

## Characterization of Cerate-Zirconate Ceramics Powder Prepared by Three Different Methods: A Comparative Study

(Pencirian Serat-Zirkonat Seramik Serbuk Disediakan oleh Tiga Kaedah yang Berbeza: Suatu Kajian Perbandingan)

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### ABSTRACT

Ceramics powder of  $BaCe_{0.54}Zr_{0.36}Y_{0.1}O_{2.95}$  (BCZY) was synthesized using three different methods namely sol-gel (SG), supercritical fluid (SC) and supercritical fluid assisted sol-gel (SCSG). The respective prepared samples were denoted as S1, S2 and S3. TG thermogram of the dried powders for all samples showed three stages of weight loss. Each stage was corroborated by one or two exothermic peaks as shown in DTG signal. Complete thermal decomposition for all the samples was almost accomplished at 1000°C for about 2 h. At calcination temperature of 1100°C, S1 showed a single-phase of perovskite-type oxides as proven by XRD result. Morphology of the calcined powders by SEM micrograph showed that S1 is in spherical shape, S2 is in cubic structure and S3 has a mixture of spherical and rod-like structure. Therefore, as comparison, SG method gives better characteristics of cerate-zirconate ceramics powder compared to SC and SGSC.

*Keywords:* Cerate-zirconate ceramic; sol-gel; supercritical fluid; supercritical fluid assisted sol-gel

### ABSTRAK

Serbuk seramik  $BaCe_{0.54}Zr_{0.36}Y_{0.1}O_{2.95}$  (BCZY) telah disintesis menggunakan tiga kaedah yang berbeza iaitu sol-gel (SG), bendalir lampau genting (SC) dan bendalir lampau genting dibantu sol-gel (SGSC). Sampel masing-masing telah ditandakan sebagai S1, S2 dan S3. Isyarat TG bagi semua serbuk sampel yang dikeringkan telah menunjukkan tiga peringkat kehilangan berat. Setiap peringkat kehilangan diiringi oleh satu atau dua puncak eksotermik seperti ditunjukkan oleh isyarat DTG. Penguraian terma yang lengkap untuk semua sampel didapati hampir sempurna pada suhu 1000°C selama 2 jam. Pada suhu pengkalsinan 1100°C, S1 menunjukkan satu fasa tunggal oksida perovskite seperti dibuktikan oleh analisis XRD. Morfologi serbuk yang dikalsinkan menggunakan mikrograf SEM mendedahkan S1 adalah dalam bentuk sfera, S2 berbentuk kubus dan S3 adalah campuran bentuk sfera dan rod. Sebagai perbandingan, didapati kaedah SG memberi ciri serbuk seramik serat-zirkonat yang lebih baik daripada SC dan SGSC.

*Kata kunci:* Cecair genting lampau; cecair genting lampau membantu sol-gel; serat-zirkonat seramik; sol-gel

### INTRODUCTION

Ceramic perovskite-type oxides are well known as proton conductor in atmosphere containing hydrogen and/or water vapour at elevated temperatures. Typical examples of proton conducting ceramics based on perovskite-type oxides are  $BaCeO_3$ ,  $SrCeO_3$ ,  $BaZrO_3$ ,  $SrZrO_3$ , and  $CaZrO_3$ . The first two showed higher conductivity and the latter are superior with respect to their chemical and mechanical stability. However, substitution of zirconium at cerium sites or vice-versa to become cerate-zirconate compound is known to improve proton conductivity as well as chemical and mechanical stability (Fabbri et al. 2008). Cerate-zirconate powders have been mostly prepared by solid-state reaction (SSR) method which requires repeated ball milled, fired at high temperature and needs longer annealing time. These process cannot control the microstructure, grain size distribution, or shape of the resulting powders leading to hard agglomeration and contamination (Ryu & Haile 1999). Compared to SSR, wet chemicals methods (WCMS) are preferable to synthesize ceramics compound

(Lazarević et al. 2010; Piticescu et al. 2006). One of the most attractive methods is a modified Pechini or sol-gel because the resulting powders have well crystallized structure and narrow size distribution due to the mixing of reagents at molecular or atomic level. However, a complete decomposition of the citrate complex takes a very long time and several washing steps are necessary to remove additional phase of  $BaCO_3$  that is still presence even after calcined at  $T \geq 1100^\circ\text{C}$  (Zhong 2007).

