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Synthesis of Al_2O_3-ZrO_2 Nanocomposite by Mechanical Activated Self-propagating High Temperature Synthesis and Ignited via Laser

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ABSTRACT

Al_2O_3-ZrO_2composite has unique properties such as high toughness, high wear resistant and relative low thermal expansion. In this study, nanocomposite of Al_2O_3-ZrO_2 was produced by Mechanical activated Self propagating High-temperature Synthesis (MASHS) using laser beam for ignition. Al and ZrO_2 powders were mixed in the mole ratio of 1:1 and milled for 1, 3 and 6 hours. The mixtures were pressed and were exposed by continuous wave (CW) CO_2 laser for combustion reaction to start. In order to characterize the products, X-ray diffraction (XRD), scanning electron microscopy (SEM) and Energy-Dispersive X-ray spectroscopy (EDX) were implemented. The XRD results show that samples have α-Alumina and also cubic and monoclinic phases Zirconium oxides together. The dendrite structures were formed during the process.

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1. INTRODUCTION

Recent decades, increasing attention has been given to the production of nanometer powders and nanophase materials; because nanophase materials have novel electronic, optical, and magnetic, transport and mechanical properties. Novel mechanical properties include sinter ability at low temperature, high plasticity, high strength and easily machining of sintered products [1].

Engineering ceramics is known worldwide because of its high operating temperature, corrosion and wear resistance [2]. The Al_2O_3-ZrO_2 system was chosen as a model system for its many applications such as oxygen sensor, high temperature fuel cells, thermal barrier coatings, catalyst support and toughened ceramic [1].There are many methods for preparation of nanomaterials, e.g., high-energy milling [1], mechanochemical– hydrothermal and sol-gels [3] methods. The major problem associated with these techniques is process duration [4]. It is well recognized that a combustion–based technique as Self propagating High-temperature Synthesis (SHS), is an effective energy and time saving method for synthesis of a variety of advanced materials [4].The SHS process has two basic steps: ignition of reaction, and propagation of reaction. The ignition is first initiated by an external heating source such as laser beam irradiation, radiant flux, resistance heating, spark or chemical oven, and then induced by the chemical reaction inside the heated materials [5]. Recently, SHS has been combined with mechanical alloying leading to better and faster combustion reactions [6]. In this study, laser beam was used as external heating source for ignition, and in order to approach nanoscale materials, a short duration milling was used

2. MATERIALS ANDMETHODS

Micron size Al powder (Fluka,No.06140, purity>99%) and micron size ZrO_2 (purity>98.5%) were mixed in the mole ratio of 1:1 and milled for 1, 3 and 6 hours at 300 rpm and ball to powder ratio (BPR) 5:1 in a planetary mill (Retsch, model: PM100) in Ar atmosphere. Then, the mixtures were pressed into a 10 mm diameter disks. A continuous wave (CW) CO_2 laser with following

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parameters: 10.6μm, maximum output power - 150W, beam diameter 1mm, focal lens length 5cm, and was carried out to combustion reactions under oxygen atmosphere. After cooling, the density of 6 hours milling sample was measured by Archimedes method according to ASTM C373-88 standard. The microstructure of the synthesized samples were observed and analyzed by scanning electron microscopy (SEM) (VEGA\\TESCAN) equipped with an energy-dispersive spectroscope (EDX). The X-ray diffraction (XRD) (Philips pw3710) with Cu K_α radiation at 30kV and 25mA was used to identify the phase constitution synthesized samples.

3. RESULTS AND DISSCUSSIONS

3. 1. Mechanical Activating In this step, to make sure about that no forming another phase during milling, XRD analysis was taken from sample (Al+ZrO_2) which was milled for 6 hours. Figure 1 shows the XRD pattern of mixture of powders milled for just 6 hours.

The results of XRD pattern show that there is no unwanted phase and only the peaks of Al and ZrO_2 appear. In order to calculate crystallite size, there are many methods such as Scherer, Williamson-Hall, Warren Averbach and Rietveld Refinement Method [7]. After 6 hours of milling, the crystallite size was measured by Rietveld method [8]. In this method, Equation (1) was used to determine the crystallite size

$$D_i = \left(\frac{180}{\pi}\right)\frac{\lambda}{(W_i - W_{std})^{0.5}} \quad (1)$$

In this equation, D_i, λ, w_i and w_{std} are crystallite size, wavelength, the parameter of peak intensity of the analyzed sample and the parameter of peak intensity of the standard sample, respectively. Table 1 gives the results.

