Journal of Nuclear Materials 512 (2018) 227–238

Contents lists available at ScienceDirect

Journal of Nuclear Materials

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

Influence of mechanical alloying and extrusion conditions on the microstructure and tensile properties of Low-Cr ODS FeCrAl alloys☆

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HIGHLIGHTS

- 10Cr ODS FeCrAl alloys exhibit higher ductility than 12Cr alloys with higher oxygen content.
- Zr solute additions sequester impurity C and N added during alloy fabrication.
- Alloy strengthening is well described through the root mean square (RMS) superposition model.

ARTICLE INFO

Article history:
Received 22 July 2018
Received in revised form
9 September 2018
Accepted 10 October 2018
Available online 11 October 2018

Keywords:
Oxide dispersion strengthened (ODS) alloy
Strengthening mechanisms
Precipitation
Mechanical properties

ABSTRACT

Low-chromium (<10%Cr) high strength oxide dispersion strengthened (ODS) FeCrAl alloys are considered promising candidates for accident tolerant fuel cladding in light water fission reactors. These alloys are designed to combine the beneficial high temperature mechanical properties of ODS materials with the exceptionally high temperature oxidation resistance of FeCrAl in comparison to conventional Zr-containing cladding materials, while also providing good mechanical and aqueous corrosion behavior under normal light water reactor operating conditions. The initial (1st generation) ODS FeCrAl alloys combined gas atomized FeCrAl powders with yttria and other oxides for alloying element additions using the mechanical alloying approach. These alloys exhibited low ductility but excellent high temperature tensile strength while maintaining good oxidation resistance at temperatures up to 1400 °C. In an attempt to improve alloy ductility for accident tolerant fuel cladding applications, new low-Cr ODS FeCrAl alloys with decreased oxygen content were developed with the Zr alloying solute already gas atomized into the powder prior to mechanical alloying. The resultant Fe-10Cr-6.1Al-0.3Zr+0.3Y_2O_3 (106ZY) powders were ball milled for 10, 20, and 40hr followed by consolidation via hot extrusion at temperatures ranging from 900 to 1050 °C. Increasing the mechanical alloying time decreased the resultant grain size and improved high temperature tensile properties. Decreasing the extrusion temperature refined the grain size and subsequently strengthened the ODS FeCrAl 106ZY alloys while lowering the ductility. Scanning transmission electron microscopy (STEM) and energy dispersive spectroscopy (EDS) demonstrated the Zr solute addition effectively sequestered impurity C and N within the matrix. The root mean square (RMS) hardening superposition model for yield strength shows good agreement with experimental results. Errors between predicted and experimental values are discussed within the scope of processing parameters. These 2nd generation 106ZY alloys show a distinct increase in alloy ductility without sacrificing the high temperature tensile properties characteristic of legacy or 1st generation ODS FeCrAl alloys.

☆ 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, worldwide 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).

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https://doi.org/10.1016/j.jnucmat.2018.10.017
0022-3115/© 2018 Published by Elsevier B.V.

1. Introduction

Since the devastating events at the Fukushima Daiichi nuclear reactor facility in 2011, research has been aimed at developing advanced materials that provide enhanced strength and oxidation resistance in the event of a nuclear accident scenario [1,2]. These accident tolerant fuel (ATF) cladding materials must outperform existing Zr-based alloys with respect to their oxidation resistance in steam and their subsequent hydrogen generation in the event of a design basis or beyond design basis accident, while retaining acceptable neutronic and corrosion performance under prolonged normal operating conditions. Furthermore, ATF cladding materials must exhibit many of the desirable qualities for fuel cladding applications under normal operating and accident conditions, such as having good mechanical strength and ductility, chemical compatibility with fuel and coolant selections, both thermal and irradiation stability, a minimal neutron absorption penalty, high thermal conductivity, and a sufficiently high melting temperature [2].

To address these challenges, high strength wrought FeCrAl alloys have been developed that exhibit 100 × lower oxidation kinetics in high temperature steam than currently used Zr-based claddings [3–5]. The presence of aluminum in the matrix promotes the growth of a stable aluminum oxide film at high temperatures while simultaneously allowing for a lower chromium content (≤10%Cr) in the alloys [6]. The lower Cr content is imperative due to the well-known irradiation induced precipitation of the deleterious and embrittling Cr-rich α' phase that occurs for higher Cr contents at the ~300–350 °C operating temperature for fuel cladding in existing light water reactors [7,8]. The higher strength of these FeCrAl alloys is imperative due to the added neutronic penalty of Fe in comparison to Zr in the reactor core, which will require a substantial thinning of existing cladding geometries to maintain efficient neutron economy [2]. Recent loss of coolant accident burst experiments on as-fabricated FeCrAl alloys have shown that the onset of burst for these Fe-based alloys is attributed to exceeding the high temperature ultimate tensile strength (UTS) for the alloy [9]. This suggests that further improving the UTS of existing FeCrAl alloys can prove beneficial not only for cladding thinning requirements for drop-in replacements for existing reactor assemblies, but it may also improve the burst properties of these cladding tubes in the event of a nuclear accident condition.

