

PHASE ANALYSIS AND PHYSICAL PROPERTIES OF B₂O₃-ADDED ZIRCON CERAMICS SINTERED AT 1300 °C

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ABSTRACT

This study was aimed to know the effect of B₂O₃ (boria) addition on the phase composition and physical properties of zircon ceramics. The raw zircon powder used in the study was a purified natural zircon sand from Kereng Pangi, Central Kalimantan, Indonesia. The zircon ceramics were prepared by a solid state reaction method with variation of B₂O₃ addition of 3 wt%, 6 wt% and 9 wt% and sintered at 1300 °C for 5h. The phase composition, density and microstructure were characterized using X-ray diffraction (XRD), densimeter and Scanning Electron Microscope (SEM), respectively. Vickers Hardness measurement was performed at the polished surface of the ceramics. Results showed that all samples contained pure zircon phase, i.e. there was no effect of B₂O₃ addition on the phase composition. In general, the density and hardness increased with increasing B₂O₃ addition, but addition up to 9 wt% is not optimum to achieve ultra-dense zircon ceramics. Furthermore, the SEM image also showed no significant difference in average grain size. The crystallite size has grown nearly eight times (325 nm) of its original powder. The Vickers hardness of the ceramics is not significantly influenced by the addition of boria. It appears that the boria failure to increase densification also results in the extent of contact between grains which then produces relatively large zircon grains.

Keywords: Ceramic; Zircon; Boria; Natural Sand

Introduction

Ceramics is a material widely used to daily activities. Ceramics became an important material because it is very useful in the industry of mechanical, electronic, refractory, etc. Zircon (ZrSiO₄) is one example of functional ceramics. Zircon has potential as a reliable ceramic material because it has excellent physical properties, such as low thermal conductivity (5.1 W/m °C at room temperature and 3.5 W/m °C at a temperature of 1000 °C), the low coefficient of thermal expansion (4.1 × 10⁻⁶ °C in range 25 and 1400 °C) and high mechanical resistance to heat (more than 1400 °C).¹⁻⁴ Based on these superior thermo-mechanical properties, zircon can be used for various

purposes such as household appliances, industry, electronics, refractories, and space technology.⁵

Other hand, Indonesia is one of the largest country in the world with natural potential, including natural sand. One potential Indonesian natural sand explored are zircon (ZrSiO₄). The existence of zircon sand in Indonesia spread in some regions such as the island of Kalimantan, Riau and Bangka Belitung.⁶ Mining Law on Mineral and Coal in 2009 have restricted the export of raw materials overseas, so it needed a simple way to process natural minerals become more functional materials.⁶ Thus, one solution is to develop a simple technique to process zircon sand into zircon powder and ceramic that has

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the physical properties that make it a functional material.

High density zircon ceramics are still difficult to form. This is due to the high melting temperature of zircon at 2550 °C.⁷ To increase density and reduce porosity of zircon ceramics, some additive materials can be used, including TiO₂,⁸ SiO₂ and Al₂O₃.⁹ The addition of these materials can reduce the sintering temperature, but tends to reduce the value of ceramic hardness.¹ B₂O₃ (boria) was chosen to reduce ceramic porosity. In previous studies, the addition of boria as much as 3 wt% was known to reduce ceramic porosity of ZrO₂-SiO₂ at a temperature of 1200 °C and a density of 3.60 g / cm³ was achieved [10]. On the other hand, the addition of boria also been carried out for the synthesis of forsterite ceramics. With the addition of 4 wt.% Boria, and 3 wt.% Polyvinyl alcohol powder as sintering agent, high density forsterite ceramics were successfully formed with 0.3% porosity at a sintering temperature of 1200 °C.¹¹ From this frame of mind, this study was then designed to offer an opportunity to realize the value added of Central Kalimantan zircon sand as a ceramic-making material with the addition of B₂O₃ and the characterization performed is the structure and mechanical properties of density and ceramic hardness.