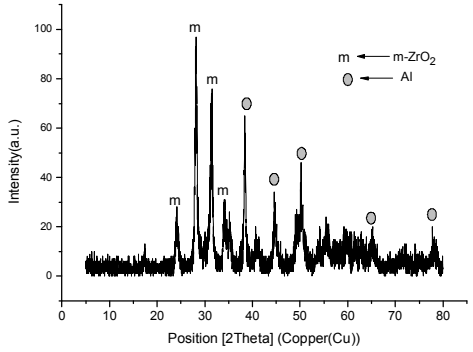


Figure1. XRD pattern of mixture of Al and ZrO_2 powders were milled 6 hours (m: monoclinic-ZrO_2)

TABLE 1. Crystallite size measurement by Rietveld method

Material	Particle size before milling(μm)	Average of crystallite size (nm)
Al	100-200	23
ZrO_2	<300	16

Mechanical alloying can be explained by two mechanisms:

1. Gradual diffusion of components and formation of more products by increasing milling time.
2. Sudden formation of products in a short period of milling time, and consequent occurrence of mechanically alloyed self-sustaining reaction (MSR) [9].

During high-energy milling, the force of the ball to the powder particles makes the powder particles to repeatedly flatten, cold weld, fracture and rewelds. Because of repeated flattening and cold welding, the fine structure has been reviewed and as a result, the distance between layers is reduced. On the other hand, increasing the local temperature during particle collision leads to increase in diffusion rate in the structure [10].

3. 2. Thermodynamic Principles The reaction between aluminum and oxygen are as follows:

$$2Al + \frac{3}{2}O_2 \rightarrow Al_2O_3 \qquad \Delta H^{\circ}_{298} = -1675.7\,\mathrm{KJ/mol} \quad (2)$$

Negative enthalpy change of this reaction shows that this is very exothermic reaction and according to the thermodynamic calculations, the ΔG^{o}_{298} of reaction (2) was calculated (ΔG^{o}_{298}= -1581.77 kJ). This negative value confirms the thermodynamic possibility of the reaction even at low temperatures. Adiabatic temperature was calculated to be 7294°K by using thermodynamic data; this is greater than the critical value (1800K) of self-propagating reaction [11]. So, we can conclude that there action is performed in a self-propagating condition and it only needs basic activation energy which can be supplied mechanically or thermally.

Then, the activated powders were pressed by cold press. In the final stage, tablets were exposed to laser beam in atmospheric oxygen at ambient temperature. Figure 2 shows the XRD patterns for synthesized sample after using laser to expose.

3. 3. Phase Changes Zirconia has three polymorphs: monoclinic, tetragonal and cubic phase. Monoclinic phase with space group of P21/c is stable up to about1478°K and then transforms to tetragonal phase with space group P42/nmc. Tetragonal phase is stable at 2650°K and cubic phase with space group Fm3 is stable up to the melting point, 2983K [12]. The great amount

of heat released due to oxidation reaction of aluminum with oxygen (about -1675.7 KJ.mol^{-1} at 993K) [13] increases the temperature of the sample, and monoclinic zirconia transforms to the high temperature cubic phase.

According to XRD patterns in Figure 2, the alumina, monoclinic and cubic zirconia phases were synthesized. The stabled cubic phase was formed due to high rate of solidification. In other words, synthesized phases had not enough time to change to another phase. Also, alumina phase creates stress to prevent phase retransformation. Thus, the cubic phase stabled at ambient temperature. After synthesis of nanocomposite Al_2O_3-ZrO_2, the crystallite sizes of phases were estimated by using Rietveld method. The results are given in Table 2.

The data in Table 2 shows that the synthesized samples are nanostructure Al_2O_3-ZrO_2 composites. The bulk density of 6 hours milling sample synthesized was measured 3.022 g/cm^3.

The backscattered SEM images and EDAX analyses of Al_2O_3-ZrO_2 nanocomposite synthesized by laser-assisted MASHS, at different milling times, are shown in Figures 3-5.

