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Ferritic oxide dispersion strengthened alloys by spark plasma sintering

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ABSTRACT

Spark plasma sintering (SPS) was used to consolidate a Fe–16Cr–3Al (wt.%) powder that was mechanically alloyed with Y_2O_3 and Ti powders to produce 0.5 Y_2O_3 and 0.5 Y_2O_3–1Ti powders. The effects of mechanical alloying and sintering conditions on the microstructure, relative density and hardness of the sintered oxide dispersion strengthened (ODS) alloys are presented. Scanning electron microscopy indicated a mixed fine-grain and coarse-grain microstructure that was attributed to recrystallization and grain growth during sintering. Analysis of the transmission electron microscopy (TEM) and atom probe tomography (APT) data identified Y–O and Y–O–Ti nanoclusters. Elemental ratios of these nanoclusters were consistent with that observed in hot-extruded ODS alloys. The influence of Ti was to refine the grains as well as the nanoclusters with there being greater number density and smaller sizes of the Y–O–Ti nanoclusters as compared to the Y–O nanoclusters. This resulted in the Ti-containing samples being harder than the Ti-free alloys. The hardness of the alloys with the Y–O–Ti nanoclusters was insensitive to sintering time while smaller hardness values were associated with longer sintering times for the alloys with the Y–O nanoclusters. Pressures greater than 80 MPa are recommended for improved densification as higher sintering temperatures and longer sintering times at 80 MPa did not improve the relative density beyond 97.5%.

1. Introduction

Several next generation fast reactor concepts are being designed for power production and used fuel transmutation applications which contain fuel forms encapsulated in high performance cladding materials capable of reaching significantly higher burn-up levels than historically possible [1]. The components used in these reactors, as well as in other advanced reactors, will require material attributes under irradiation such as dimensional stability, high strength and toughness, resistance to creep, to creep rupture, and to fatigue crack growth, in addition to excellent chemical compatibility with coolants [1]. Ferritic/martensitic (F–M) steels are candidate cladding materials for the development of advanced reactors where operational conditions include temperatures of 350–700 °C and up to 200 dpa exposure [2,3]. Historically, F–M steels have shown good swelling resistance compared to the austenitic stainless steels [4]. A preliminary choice of the cladding material is the F–M steel HT-9, however, there are concerns with the potential for radiation hardening and embrittlement at temperatures near or below 400 °C, and relatively low strength at higher temperatures [5]. Consequently, cladding materials, other than HT-9, are being sought as materials for future reactor designs.

Oxide dispersion strengthened ferritic steels are candidate cladding materials for nuclear applications due to demonstrated resistance to thermally-induced creep, and excellent strength retention [3]. Favorable properties are attributed to the dispersed oxide nanofeatures in ODS alloys. The earliest documented work in the development of ODS alloys began with Benjamin who developed mechanical alloying as a means for producing intimately mixed metal–ceramic powders for subsequent processing in 1976 [6]. In subsequent years, a variety of commercial alloys were developed including several commercial Ni- and Fe-based superalloys (MA754, MA758, MA6000 and MA956 and MA957, respectively) [7]. ODS alloys are generally processed by mechanical alloying powders via high energy ball milling and subsequent hot powder consolidation by hot extrusion or hot isostatic pressing followed by hot rolling and forming [7]. This process is relatively time consuming compared to most conventional metal fabrication processes and results in high alloy costs, as well as potentially undesirable texturing of the alloy, giving an anisotropic microstructure and undesirable defects such as stringers [8]. Post

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processing heat treatments have been assessed for removing deformation texture and to improve inferior biaxial creep-rupture strength [9,10]. In addition a number of studies have examined the effects of alloy composition on mechanical properties [11].

The use of spark plasma sintering (SPS), also known as a field assisted sintering (FAS) or pulsed electric current sintering (PECS), for powder metallurgy processing is growing due to lower sintering temperatures, shorter hold times and improvement in properties of consolidated samples when compared to conventional processing methods [12]. Although SPS is widely used, the fundamental roles of current on mass transport, pulsing, applied pressure and heating rate are only recently being reported. SPS processing been has shown to increase densification rates and permit the fabrication of materials that required an applied pressure during sintering. SPS has been used not only to fabricate near net-shape components but can also be used to fabricate cellular solids, nanograined materials, composites, coatings, and joints [7]. The development of SPS as a possible replacement for hot extrusion and hot isostatic pressing during ODS fabrication can yield a rapid and economic process that mitigates anisotropy and contamination.

There are limited reported applications of SPS in the processing of ODS alloys, leaving unique opportunities provided by this processing largely unexplored. Microstructural characterization and mechanical properties have been reported by Heintze et al. [13–15] for the Fe–9Cr ODS alloy processed by SPS. The microstructural characterization by small angle neutron scattering (SANS), APT, and TEM demonstrated that the Y–O nanoclusters were 4.2 nm in size and a ratio of Y to O between 1:3 and 2:3 [13,14]. The mechanical properties were found to be a function of composition and SPS processing parameters [15]. An effort to demonstrate the mechanisms involved in the formation of the ODS alloys through SPS was reported by Ji et al. [16] for a Fe–39.78Al–0.054Zr–0.01B (at.%) alloy used TEM and energy dispersive X-ray spectroscopy (EDS). A heterogeneous microstructure was observed where distinct regions that included small grains with nanoclusters enriched in yttrium and large grains with and without yttrium were identified. The small grain formation was attributed to a recrystallization phenomenon during the SPS process while the large grains were attributed to localized melting and insufficient milling.

