



# Studies on the synthesis of nanocrystalline yttria powder by oxalate deagglomeration and its sintering behaviour

S. Balakrishnan<sup>a</sup>, K. Ananthasivan<sup>a,\*</sup>, K.C. Hari Kumar<sup>b</sup>

<sup>a</sup>Chemistry Group, Indira Gandhi Centre for Atomic Research, Kalpakkam, India

<sup>b</sup>Department of Metallurgical and Materials Engineering, Indian Institute of Technology Madras, Chennai, India

Received 30 August 2014; received in revised form 31 October 2014; accepted 17 November 2014

Available online 24 November 2014

## Abstract

Nanocrystalline yttria powders were synthesized from deagglomerated yttrium oxalate. The precipitation of this oxalate was carried in two different modes *viz.*, addition of aqueous oxalic acid into yttrium nitrate solution (forward strike) and vice versa (reverse strike) followed by ultrasonication in acetone and water. Nanocrystalline yttria was obtained by calcining the oxalate in air at 1073 K. The bulk densities, specific surface area, X-ray crystallite size, size distribution of particles as well as the quantity of carbon residue in these powders were determined. The influence of the deagglomeration medium on the powder properties was analyzed. Scanning electron microscopy (SEM) showed that these powders comprised irregular agglomerates while the high resolution transmission electron microscopy (HRTEM) revealed that the constituent units of these agglomerates were randomly oriented cuboidal nanocrystallites (20–40 nm). These powders were compacted at 120 MPa without any lubricant or binder and their sinterability was studied. Pellets with sintered density as high as 97.5% T.D. (theoretical density) could be obtained at a relatively low sintering temperature of 1873 K. Synthesis of nanocrystalline yttria powders by oxalate deagglomeration route as well as the systematic studies of their properties and sinterabilities are being reported for the first time. It was further demonstrated in this study that higher sintered densities could be obtained with less number of process steps and at a much lower compaction pressure. Samples prepared by reverse strike yielded a powder with characteristics most suitable for fabricating high density yttria bodies. 1673 K would be the optimum temperature for sintering the compacts made out of this powder.

© 2014 Elsevier Ltd and Techna Group S.r.l. All rights reserved.

**Keywords:** Yttria; Oxalate; Sintering; Precipitation; Deagglomeration

## 1. Introduction

Yttrium oxide finds extensive use in a variety of applications. It is used in the manufacture of optical components, and as a transparent material for infrared applications [1–3]. It is used as a container for melting reactive molten alloys, salts [4–8] and as a crucible for holding molten uranium alloys owing to its inertness and higher thermodynamic stability [9–14]. Yttria doped ceramics are also being considered as inert matrices for burning minor actinides in nuclear reactors [15].

Synthesis of yttria by precipitation, combustion, polymerization, solvothermal technique, explosive decomposition,

non-aqueous method (xerogel based) and electrospray pyrolysis has been investigated extensively by different authors [1,14,16–46]. Among these, the precipitation route is widely used [1,16–19,23–26,30,46,47]. Various authors studied the sinterability of the yttria powders either in the context of fabricating transparent yttria ceramic bodies or obtaining consolidates with a high sintered density. The precipitation synthesis that employs oxalates, carbonates and hydroxides as precursors are the most frequently used [16]. The morphology and characteristics of the precursors as well that of the final powder could be changed by altering the mode and conditions of precipitation [16,30] *viz.*, addition of precipitant into yttrium nitrate solution (forward strike) or vice versa (reverse strike). Removal of water from these precursors is also an important aspect of the synthesis that would have a strong influence on

\*Corresponding author. Tel.: +91 44 2748 0500x24069.

E-mail address: [asivan@igcar.gov.in](mailto:asivan@igcar.gov.in) (K. Ananthasivan).

the characteristics of the final product including the degree of agglomeration and sinterability of the latter [16–18,46]. The degree of agglomeration could be reduced by washing the precursors with different solvents or by using an appropriate drying procedure [1,16–18,25,46]. Mouzon et al. [46], Ananthasivan et al. [48,49], Yasrebi et al. [50] and Oktay et al. [51] used ultrasonication for bringing about the deagglomeration. All these process steps strongly influence the powder properties and its sinterability significantly.

Tiny yttria crucibles (typically of about 9 mm outer diameter, 8 mm inner diameter and 7 mm height) are required for some experimental investigations that are being currently pursued in our laboratory [52,53]. A larger number of these are to be used in experiments that necessitate the containment of molten uranium bearing alloys. We have devised a method to fabricate crucibles in-house by die casting nanocrystalline yttria powders and sintering at relatively low temperatures [44]. Although powders derived from citrate gel combustion [44] yielded high density sintered bodies, they were found to contain isolated pores distributed throughout the matrix. This led to the percolation of the molten alloy and rendered them less desirable for the current application. Hence, an attempt has been made in this work to devise an alternate method to produce powders in bulk quantities that yielded sintered samples of less porosity. Ananthasivan et al. [48,49] reported that oxalate deagglomeration yields highly sinterable nanocrystalline powders and fabricated thoria crucibles from them [49]. So far no systematic studies have been reported on the oxalate precipitation synthesis of yttria, its characterization and subsequent sintering. Hence, in this study nanocrystalline yttria powders were synthesized by using the oxalate precipitation and ultrasonic deagglomeration method. The dependence of the characteristics of the powder on the deagglomeration medium and their sinterabilities were studied.