Morphological observation using SEM with a backscattered mode revealed that with increasing the milling time from 1 to 3 and 6 hours, the morphology was changed from spherical and flower-like to acicular and fine dendrites. The microstructure for Al_2O_3-ZrO_2 nanocomposite is composed of primary α, Al_2O_3-rich phase, (Figure 3b) and (α + β) eutectic phase, in which β is ZrO_2–rich phase. Increasing the milling time causes an increase in the internal energy of nanocomposites, and therefore the equilibrium conditions is diminished. In the 1 hour milled specimen, the milling time is low and the equilibrium conditions can exist. Thus, the stable shape of dendrites inclines to be sphere shaped because of ratio of surface area to volume is lowest (see Figure 3a). With increasing the milling time, the equilibrium condition is not dominant anymore and the stable shape will be non-spherical or acicular shape (Figures 4 and 5).When laser beam is applied for MASHS, synthesis of nanocomposite is rapidly performed and thus, the non-equilibrium situation is established.

TABLE 2. Crystallite size after MASHS synthesis according to nm

Phase	1 h. Milling	3 h. Milling	6 h. Milling
Al_2O_3	58	57.5	60
C-ZrO_2	13	15	13
m-ZrO_2	33	37.5	32.5

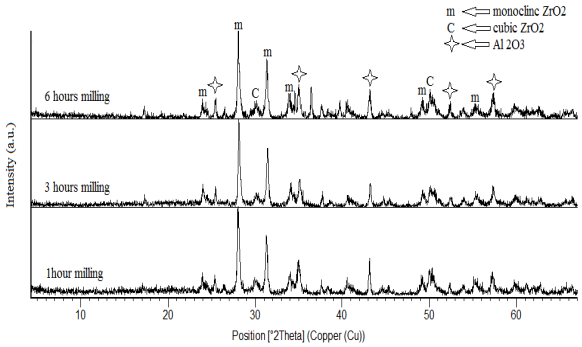
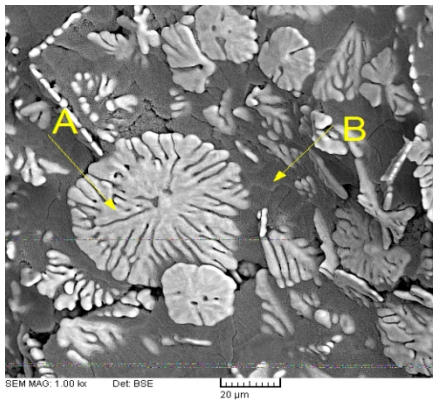
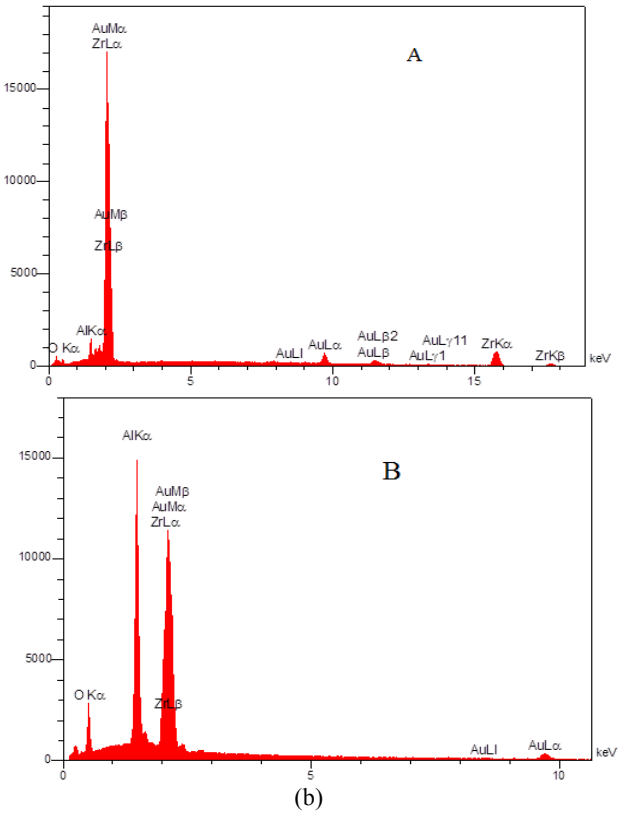


Figure2. XRD patterns of Al_2O_3-ZrO_2composite.



(a)



(b)

Figure 3. Backscattered SEM image (a) and EDAX analyses of regions A and B (b) of powder milled at 1 h.