In order to overcome the disadvantages of the SSR and WCMS, supercritical fluids can be adopted as another potential method to produce better characteristics of ceramic powders. A few reports demonstrate the synthesis of ceramics compound using supercritical fluids involving growing the particles in controlled manner to attain the desired morphology unlike conventional particle formation methods. Supercritical (SC) fluids have been used in synthesis and processing metal oxides as they have tuneable properties which enable the control of parameter in reaction media. The high diffusivity of the

reactants in SC medium, fast reaction rate and high degree of supersaturation due to low solubility of starting materials makes SC fluids as one of the alternative methods for many researches worldwide (Bozbag & Erkey 2012; Byrappa et al. 2008). A variety of supercritical fluids for examples carbon dioxide, alcohols and water have been used to synthesis ceramic materials. Particularly, alcohols have been proven to be efficient in the preparation of diverse ceramic oxides. It is believed that a low carbon number alcohol such as methanol or ethanol acts both as solvent and reducing agents at SC condition. The vital stage in this SC batch process is to eliminate the carbonate impurities by raising fluids temperature and pressure in order to obtain a crystalline powder through the effect of SC alcohol as reaction media.

Recent studies have demonstrated the efficiency of assisted SC fluid synthesis to produce ultra-fine ceramic powders at lower temperature (Ru et al. 2008). Most of the works are reported for the synthesizing ceramics compound with three metal elements (Aymonier et al. 2006; Kim et al. 2009). However, to the best of our knowledge this route has not yet been carried out to synthesize ceramics compounds with four metal elements. Considering the advantages of WCMS and SC fluids properties, an approach towards the combination of both synthesis routes was used in this work.

## METHODS

### CHEMICALS

Precursors used for the three methods of sample preparation were metal nitrate salts. A stoichiometric amount of  $\text{Ba}(\text{NO}_3)_2$  (99%, ACROS),  $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$  (99.5%, ACROS),  $\text{Zr}(\text{NO}_3)_2 \cdot x\text{H}_2\text{O}$  (99.5%, ACROS) and  $\text{Y}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$  (99.9%, Aldrich) were used as starting materials. The chelating agent used for sol-gel method was triethylenetetramine, TETA (60%, ACROS). Ethanol (95%, HmbG) was used as the solvent and reaction media in supercritical fluids method. The samples for sol-gel, supercritical fluids and sol-gel assist supercritical fluids were denoted as S1, S2 and S3, respectively.

### PROCEDURE

*Sol-gel Method (S1)* A stoichiometric amount of starting materials were dissolved in deionized water and continuously stirred on a hot-plate to make a transparent nitrate solution. During stirring, TETA was slowly added into metal nitrate salts solution. Ethylene glycol was then added into the complex solution to adjust the viscosity of the solution and control the moving velocity of the metal ions, resulting in the gelation of reaction mixture. A concentrated ammonia solution was used to neutralize the complex solution as reported by Osman et al. (2006). The gel was dried at  $325^\circ\text{C}$  in a furnace to yield black flakes. Then the sample was calcined at  $1100^\circ\text{C}$  with heating rate of  $10^\circ\text{C min}^{-1}$  for 10 h to produce light-yellow powder.

*Supercritical Method (S2)* Each metal nitrate salts were dissolved in stoichiometric amount with ethanol. The raw materials were loaded inside a 350 mL vessel and were synthesized by a high pressure-high temperature (HP-HT) batch wise reactor system. The temperature and the pressure were fixed at  $300^\circ\text{C}$  and 11 MPa, respectively, using a controller. The amount of solvent depends on the percentage of liquid level filling or percent of reactor-free volume as reported by Ishak (2007). The facility involved in monitoring the parameter are flow-meters, temperature controller (thermocouple type-K located inside the vessel) and a back-pressure regulator (Unijin). After a fixed period, the vessel was transferred into the water bath to quench the reaction immediately. The obtained slurry was subjected to a vacuum suction to separate solid from the slurry. Rotary evaporator was used to evaporate the remaining solvent and the solid product was further calcined at  $1100^\circ\text{C}$ .

