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Original Research Paper

Mixed fuel synthesis of Y_2O_3 nanopowder and their applications as dispersoid in ODS steel

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ABSTRACT

Yttrium oxide (Y_2O_3) is one of the most stable oxides to withstand at high temperature and suitable for use as a dispersoid in oxide dispersion strengthened (ODS) steel alloy. In this paper, Y_2O_3 nano-powders are synthesized by three different techniques namely mechanical milling, single fuel combustion synthesis and mixed fuel combustion synthesis. In mixed fuel, various combinations of fuels are used instead of a single fuel (glycine, citric acid, urea, and ethylene glycol). Thermal characteristics and crystalline nature of Y_2O_3 nano powders produced by different techniques are analyzed extensively. Y_2O_3 nano powders with amorphous to nano-crystalline structure and having 8–80 nm crystallite size are obtained from the mixed fuel process, that is suitable for use as dispersiod in ODS steel. Further, these powders are dispersed in ferritic steel matrix using an innovative dual drive planetary ball mill to produce Y_2O_3 dispersed ODS steel powder within a short time period. Transmission electron microscopy analysis of milled oxide dispersion strengthened steel powder reveals a homogeneous distribution of Y_2O_3 nano powders in ferritic steel matrix after milling time of 5 h.

1. Introduction

Yttrium oxide (Y_2O_3) exhibits interesting properties like high melting point (2410 °C, high chemical and thermal stability, low volatility in vacuum, high thermal conductivity, low thermal expansion coefficient, wide optical transmission range, low neutron absorption cross section, and good resistance to erosion and thermal shock. Y_2O_3, either in its pure form or doped with other cations is being widely used in several well-known applications. Solid state lasers, lighting and cathode-ray phosphors, high-density magnetic recording are few to mention. Use of Y_2O_3 for dispersion strengthening of steels, and as sintering additives for Al_2O_3 and Si_3N_4 ceramics is also very popular [1].

For the purpose of dispersion strengthening of steels, Y_2O_3 is the most preferred material because it is thermodynamically more stable than nitrides, carbides and even other oxide materials. For example, CeO_2 undergoes reduction ($Ce^{4+} \rightarrow Ce^{3+}$) at higher temperatures and the oxygen released in this process tends to retard the densification behavior. Y_2O_3 has high temperature phase stability as well as low neutron absorption than any of the rare earth oxide [2]. Nano Y_2O_3 can be synthesized by bottom-up as combustion synthesis (CS) or top-down approach as mechanical milling (MM). CS is a simple process with less time consuming, cost-effective, and reproducible with high yield. This method has been used for the preparation of a wide variety of materials, including advanced ceramics, composites, intermetallics, semiconductors, insulators, sensors, phosphors and many more [3]. MM is also a well-established and potential method for producing bulk amounts of nano-sized powders. Commercial micron sized powders are generally used as the raw materials in this process. Numerous studies have been conducted on the CS of nano-powders using individual fuels. But employing the mixture of fuels in CS is a relatively a different approach. A mixture of fuels is generally used to control the flame temperature and thus to have a control over the stability of various phases that are formed. The control over flame-temperature is achieved by mixing a fuel of low-flame-temperature to a fuel of high-flame-temperature [4] and is universally known as mixed fuel combustion synthesis (MFCS). The potentialities of MFCS in tuning the powder properties by controlling the energy of combustion reaction have successfully been exploited in some recent studies. Some of the remarkable outcomes of MFCS are well controlled enthalpy and exothermicity of the combustion reaction [5], improved particulate properties,

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sinterability of the final product [6], in-situ formation of plasma sprayable grade CeO_2, single-phased $CeAlO_3$, $Ca_3Al_2O_6$, and $Y_3Al_5O_{12}$ powders [7–10]. Nanocrystalline Y_2O_3 has been synthesized by combustion technique using glycine [11], citric acid [$C_6H_8O_7$] [12], urea [CH_4N_2O] [13], ethylene glycol [$C_2H_6O_2$] [14], and several hydrazine-based [15] individual fuels. It appears that no report on the preparation of nano-sized Y_2O_3 by the MFCS. Thus, considering its capability to tailor the product properties, there is a compelling interest to explore this new, modified approach for synthesizing nano-crystalline Y_2O_3 with controlled powder characteristics.The present investigation is directed to make a systematic study of MM, CS and MFCS for the preparation of nano-crystalline Y_2O_3. Further, the nano Y_2O_3 produced with one of the optimized process was used as a dispersoid for the preparation of Oxide dispersion strengthened (ODS) ferritic steel. The purpose of adding nano Y_2O_3 in steels is to improve its high temperature stability; especially in case nuclear reactors where the low neutron absorption cross-section of Y_2O_3 assumed to play its role [16].