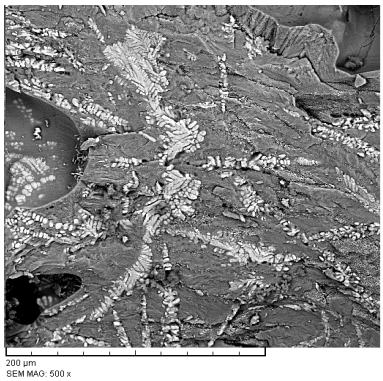
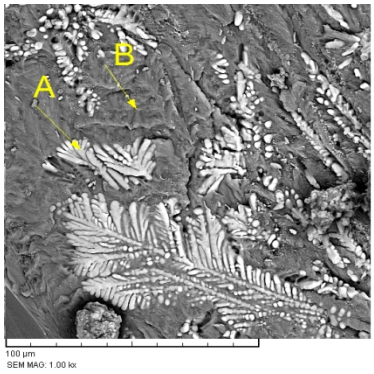
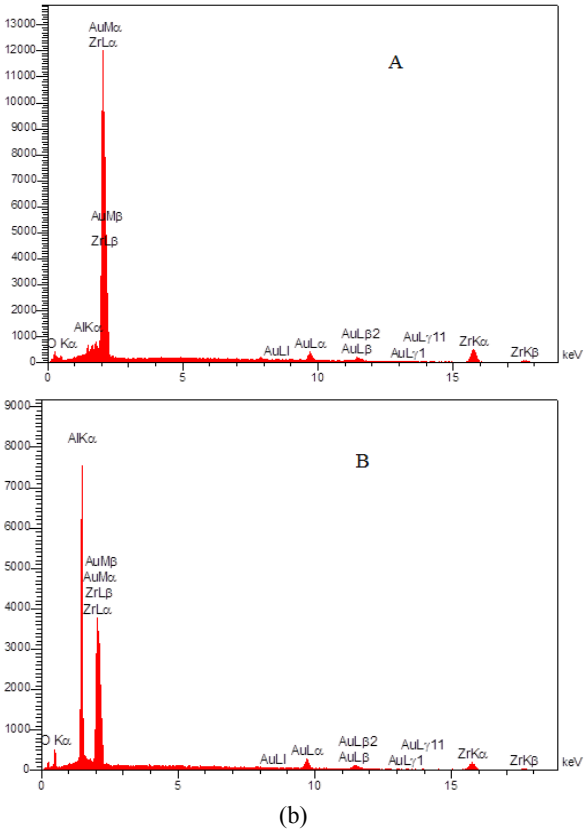


Figure 4. Backscattered SEM image of powder milled at 3 h.



(a)



(b)

Figure 5. Backscattered SEM image (a) and EDAX analyses of regions A and B (b) of powder milled at 6 h.

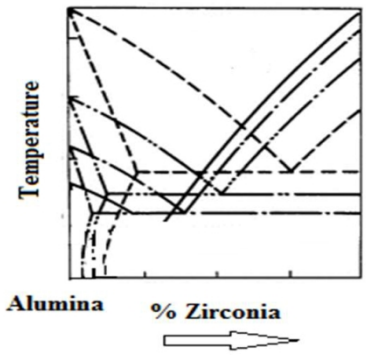


Figure 6. The effect of heating rate on variation of Al_2O_3-ZrO_2 binary phase diagram

Although laser beam has been equally applied on all specimens, the effect of heating rate on the specimens milled for 6 hours is higher than that milled for1 hour; therefore, the heating rate is higher. The non-equilibrium or rapid heating originated by laser beam ignition causes to displace the eutectic point, solidus and liquidus curves. Figure 6 shows the effects of rapid, non-equilibrium, and heating on variation of Al_2O_3-ZrO_2 binary phase diagram. It can be seen from Figure 6 that eutectic point, solidus and liquidus curves move toward ZrO_2 direction significantly with an increase of heating rate. When the heating rate increases, the weight percent of eutectic phase decreases (calculated from Lever rule) and thus, the volume percent of eutectic phase decreases with the milling time (Figures 3- 5).

4. CONCLUDING REMARKS

1. Al_2O_3-ZrO_2 composite was successfully synthesized by laser assisted MASHS.
2. The results of XRD patterns show that the samples have α-Alumina, and also cubic and monoclinic Zirconium oxides phases
3. The results of Rietveld method show the composite is nanostructure and crystallite size smaller than 50nm.
4. SEM images show eutectic structure. By increasing milling time from 1 to 6 hours, particle morphology changed from snowy shape to dendritic structure.

