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Short communication

Flame aerosol synthesis of phase-pure monoclinic Y_2O_3 particles via particle size control

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ABSTRACT

In this study, for the first time, a particle size effect on crystal structure of Y_2O_3 particles was exploited to synthesize phase-pure monoclinic Y_2O_3 particles. In the synthesis process, a precursor aerosol consisting of H_2 fuel gas and precursor droplets passed through an impactor before it entered a flame to form yttria particles. A round-jet impactor was used to remove the large precursor droplets, so that the product Y_2O_3 particles were all smaller than a critical size of approximately 1.5 μm. Due to the particle size effect on crystal structure, the Y_2O_3 particles thus obtained were essentially phase-pure with the monoclinic structure. The result shows that, by using an impactor to alter the particle size distribution, it is possible to control the crystal structure of Y_2O_3 particles while maintaining relatively high synthesis yield.

1. Introduction

Y_2O_3 is a material that finds applications in phosphors, catalysts and optical window materials. Y_2O_3 has multiple crystal structures with differing properties. Mainly two phases of Y_2O_3 are possible from regular synthesis processes, namely the C-type cubic phase and the B-type monoclinic phase [1]. The two crystal structures have significantly different thermophysical and optical properties. For example, the density of monoclinic Y_2O_3 is significantly higher than that of the cubic phase [2]; the fluorescence properties of Eu-doped Y_2O_3, an important phosphor material, are strongly dependent upon the Y_2O_3 crystal structure [3]. So far the application for monoclinic Y_2O_3 particles has not been extensively explored. One apparent reason is simply that, until now there has not been a feasible synthesis method to produce this material in large quantities.

Nevertheless, the monoclinic structure is an important phase for Y_2O_3 and other rare earth sesquioxides [6]. A number of researchers have studied the synthesis of monoclinic Y_2O_3 using various methods. A summary of the reported methods for synthesizing monoclinic Y_2O_3 particles is given in Table 1. High-pressure processes require special equipment and are not capable of producing nanoparticles or microparticles [1]. Furnace-based heating–condensation methods require two steps, can only produce monoclinic Y_2O_3 particles <10 nm, and are not suited for continuous synthesis of significant quantities of material [2]. In a previous study, a flame process with a gas-phase precursor was used to synthesize monoclinic Y_2O_3 particles, but the particle size was limited to below 90 nm due to the gas-phase precursor. The batch mode precursor loading also makes it difficult to achieve continuous synthesis with that method. In addition, the precursor used in that method is costly [4]. More recently, a flame spray pyrolysis (FSP) method was used to synthesize monoclinic Y_2O_3 particles with diameters up into the micrometer range. However, the polydisperse Y_2O_3 particles had mixed cubic and monoclinic phases. A critical particle diameter of approximately 1.5 μm was found. At the critical diameter, the probability was 50% for a particle to be either cubic or monoclinic. Particles significantly smaller than the critical diameter were all monoclinic, while those significantly larger were all cubic. [5].

To explore the potential applications for monoclinic Y_2O_3, one must first be able to synthesize phase-pure Y_2O_3 in sufficient quantities. This synthesis capability is also key to studying the interplay between surface energy and polymorphism [7]. The relationship between surface energy and polymorphism is a topic of profound importance in materials formation, especially on the nanometer scale [8]. This work was motivated by the above-mentioned reasons. The basic operating principle used in this study was that, if all the Y_2O_3 particles produced from the flame synthesis process were smaller than the critical size, then they would all have the monoclinic structure. Therefore phase-pure monoclinic Y_2O_3 particles may be generated via controlling the particle size. Herein we report the experimental methods for particle size control and the respective results. In particular, we describe the design and the successful use of a real impactor for achieving particle size control. To the

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Table 1
Summary of methods for synthesizing monoclinic Y_2O_3 particles

Author	Method	Maximum process temperature (°C)	Critical particle size[a]	Yield
Hoekstra [1]	High pressure diamond anvil	~1000 °C	∞ (Bulk sample)	Unspecified
Krauss et al. [2]	Evaporation–condensation	350 °C	~10 nm	~150 mg/batch (inferred from sample description)
Guo et al. [4]	Flame aerosol process with gas-phase precursor	~2700 °C	Not observed (All particles monoclinic largest ~90 nm)	40–120 mg/h (batch mode; each run approximately 15 min)
Guo and Luo [5]	Flame aerosol process with droplet precursor (flame spray pyrolysis)	~2700 °C	~1.5 μm (Mixed with larger Y_2O_3 particles that were cubic)	200 mg /h (0.65 M precursor solution) and 10 mg/h (0.026M precursor solution)

[a] Critical size is the size below which the Y_2O_3 particles had the monoclinic structure, and those larger had the cubic structure.

best of our knowledge, this is the first report of a real impactor being used in flame spray pyrolysis to obtain phase-pure product particles.