2. Experimental details

2.1. Synthesis of Y_2O_3 by CS and MFCS

Nanocrystalline Y_2O_3 is synthesized by single fuel combustion of aqueous solutions containing stoichiometric amounts of the corresponding metal nitrate and glycine. These raw materials are of high purity, analytical reagent grade. For the synthesis of different batches calculated amount of yttrium nitrate [$Y(NO_3)_3{\cdot}6H_2O$, 99.9%, Alfa Aesar, USA] and glycine [$C_2H_5NO_2$, 99.9%, Himedia, France] are taken corresponding to a G/N ratio of 0.553 and then dissolved in a minimum volume of water to obtain transparent aqueous solutions. G/N refers to glycine-to-nitrate ratio ($G/N = M_{glycine}/M_{nitrate}$, M = molar amount of the compound). The G/N ratio is varied from 1:1 to 1:2 in this work. The resulting translucent solutions are taken in a glass beaker (1000 ml), heated on a hot plate at 110 °C with constant stirring, using a magnetic stirrer to remove the excess solvent. These homogeneously mixed solutions, after thermal dehydration, result in the viscous liquids. Then the viscous gels are heated and evaporated continuously at around 250–300 °C, for 30 min to 1 h for producing 5 g of powder in each batch. During heating, the viscous liquid swelled and auto-ignited with the rapid evolution of a large volume of gases (CO_2, N_2, H_2O) to produce voluminous yellowish white ash powders. The powders obtained after auto-ignition are calcinated at 600 °C for 2 h in a muffle furnace.

Glycine, citric acid [$C_6H_8O_7$, 99.9, Himedia, France], and ethylene glycol [$C_2H_6O_2$, 99.9%, Himedia, France] are used as fuels for MFCS, taking two at a time, for the synthesis of nanocrystalline Y_2O_3. The procedure followed for the synthesis of Y_2O_3 is same as discussed for single fuel system.

2.2. Mechanical milling of Y_2O_3 powders

The mechanical milling is carried out on commercial micron sized (3–5 μm) Y_2O_3 (99% pure, HiMedia, India) using a high energy planetary ball mill (InSmart systems Inc., India). The milling media (balls and vials) are made up of stainless steel and the ball diameter used for milling is 10 mm. The mill speed is 300 rpm and the ball to powder ratio is 10:1 by weight. Toluene (Merck, Germany) is used as process control agent and it promotes fracture as well as provides a protective atmosphere. The powder blends are milled up to 10 h and samples collected at an interval of 1, 3, 5, and 10 h of milling, washed with distilled water and then with ethyl alcohol followed by drying then taken for characterization.

2.3. Mechanical alloying of nano Y_2O_3 dispersed ferritic steel powder

An efficient dispersion of nano-oxides in ODS ferritic steels is achieved by employing high energy ball milling technique using dual drive planetary ball (DDPB) mill. The starting material used in this study is pre-alloyed ferritic steel powder supplied by M/s Hoganas, Belgium. The chemical composition along with relevant powder properties are given in Table 1. The Y_2O_3 dispersoid is prepared by CS route in the form of particles between ~30–50 nm in size. The milling experiments have been carried out by using DDPB mill and reported in detail elsewhere [17,18].

2.4. Characterization

The thermal decomposition characteristics of the precursor gels are investigated by means of simultaneous thermo-gravimetric analysis (TGA) and differential scanning calorimetry (DSC) in the temperature range of 30–800 °C using a thermal analyzer (STA, Netzsch, Germany). The diffraction patterns of combustion synthesized as well as ball milled powder samples are collected using Ni-filtered Co Kα radiation with the help of an X-ray diffractometer (XRD), (PANalytical Inc., Netherlands). The phases formed are identified by comparison of the recorded diffraction peaks with the ICDD database using X'Pert HighScore Plus software (PANalytical Inc., Netherlands). In order to determine the crystallite size of the powders, the $K_{\alpha 2}$ contribution of the XRD spectra is stripped by the Rachinger correction method. Profile fitting is carried out using the Pseudo–Voigt profile function included in the above software. In order to correct the instrumental line broadening of the measured profiles, a silicon disc is used as an external standard sample. The experimental diffraction profile is corrected for instrumental broadening by means of the following relation assuming Lorentzian shape of the peaks to calculate crystallite size:

$$B = B_M - B_S \tag{2.1}$$

where B is the true width that is, peak broadening from the intrinsic profile (pure diffraction profile), B_M is the measured width of the diffraction peak from the experimental profile and B_s is the instrumental width of the diffraction peak from the standard sample. Crystallite size is calculated from the XRD line broadening using the standard Scherrer's equation assuming spherical particles:

$$D = \frac{K\lambda}{B\cos\theta} \tag{2.2}$$

where D is the crystallite size in nm, λ is the radiation wavelength (1.7902 nm in the present case, Co target), θ is the diffraction peak angle in radians. However, to calculate lattice strain, according to the formula, broadening is calculated assuming Gaussian profile with formula:

$$B_{LS} = \sqrt{\left(B_M^2 - B_S^2\right)} \tag{2.3}$$

where B_{LS} is broadening due to lattice strain and with this, *X'Pert High Score Plus* software uses the tangent formula to calculate lattice strain given as:

Table 1
Chemical composition and powder properties.

Chemical analysis		Physical properties	
Element	%	Apparent density	2.82 g/cm^3
Carbon	0.024	Flow rate	29.8 s/50 g
Molybdenum	1.00	*Sieve analysis*	
Manganese	0.1	+150 μm	1.29%
Chromium	16.2		
Silicon	1.0	−45 μm	35.37%
Iron	Balance		