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A Critical Role of Fuel Molar Ratio in the Microwave Combustion Synthesis of Nano Size α-Al_2O_3

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Abstract
Nano size highly crystalline α-Al_2O_3 powders have been synthesized by the microwave combustion approach. A study of the evolution of crystalline phase of obtained powders and the morphology of the particles were accomplished through X-ray diffraction and High resolution transmission electronic microscopy. The effect of metal to fuel molar ratios on the crystal structure, particle size, and morphology of the powders was examined. Microwave combustion gave finer particles with very small agglomerate size as revealed by TEM analysis. Also the results obtained allow the identification of the highly crystalline single-phase αAl_2O_3 powder was obtained after microwave combustion at 900 w for 5 min with metal/fuel molar ratio as 1:3.
Keywords: Microwave Combustion, Mixed Oxide, Synthesis, Characterization.

1. Introduction

α-Al_2O_3 is a ceramic material of industrial importance, due to its promising structural, chemical and morphological properties [Lo´pez et. al., 2006]. Synthesis of α-Al_2O_3 has been prepared by using different methods such as hydrothermal methods [Gao et. al., 2008], homogeneous precipitation [Xinghua et. al., 2012] and [Sugita et. al., 2001], sol–gel method [Shokry et. al., 2014], and [Mirjalili et al., 2010], plasma spray synthesis [Tok et. al., 2006], organic precursor route [Zaki et al., 2012] and [Hassan et al., 2012], reverse micro-emulsion route [Jingjing and Bolin 2013] and combustion synthesis [Chen et al., 2005] and [Kiranmala et al., 2012] a part from the conventional solid state reaction [Amartya et al., 2010]. Each method has its own advantages and disadvantages. Traditional method of fabricating α-Al_2O_3 includes direct sintering of the transitional alumina phases. This method requires very high temperature, which inevitably results in a considerable degree of particle coarsening with small surface area [Pati et al., 2000].

Combustion synthesis is an effective, low-cost method for the synthesis of a wide variety of industrially useful materials [Rekha et al., 2009] and [Alexander et al., 2007]. Combustion synthesis does not require special igniting equipment and the operation is simple and easy [Guanghua and Kexin, 2009]. It involves a self-sustained reaction containing a metal nitrate and an organic fuel. In a combustion synthesis, the initial reaction media is in liquid form, which allows molecular level mixing of the reactants that result in a homogeneous product. The evolution of large amount of gases such as CO_2, NOx, and H_2O results in fine porous powder, which is amenable for further processing.

Here reporting a rapid and novel single step synthesis of α-Al_2O_3 by microwave combustion method and effect of metal/fuel molar ratio was studied. Since urea has been proven to be the best fuel for combustion of aluminum nitrate [Zhai et al., 2006] so aluminum nitrate and urea were used as the starting materials.

2. Experimental

$Al(NO_3)_3 \cdot 9H_2O$ (99%, Aldrich) and urea (99, Merck) were weighed to obtain their molar ratio of 1:3, 1:2 and 1:1. The mixture was dissolved in D/W to make a clear solution. The sol was treated in a domestic microwave working at 850 W, 2.45 GHz for 3–5 min. The combusted powders were characterized by X-ray diffraction (XRD) on a Shimadzu XD-1 diffractometer using Cu-target & Ni-filtered radiation. High resolution transmission electron microscope (HRTEM) is a JEOL 2100F TEM at an accelerating voltage of 200 kV with Electron Diffraction (SAED).