5. REFERENCE

1. Yang, X.-C. and Riehemann, W., "Characterization of Al_2O_3-ZrO_2 nanocomposite powders prepared by laser ablation", ***Scripta Materialia***, Vol. 45, No. 4, (2001), 435-440.
2. Yu, G., Yan, W., Wang, S., Su, B. and Tan, B., "Toughening mechanism of lined Al_2O_3-ZrO_2 multiphase ceramics in SHS

composite pipes", ***Journal of University of Science and Technology Beijing, Mineral, Metallurgy, Material***, Vol. 13, No. 2, (2006), 178-182.

3. Chen, Y.-W., Yen, T.-M. and Li, C., "Preparation of alumina-zirconia materials by the sol-gel method from metal alkoxides", ***Journal of non-crystalline solids***, Vol. 185, No. 1, (1995), 49-55.
4. Mukasyan, A. and Dinka, P., "Novel approaches to solution-combustion synthesis of nanomaterials", ***International Journal of Self-Propagating High-Temperature Synthesis***, Vol. 16, No. 1, (2007), 23-35.
5. Shen, P., Guo, Z., Hu, J., Lian, J. and Sun, B., "Study on laser ignition of Ni-33.3 at% Al powder compacts", ***Scripta Materialia***, Vol. 43, No. 10, (2000), 893-898.
6. McCauley, J. W. and Puszynski, J. A., "Historical perspective and contribution of US researchers into the field of self-propagating high-temperature synthesis (SHS)/combustion synthesis (CS): personal reflections", ***International Journal of Self-Propagating High-Temperature Synthesis***, Vol. 17, No. 1, (2008), 58-75.
7. Klug, H. P. and Alexander, L. E., "X-ray diffraction procedures: for polycrystalline and amorphous materials", ***X-Ray Diffraction Procedures: For Polycrystalline and Amorphous Materials, 2nd Edition, by Harold P. Klug, Leroy E. Alexander, pp. 992. ISBN 0-471-49369-4. Wiley-VCH, May 1974.***, Vol. 1, No., (1974).
8. Rietveld, H., "A profile refinement method for nuclear and magnetic structures", ***Journal of Applied Crystallography***, Vol. 2, No. 2, (1969), 65-71.
9. Mossino, P., "Some aspects in self-propagating high-temperature synthesis", ***Ceramics International***, Vol. 30, No. 3, (2004), 311-332.
10. Suryanarayana, C., "Mechanical alloying and milling", ***Progress in Materials Science***, Vol. 46, No. 1, (2001), 1-184.
11. Yi, H. and Moore, J., "Self-propagating high-temperature (combustion) synthesis (SHS) of powder-compacted materials", ***Journal of Materials Science***, Vol. 25, No. 2, (1990), 1159-1168.
12. Deb, A., Chatterjee, P. and Sen Gupta, S., "Synthesis and microstructural characterization of α-Al_2O_3-ZrO_2 composite powders prepared by combustion technique", ***Materials Science and Engineering: A***, Vol. 459, No. 1, (2007), 124-131.
13. Gaskell, D. R., "Introduction to the Thermodynamics of Materials: CD-ROM, Taylor & Francis US, Vol. 2, (2008).

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چکیده

با توجه به خواص بی نظیر کامپوزیت آلومینا–زیرکونیا (Al_2O_3-ZrO_2) مانند چقرمگی شکست و مقاومت به سایش بالا و ضریب انبساط حرارتی پایین،در این تحقیق نانوکامپوزیت آلومینا–زیرکونیا با روش سنتز خود انتشار دما بالای فعال‌سازی مکانیکی شده به کمک پرتو لیزر موجب افروزش تولید شد. ابتدا پودرهای آلومینیوم و زیرکونیا با نسبت مولی ۱:۱ مخلوط شدند، سپس پودرهای مخلوط شده به مدت زمان‌های ۱ ، ۳ و ٦ ساعت آسیاب و پرس شده و در معرض پرتو لیزر جهت شروع واکنش احتراق قرار گرفتند. به منظور**مشخصه‌یابی** محصولات از آنالیزهای EDX، SEM و XRD استفاده شد. نتایج آنالیز XRD نشان داد که نمونه‌ها حاوی فاز α– آلومینا و فازهای زیرکونیای مونوکلینیک و مکعبی می‌باشد. ساختارهای دندریتی در طی واکنش به وجود آمدند

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