## 2. Experimental

### 2.1. Chemicals

Yttrium nitrate hexahydrate with a purity better than 99.9% supplied by MFTC Group, China and oxalic acid (> 99.5%) supplied by Merck were used in the synthesis of yttria. Acetone (AR grade) was supplied by Merck.

### 2.2. Experimental procedure

In a typical experiment, appropriate amounts of yttrium nitrate and oxalic acid were weighed and dissolved in 100 and 300 mL of double distilled water, respectively. Yttrium oxalate was precipitated by using both the modes of precipitation *viz.*, addition of oxalic acid to yttrium nitrate solution (forward strike, F) and vice versa (reverse strike, R) at the room temperature (300 K). The precipitate was filtered by using a funnel fitted with G3 sintered glass frit for about 30 min under suction. The completion of precipitation was ascertained by ensuring that the filtrate did not yield any precipitate when treated with the oxalic acid solution again. Subsequently, the precipitate thus obtained was dispersed in a medium (water or acetone) and was ultrasonically

deagglomerated for about 15 min and then filtered. This process of deagglomeration and subsequent filtration is termed as a cycle and each experiment comprised three such cycles. The total duration of the precipitation and deagglomeration was about 3 h. These oxalate precipitates were dried under an infrared (IR) lamp for about 1 h. The procedure followed in all these syntheses is depicted in the flowchart (Fig. 1). The decomposition temperature of the oxalate was identified by thermogravimetry coupled with differential thermal analysis (TG-DTA) at a heating rate of 10 K min<sup>-1</sup> and the oxalates were calcined at 1073 K for 4 h in air.

The following nomenclature has been used to identify the samples prepared in this study. The alphabets “F” and “R” denote the mode of precipitation *viz.*, forward and the reverse strike, respectively. “A” and “W” were suffixed to indicate the medium used for deagglomeration *viz.*, acetone and water, respectively. The powders that were not subjected to the deagglomeration treatment were identified with the suffix “N”. For example, the label “RN” indicates a powder that was prepared by using the reverse strike and without deagglomeration.

The calcined powders were then compacted at 120 MPa in a die lined with tungsten carbide into pellets having a diameter of about 10 mm and a thickness of about 2–3 mm by using punches made out of tungsten carbide. An automated double action hydraulic press supplied by Bemco Ltd. India, was used for carrying out the compaction. They were subsequently sintered in air at 1473, 1673 and 1873 K for 4 h with a heating and cooling rate of 5 K min<sup>-1</sup>. A furnace equipped with molybdenum di-silicide heating elements was used for sintering.

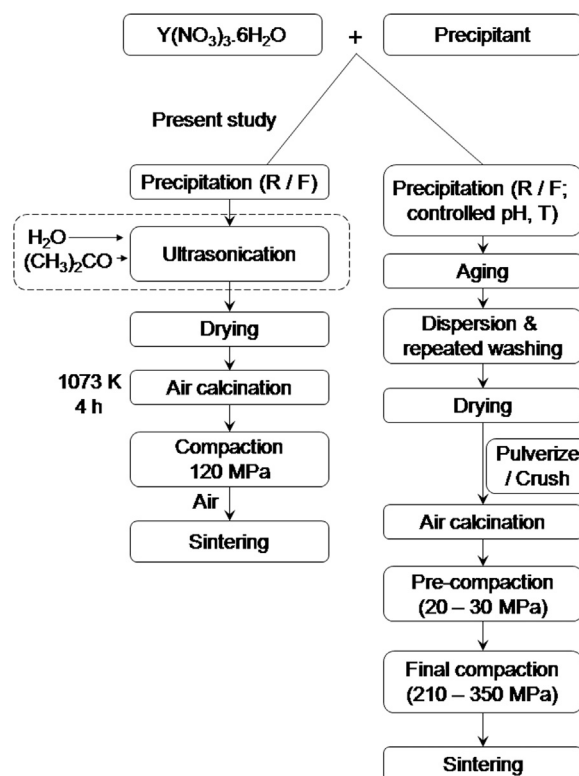


Fig. 1. Flow chart of the present work vis-à-vis general flow chart followed by other authors.

Download English Version:

<https://daneshyari.com/en/article/1460095>

Download Persian Version:

<https://daneshyari.com/article/1460095>

[Daneshyari.com](https://daneshyari.com)