Methods

The experiment used zircon natural sand which was taken from Kalimantan Tengah, Indonesia as the raw material. The zircon sand was purified by washing, milling, magnetic separation, soaking in 2M chloride acid, reacting with NaOH and finally drying at room temperature to obtain zircon powder.¹² Furthermore, zircon powder was milled for 10 hours and annealed 2 times to obtain zircon powder ≈40 nm.¹³ Then, the zircon powder is mixed with B₂O₃ of 3 wt%, 6 wt%, and 9% wt using ball milling for 1 hour at 150 rpm. Then, the samples were compacted and sintered at 1300 °C for 5 hours. Afterward, the structure and phase composition of the ceramics were characterized by X-ray Diffraction (XRD).

Zircon crystallite size was determined using the Scherrer's approach.¹⁴

$$D = \frac{\lambda}{H_L - H_{L,S}} \quad (1)$$

where D is the crystal size, λ is the x-ray wavelength, H_L is the Lorentzian component output from Rietica and H_{L,S} is the Lorentzian Component which is the result of the calibration of the test equipment. In this calculation H_{L,S} is used at 0.005.¹⁵ Characterization of physical properties were done using densimeter and Vickers hardness tester to obtain density and hardness. The morphological observation were done using Scanning Electron Microscope (SEM).

Result and Discussion

Fig. 1 presents the XRD pattern (Cu-Kα radiation) of the sintered B₂O₃-added ceramics sintered at 1300 °C. A ceramic sample with 0 %wt B₂O₃ (denoted as M0) was also examined. Phase identification showed that the ceramics exhibited pure zircon (ZrSiO₄ – ICSD 09-582). The highest XRD peak of the ceramics is found at 2θ ≈ 27° for [020] plane, similar to others, e.g. Puclin.¹⁶ It is obvious that B₂O₃ did not react with ZrSiO₄. In addition, B₂O₃ is not detected as a crystalline phase since it melts at approximately 450 °C.¹⁷ Therefore, addition of B₂O₃ has no effect on the phase formation or decomposition of zircon.

The density-porosity and hardness characters of the ceramics are presented in Table 1. In general, the density of the ceramic slightly increases (therefore a decrease in porosity) with the addition of boria. The highest theoretical density of 91.1% has been achieved by 9 wt% boria addition. The densification process began when boria melted at approximately 450 °C¹⁷ far below the sintering temperature, i.e. 1300 °C. However, the melting of boria was not able to effectively fill the pores between zircon grains – see Figure 2. It can be seen from the SEM micrographs that the ceramics are relatively porous. This SEM observation is in agreement with the bulk density measurement in Table 1. Our previous study

showed that the addition of boria could decrease the porosity of forsterite ceramics up to 0,3%.¹¹ Therefore, the addition of boria

increases the density of zircon ceramics, but addition up to 9 wt% is still not optimum to achieve ultra-dense zircon ceramics.

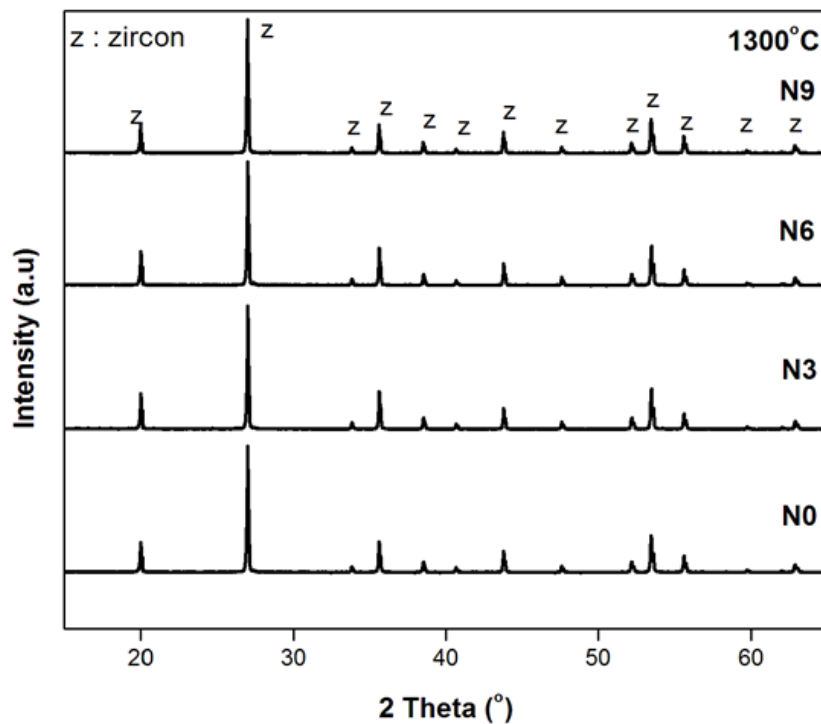


Figure 1. XRD pattern (Cu-K α radiation) of B₂O₃-added zircon ceramics. z = zircon phase (ZrSiO₄).