To improve the strength of existing FeCrAl alloys, ODS alloys utilize a high number density of nano-precipitates within the metal matrix that pin grain boundaries during alloy consolidation, leading to a refinement in grain size and a significant increase in both strength [10]. Nanoprecipitates within the as-consolidated alloy also inhibit dislocation motion and grain boundary sliding through a Zenner pinning effect, thus improving high temperature mechanical properties and creep resistance [11]. However, legacy ODS FeCrAl alloys developed originally for fossil applications such as MA956 and PM2000 are highly prone to the formation of α', Cr-rich precipitates due to their high Cr content (~20%Cr) [12–14]. In an attempt to reduce the precipitation of this Cr-rich phase, low-Cr (10–12% by weight) ODS FeCrAl alloys have been previously developed for fission and fusion applications by milling FeCrAl gas atomized powder with Y_2O_3 and either ZrO_2, TiO_2, or HfO_2 for the addition of alloying elements Zr, Ti, and Hf [15–17]. The resulting alloys exhibited beneficial oxidation resistance and a fine-grained microstructure with corresponding grain sizes ranging from 100 to 500 nm [15]. Though these alloys demonstrated exceptional tensile strength at high temperatures, the corresponding unirradiated total deformation ($\varepsilon_{tot} < 10\%$) was poor for the alloys at temperatures characteristic of light water reactor environments ($T < 400°C$). The high oxygen content of these 1st generation ODS FeCrAl alloys was of concern since previous studies on ODS FeCr alloys have suggested that the oxygen availability in the lattice strongly influences the types of nano-precipitates that can form during alloy consolidation [18,19].

In an attempt to more tightly control the oxygen content and to fabricate new alloys with better ductility, new 2nd Gen. ODS FeCrAl alloys with only 10% Cr by weight are being developed at ORNL. Instead of the addition of alloying elements as oxides during the mechanical alloying process, FeCrAl powders were procured with Zr already homogeneously gas atomized into the powder prior to adding Y_2O_3 during the ball milling stage. The addition of Zr to the metal matrix allows for the careful assessment of zirconium's role in the formation of precipitates in the resulting ODS alloys and also decreases the amount of oxygen in the ODS FeCrAl lattice. To optimize the processing methodology for the production of these ODS FeCrAl alloys, parametric evaluations of the ball milling durations and extrusion temperatures were performed. By varying the milling time, the impurity content can be balanced with the effectiveness of Y_2O_3 incorporation into the lattice. Lastly, the effect of the extrusion temperature on the mechanical properties of these alloys is presented with the specific goal of optimizing the strength and ductility of the resulting ODS FeCrAl alloy.

2. Methodology

2.1. Experimental approach

Gas atomized powder from ATI Powder Metals (mesh −100/+325 i.e. 44–149 μm particle size) with nominal composition Fe-10Cr-6.1Al-0.3Zr (all numeric values are in wt. %) was mixed with 0.3% by weight of nanocrystalline Y_2O_3 (17–31 nm crystallite size). The powder mixture was mechanically alloyed in a high energy Zoz Simoloyer CM01 ball mill in an Ar atmosphere using low-C steel milling media with a ball to powder ratio of 10:1. The milling was performed using rotational speeds of 400 rpm/900 rpm for either 10, 20, or 40 h. The resulting Fe-10Cr-6.1Al-0.3Zr+0.3Y_2O_3 (106ZY) ODS FeCrAl powders were packed and degassed for 24 h at 300 °C in a 4.83 cm (1.9") extrusion can. The 106ZY powders were extruded at 950 °C through a circular 2.22 cm (7/8") die. Furthermore, the 106ZY powder milled for 40hr was also extruded at temperatures 900 °C, 1000 °C, and 1050 °C to assess the temperature dependence of microstructural and mechanical properties for the alloy.

The chemical compositions of the various extruded alloys were measured using inductively coupled plasma optical emission spectroscopy (ICP-OES) and combustion analysis by DIRATS. The results of this compositional analysis are summarized in Table 1 for each alloy. In comparison to the 1st Gen ODS FeCrAl alloy 125YZ which had an oxygen content of 1920 ppm by weight [15], all of the new extruded 106ZY alloys had lower oxygen contents. The 106ZY alloys extruded at 950 °C after milling for 10hr (1H95C), 20hr (2H95C), and 40hr (4H95C) show an increasing C content with increasing milling time. The O and N contents vary somewhat between specimens, which will be discussed below.

2.2. Mechanical property evaluation

Mechanical property evaluation was performed using an MTS hydraulic tensile machine using a strain rate of 10^{-3} s^{-1}. Sub-sized (SS-3) tensile specimens with a 7.62 mm gauge length and a cross section of 1.52 mm × 0.762 mm were machined from each 106ZY sample listed above in Table 1. For consistency, all specimens were machined with the tensile axis parallel to the alloy extrusion direction. Tensile testing was performed at temperatures varying from room temperature to 800 °C in air, with at least one specimen being tested at each respective temperature. Due to the limited