3. Results and Discussion

3.1 X-ray diffraction analysis

The structural analysis of the powders derived is investigated by XRD and shown in Figure 1. As indicated by Fig. 1, at metal/fuel molar ratios are 1:0.5, 1:1and 1:2, the patterns are ascribed to an amorphous phase. It has been observed that the XRD pattern of the powder is highly crystalline JCPDS 10-0173, it confirmed that complete formation of single phase α-Al_2O_3 took place in the presence of a high concentration of urea (i.e. 3 molar ratio with respect to Al) because the material is derived from the higher organic precursors as fuel fulfil the equivalence theory where ratio of oxidizing valency to reducing valency is equal to unity [Jain et al., 1981]

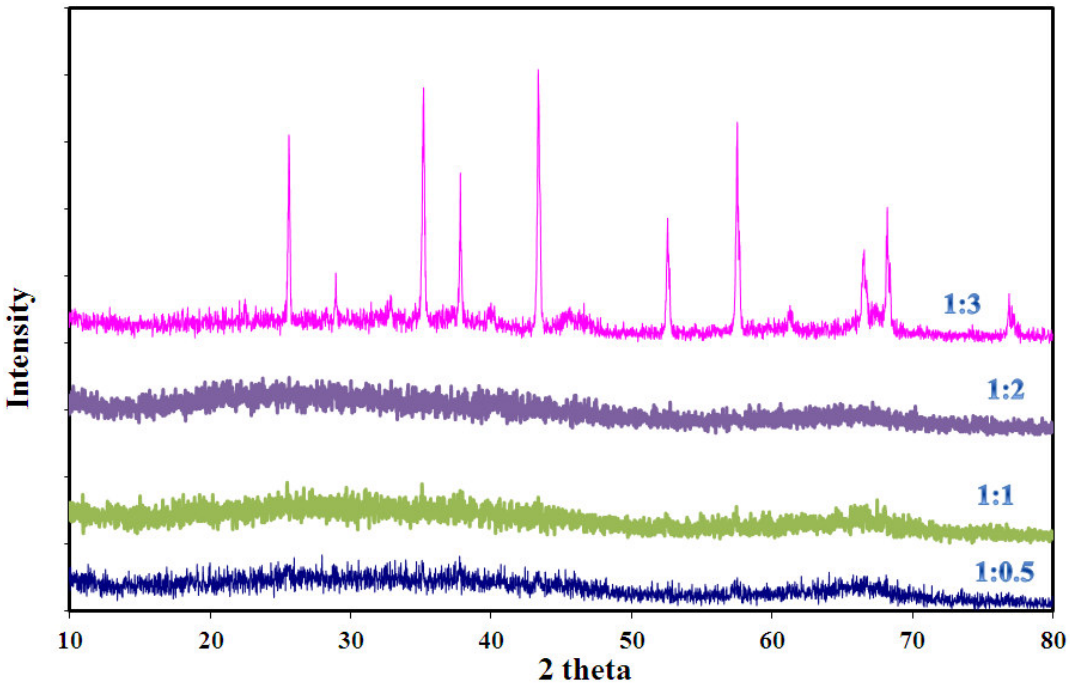

Figure 1. X-ray diffraction patterns for alumina samples with different metal/fuel ratios

3.2 High Resolution Transmission Electron Microscopy (HRTEM)

The images of transmission electronic microscopy (TEM) displayed in Fig 2. TEM (Fig. 2a) of the microwave combusted powder showed very fine particles in the range of 1.4–2 nm with close to spherical and uniform morphology on the metal: fuel molar ratio equal 1:3. Further the particles form very small agglomerates of ~100 nm whereas particles at metal/fuel molar ratio is 1:1 obtained in the range of 50–90 nm with polyhedral morphology and quite big agglomerates (Fig. 2b). HRTEM image shows dependence of fuel concentration on the particle size of the samples decreases with the increase of the fuel ratio. This is due to the crystallization of α-Al_2O_3 phase, whose particles are typically ultrafine.

Electron Diffraction (SAED) patterns in Fig. 2 has shown the crystallization processes dependence on fuel ratio. The crystallization is yet completed at low urea concentration (Fig. 2d), the formation of poly-crystalline (Fig. 2c) after a high concentration of urea (i.e. 3 molar ratio with respect to Al) has been observed as confirmed with above XRD data.

