have been developed and proven to display the excellent conditions mentioned above [20–25].

The characteristics of the oxide nanoparticles, including their size distribution, chemical composition, morphology, orientation relationship, and interaction mechanisms with the dislocations, are crucial properties that influence the performance of the ODS austenitic steels as nuclear structural materials. Scanning transmission electron microscopy (STEM), equipped with a high angle annular dark field (HAADF) detector, is capable of yielding image contrast that is proportional to the square of the atomic numbers (Z^2) of the materials, and is therefore called Z-contrast. This technique can be used to identify the precipitates in ODS steels. More importantly, embedded with the spectroscopic capabilities of electron dispersive X-ray spectroscopy (EDS) and electron energy loss spectroscopy (EELS), STEM is capable of providing additional chemical information of these precipitates [26–28]. However, the resolution limitation of conventional STEM restricts its application to the investigation of nanoparticles greater than 3 nm in diameter.

Atom probe tomography (APT), which has been developed in the past decades based on the previous success of atom probe field ion microscopy (APFIM), is able to collect over 37% of the atoms in a sample containing up to billions of atoms by identifying information on the atom types and reconstructing the atom positions based on a high efficiency position sensitive time-of-flight (ToF) mass spectroscopy detector. Therefore, this technique is an ideal complement to STEM in examining nanoparticles smaller than 3 nm [29]. Although the development of advanced APT algorithms recently realized the reconstruction of the lattice structure by means of 3D Hough transformations [30], it is currently still unlikely that the crystal structure of the nanoscale precipitates and their orientation relationships with the matrix can be distinguished, especially when the laser mode has to be adopted due to the poor thermal and electric conductivities of the specimens [24]. STEM is capable of providing sub-Å, or atomic resolution with a spherical aberration (C_s) correction, and has already been used to explore the morphology of the oxide nanoparticles in ODS steels [31]. However, the results are not explicit and therefore require interpretation using image simulation techniques. On the contrary, high-resolution transmission electron microscopy (HRTEM) provides phase contrast images with atomic resolution, and has been successfully applied to crystal structure and orientation relationship analyses of ODS steels [32–34,23,24].

With ultra-high energy X-rays of high intensity, synchrotron light sources offer the possibility of collecting scattering information from phases with marginal volume fractions that are unlikely to be captured by conventional X-ray sources [24]. Wide-angle X-ray scattering (WAXS) can produce diffraction information of precipitates for phase identification, whereas small-angle X-ray scattering (SAXS) can be used to analyze the size distribution of the ultra-fine precipitates [35].

The oxide nanoparticles in ODS steels enhance the mechanical strength of the material by interacting with dislocations and suppressing grain growth. The nanoparticle–dislocation interactions result in pileups of dislocations on the nanoparticles. With dislocations restraining them, the precipitates have larger lattice strains than those of the matrix and therefore can bear much more load during plastic deformation. This load partitioning phenomenon is the foundation of precipitate-strengthening mechanism. Being capable of measuring the lattice spacing of precipitate phases, WAXS investigation can be utilized to monitor the lattice strain evolution of the precipitates during tensile tests. Hence, *in situ* WAXS tensile experiments are powerful tools to explore the load partitioning phenomenon. In fact, this technique has been employed to investigate the strengthening mechanisms in a variety of precipitate-strengthened materials including ferrite or martensite steels [36–39], Ni-based superalloys [40], and ODS steels [7,41]. The success of these previous applications ensures the success of using this technique to investigate the contributions of the oxide nanoparticles to the mechanical strength enhancement of ODS 316 steel.

Another benefit is that the diffraction peaks in WAXS contain information on the grain size, dislocation density, and stacking fault/twinning portions within the phases of interest. Ungár developed a method to assess these quantities based on the diffraction peak broadening, which is call the modified Williamson–Hall (W–H) method [42]. With *in situ* experiment capabilities, this analysis technique can be applied to monitor the real-time alterations of microstructures within materials.

In this study, a coordinated combination of advanced microstructure characterization techniques was systematically utilized to retrieve comprehensive information including the size distribution, volume fraction, chemical composition, and crystal structure of all the precipitate phases, especially the oxide nanoparticles, existing in a recently developed ODS 316 stainless steel [43]. *In situ* synchrotron tensile investigations were then performed for the ODS 316 steel, allowing the load partitioning phenomenon as well as the dislocation density evolution during the plastic deformation to be analyzed to help reveal the strengthening mechanism of austenitic ODS steels. The knowledge obtained in this research effort will expand the understanding of the origin of the outstanding mechanical strength of austenitic ODS steels, permitting examination of the feasibility of using austenite ODS steels for fuel cladding in future advanced reactor systems.

2. Description of experiments

2.1. Investigated material

The ODS stainless steel 316(LN) investigated in this study has the composition listed in Table 1. The base material powders were mechanically alloyed in an argon atmosphere using a planetary ball mill at 300 rpm with a ball-to-powder ratio of 5:1 for 30 h. The milled powders were then degassed, sealed, and consolidated by hot isostatic pressing (HIP) under a pressure of 100 MPa at 1150 °C for 3 h. The manufacturer reported that the ODS 316 steel