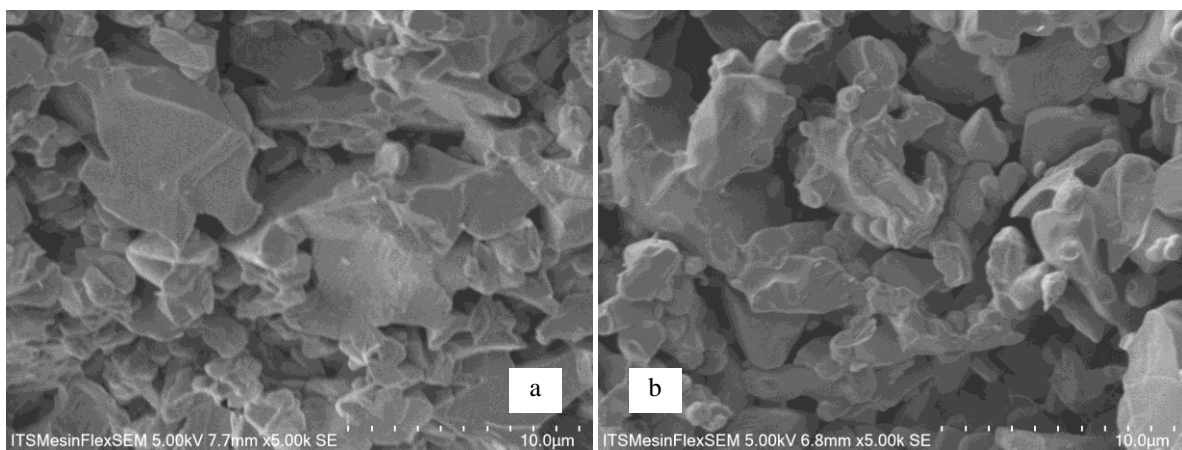


Figure 2. SEM micrographs of fracture of zircon in the B₂O₃-added ceramics. a= sample N0; b= sample N6

Table 1. Density and hardness characteristics of the B₂O₃-added zircon ceramics, as well as zircon crystallite size in the ceramics.

Sample	Density		Zircon crystal size (nm)	Hardness (GPa)
	Bulk (gr/cm ³)	Theoretical (%)		
N0	4.08±0.06	87.4±0.5	291±5	2.0±0.1
N3	4.09±0.06	87.6±0.5	325±5	2.0±0.1
N6	4.22±0.07	90.5±0.5	309±6	2.3±0.1
N9	4.25±0.07	91.1±0.5	326±6	2.2±0.1

Table 1 also shows the zircon crystallite size in the ceramics. The crystallite size has grown nearly eight times of its original powder. The addition of boria, apart from its role to fill the pores between grains, was expected to inhibit the growth of zircon grains (or crystallites) by preventing contacts between them after its melt. It appears that the boria failure to increase densification also results in the extent of contact between grains which then produces relatively large zircon grains.

The Vickers hardness of the ceramics is not significantly influenced by the addition of boria (Table 1). Only 10% increase in the hardness is achieved when boria of more than 3 wt% was added. This low increase in the hardness can be mainly attributed to the porous ceramics.

Conclusion

This study showed that zircon ceramics can be synthesized from natural zircon sand with a solid state reaction. The addition of B_2O_3 has no effect on the phase formation or decomposition of zircon. The density of zircon ceramics increase, but addition up to 9 wt% is not optimum to achieve ultra-dense zircon ceramics. The crystallite size has grown nearly eight times (325 nm) of its original powder. The Vickers hardness of the ceramics is not significantly influenced by the addition of boria. It appears that the boria failure to increase densification also results in the extent of contact between grains which then produces relatively large zircon grains.

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