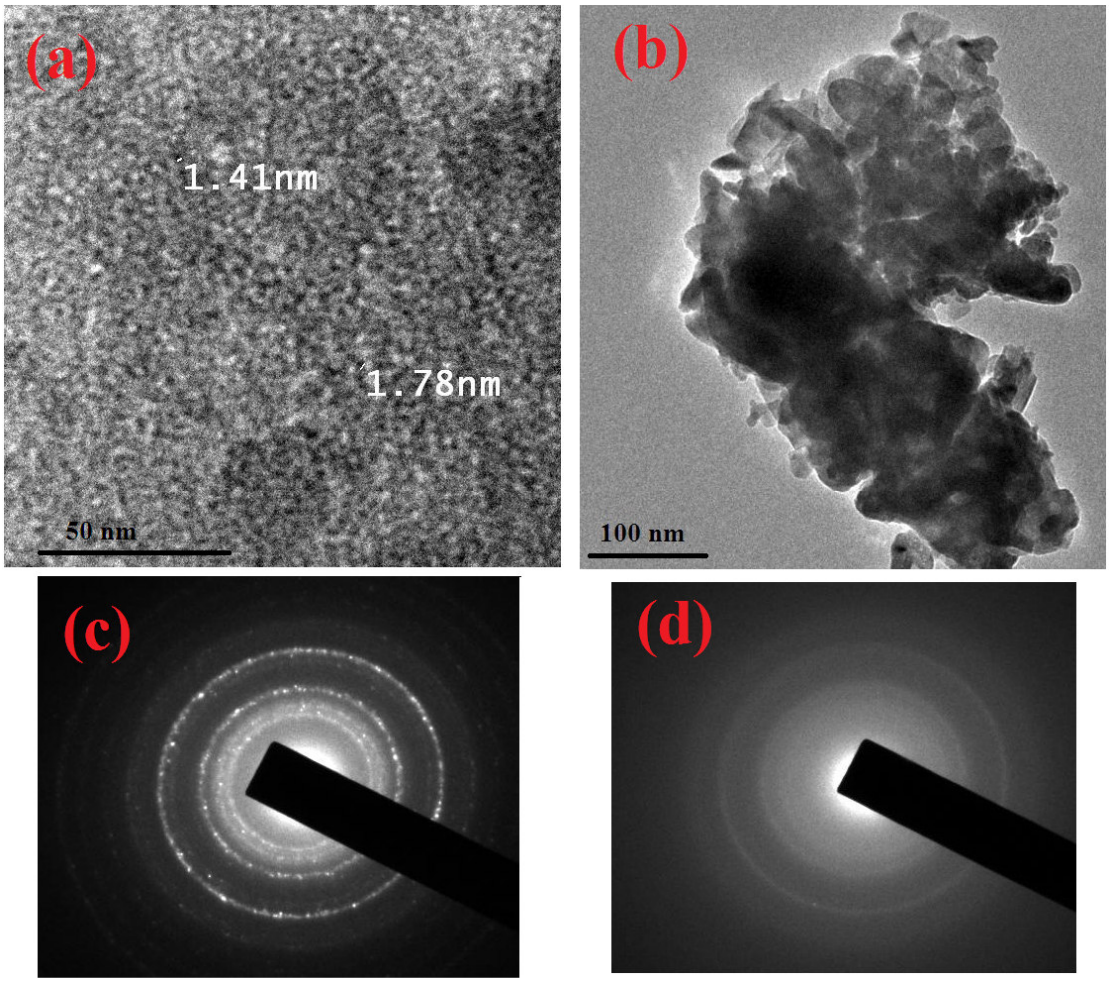

Figure 2. TEM images for alumina samples with different Mo/W ratios.

4. Conclusion

There is a critical role of fuel concentration in the microwave combustion synthesis nano size α-Al_2O_3. Microwave gel combustion resulted in formation of single phase α- Al_2O_3 nanopowders in a single step. Fine particles with size range 1.5-2 nm and close to spherical morphology were obtained by microwave gel combustion with high urea ratio compared to large particles of 50–90 nm size range and polyhedral morphology obtained from low urea ratio for direct synthesis of α- Al_2O_3. Thus microwave gel combustion method in addition to its energy and time efficiency, is easy for handling and scalable for synthesis of single phase α- Al_2O_3 nanopowders.

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