2. Experimental methods

2.1. Flame spray pyrolysis apparatus

The flame spray pyrolysis apparatus is schematically shown in Fig. 1. It is similar to the apparatus used in a previous work [5], except that an optional impactor was incorporated in the apparatus in this study. The inner/outer diameters of the burner nozzle are 1.6 mm and 9.5 mm, respectively. The apparatus consisted of a 1.7 MHz atomizer, an atomization vessel, a furnace and a burner. The furnace and the burner have been described in detail elsewhere [4]. When the optional impactor was used, the precursor aerosol had to flow through the impactor first before entering the furnace and burner. The impactor removed the large droplets from the precursor aerosol.

The precursor aerosol formed a self-sustained steady-state aerosol flame at the top of the burner. A co-flowing oxidant stream supported the flame. The furnace heated precursor aerosol to maintain a sufficiently high flame temperature. The precursor aerosol underwent chemical reactions in the flame and became a Y_2O_3 aerosol. The post-flame aerosol containing Y_2O_3 particles was drawn into a sampling tube by vacuum and the particles were collected on an alumina membrane filter (Whatman Inc., NJ). The synthesis apparatus was operated at atmospheric pressure. H_2 was used as the fuel gas at a flow rate of 1 SLM (standard liter per minute). Pure O_2 at 6 SLM was used as the oxidant stream to support the flame. The flame length was approximately 5 cm. The precursor solution was prepared by dissolving yttrium nitrate hexahydrate (chemical formula $Y(NO_3)_3{\cdot}6H_2O$, 99.9%, Alfa Aesar, Ward Hill, MA) in Nanopure® water (Barnstead, Dubuque, IO). The concentrations of the precursor solution used in this study were from 0.026 to 0.65 M.

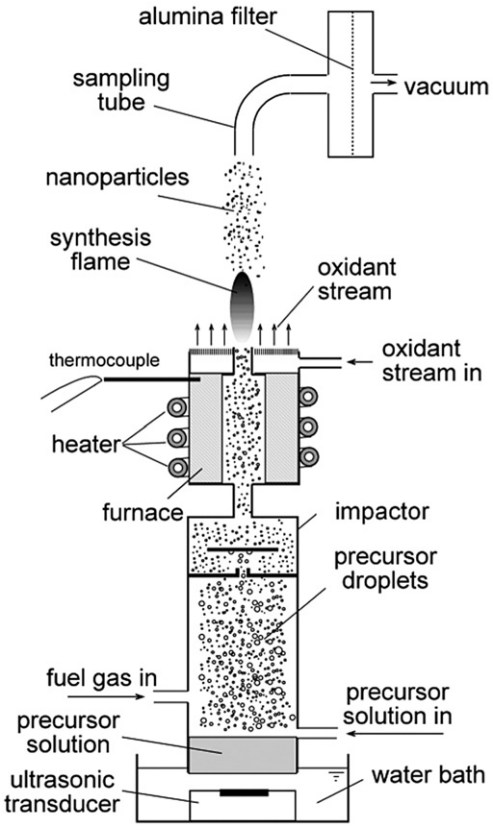

Fig. 1. Schematic of flame spray pyrolysis apparatus for Y_2O_3 synthesis.

2.2. Impactor design

The objective of the impactor design was to ensure removal efficiency greater than 80% for 6-μm droplets. With a 0.65-M precursor solution, a 6-μm droplet would produce a 1.5-μm Y_2O_3 particle, assuming that one droplet becomes one final Y_2O_3 particle. Based on mass conservation one can readily infer the relation between a precursor droplet diameter and the resultant Y_2O_3 particle diameter, knowing the precursor concentration. This relation neglects the evaporation of Y_2O_3 particles and coagulation between Y_2O_3 particles. These assumptions will be discussed later. The primary dimensions of the impactor were determined using the relations given by Marple and Willeke [9]. The design gas was H_2 at 1 SLM. The droplet density was 1120 kg/m^3 (measured density for the 0.65 M precursor solution). The impactor had a round nozzle with an inner diameter of 2.87 mm. The nozzle inner diameter was selected by trial and error, so that the precursor droplets had the proper Stokes number, and hence desired removal efficiency in the impactor. The relation between removal efficiency and Stokes number (and the Reynolds number to a lesser degree) was found in the paper by Marple and Willeke [9]. The nozzle-impaction plate distance was 3.30 mm, selected based on the empirical relation for circular jet impactors [10]. Estimated removal (collection) efficiency and the corresponding Y_2O_3 particle diameter for several droplet sizes are given in Table 2. A schematic drawing of the impactor is shown in Fig. 2.

2.3. Computational fluid dynamics simulation for the impactor

Computational fluid dynamic (CFD) simulation was used to verify the droplet removal efficiency for the impactor. The flow field through the impactor was simulated using FLUENT 6.2 (Fluent Inc., Lebanon,

Table 2
Estimated removal efficiency and the corresponding Y_2O_3 particle diameter for several droplet sizes

Droplet diameter, 0.65-M solution (μm)	4	6	6.8
Stokes number	0.20	0.45	0.58
Estimated efficiency of removal by impactor [9]	<20%	>80%	>90%
Resultant Y_2O_3 particle diameter (μm)	1.0	1.5	1.7