Microstructural characterization of ODS ferritic alloys sintered by conventional techniques has been documented in literature [17–20] along with the influence of recrystallization [21–23]. This study involved investigating the SPS processing of a base powder Fe–16Cr–3Al mechanically alloyed with Y_2O_3 and Ti additions. It was demonstrated through TEM and APT that specific milling and SPS processing conditions yielded the characteristic nanoclusters associated with ODS alloys. A heterogeneous microstructure of fine and coarse grains was observed. The influence of the processing conditions on the microstructure, hardness and relative density are reported. The influence of Ti was determined by comparing the results of the Ti-free and the Ti-containing ODS alloys.

2. Experiments

2.1. Materials

The base starting powder was a gas atomized powder with nominal composition of Fe–16Cr–3Al (wt.%). This was supplied by Goodfellow Cooperation of Coraopolis, PA, USA. As determined via laser scattering particle size analysis, the median particle size of this powder was 13 μm with the particles being spherical and platelet shaped as shown in Fig. 1a. A precursor powder was made using yttria powder supplied by American Elements of Los Angeles, California USA. The powder had a nominal 0.1 μm diameter particle size which was introduced to the base powder at 0.5 wt.% by mixer milling 30 g batches in an Ar atmosphere for 10 h in a MSK-SFM-2 Mixer Mill supplied by MTI Corporation of Richmond, California USA. A second precursor powder was made by introducing 0.5% Y_2O_3 and 1% Ti into the base powder by mixer milling 30 g batches for 10 h. These powders would be referred to as 0.5Y_2O_3 and 0.5Y_2O_3–1Ti, respectively, with the chemical compositions listed in Table 1.

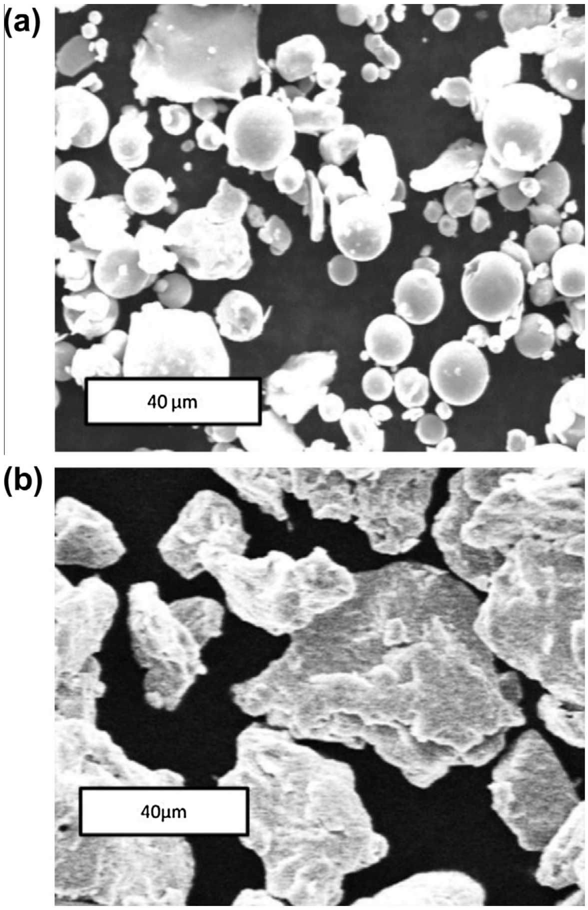

Fig. 1. Scanning electron micrographs of (a) as received powder and (b) 40 h milled powder with 0.5 wt.% yttria.

The precursor powders were mechanically alloyed in a Retsch planetary ball mill in 15 g batches in an argon atmosphere using a 250 mL stainless steel vessel and 5 mm diameter stainless steel media. The mill, media and vessel were supplied by Retsch of Newtown, Pennsylvania USA. The powders were milled for 20–40 h at 380 rpm with a ball to powder mass ratio of 10:1. These milling conditions were selected based on the powder preparation for conventional sintering of ODS alloys [24]. A scanning electron micrograph of the 40 h milled powder is shown in Fig. 1b, where the average particle size was approximately 20 μm and had a platelet-like morphology.

2.2. Spark plasma sintering

The milled powders were consolidated using a Dr. Sinter 1500 Spark Plasma Sintering System supplied by SPS Syntex Inc. of Kanagawa, Japan. A graphite die and punch assembly was used to hold approximately 3 g of powder. The cylindrical graphite die was 33 mm in external diameter with a 12 mm internal diameter cavity. Graphite foil was used to line the inner surface of the die which facilitated lubrication and acted as a release agent. A