*Sol-gel Assisted Supercritical Method (S3)* The dried as-synthesized S1 powder ( $T = 325^\circ\text{C}$ ) and ethanol were loaded in the 350 mL vessel. The temperature and pressure were fixed at  $300^\circ\text{C}$  and 11 MPa, respectively. The operating condition and controlling parameter are the same as supercritical method as discussed in S2.

## CHARACTERIZATION METHODS

The thermal decomposition process for the as-synthesized dried BCZY powders was analyzed by thermogravimetric analyzer (TA Instrument, SDT Q600). Approximately 15-18 mg of the dried powder was placed in an alumina crucible pan and gradually heated from 25 to  $1000^\circ\text{C}$  with heating rate of  $10^\circ\text{C/min}$  under purified air. The infrared spectroscopy experiment was performed in an inert atmosphere at room temperature using FT-IR Nicolet 380. The IR spectra was used to observe the formation of metal oxide as well as the remaining carbonate ions of the as-synthesized and calcined powders from 400-4000  $\text{cm}^{-1}$ . The phase identification of samples was carried out by X-ray diffraction (Shimadzu XRD-6000) equipped with Ni-filtered and graphite monochromatized  $\text{Cu-K}_\alpha$  radiation ( $\lambda = 1.5406 \text{ \AA}$ ) over a  $2\theta$  range of 10 to  $80^\circ$  with steps size at  $0.02^\circ/\text{s}$ . Morphology of the platinum-coated calcined powders was observed using scanning electron microscope (SEM) model JOEL JSM-6460 operating at 15 kV.

## RESULTS AND DISCUSSION

### THERMOGRAVIMETRIC ANALYSES (TGA)

The thermal decomposition process of  $\text{BaCe}_{0.54}\text{Zr}_{0.36}\text{Y}_{0.1}\text{O}_{2.95}$  (BCZY) dried powder was recorded by TG and DTG profile as shown in Figure 1. All the samples exhibit three stages of weight losses. The first stage of weight loss for S1 and S3 is corresponded to the presence of moisture in the samples. It can also be related to the complete chelation process with the loss of  $\text{NO}_x$  gasses. For S2, the first stage of weight loss  $\approx 4\%$  in the range of 25- $133^\circ\text{C}$  is associated

to the initial dehydration reaction of moisture and ethanol (boiling point of EtOH  $\approx$  78°C).

The larger weight loss for S1 (85%) and S3 (80%) were observed at second stage which is in the temperature range of 210-600°C and 235-590°C, respectively. The weight loss in this step as also corroborated with sharp exothermic peaks is due to the oxidation and combustion of metal-chelate complex and nitrates residue by releasing the H<sub>2</sub>O, CO<sub>2</sub> and NO<sub>x</sub> gasses (Singh et al. 2007). For S2 in temperature range from 133-410°C, with a weight loss around 15% can be attributed to combustion of ethanol excess and decomposition of high boiling point organic complexes. A small exothermic peak at 380°C is related

to the combustion of residual ethanol. This result is in-line with the work by Ru et al. (2008) for the yttrium aluminium garnet (YAG) sample using supercritical carbon dioxide/ethanol fluid drying method.

The thermal decomposition for all samples was almost completed at 410-970°C (weight loss  $\approx$  2%) due to the decomposition of remaining CO<sub>2</sub> and NO<sub>2</sub> as well as carbonaceous in the compound may burnt off (Osman et al. 2006). The amount of the excess organic released for S1 is the highest compared to S2 and S3 as summarized in Table 1. No further weight loss at T=1000°C indicates that the thermal decomposition of reaction powder might be completed prior to the formation of oxide compound.

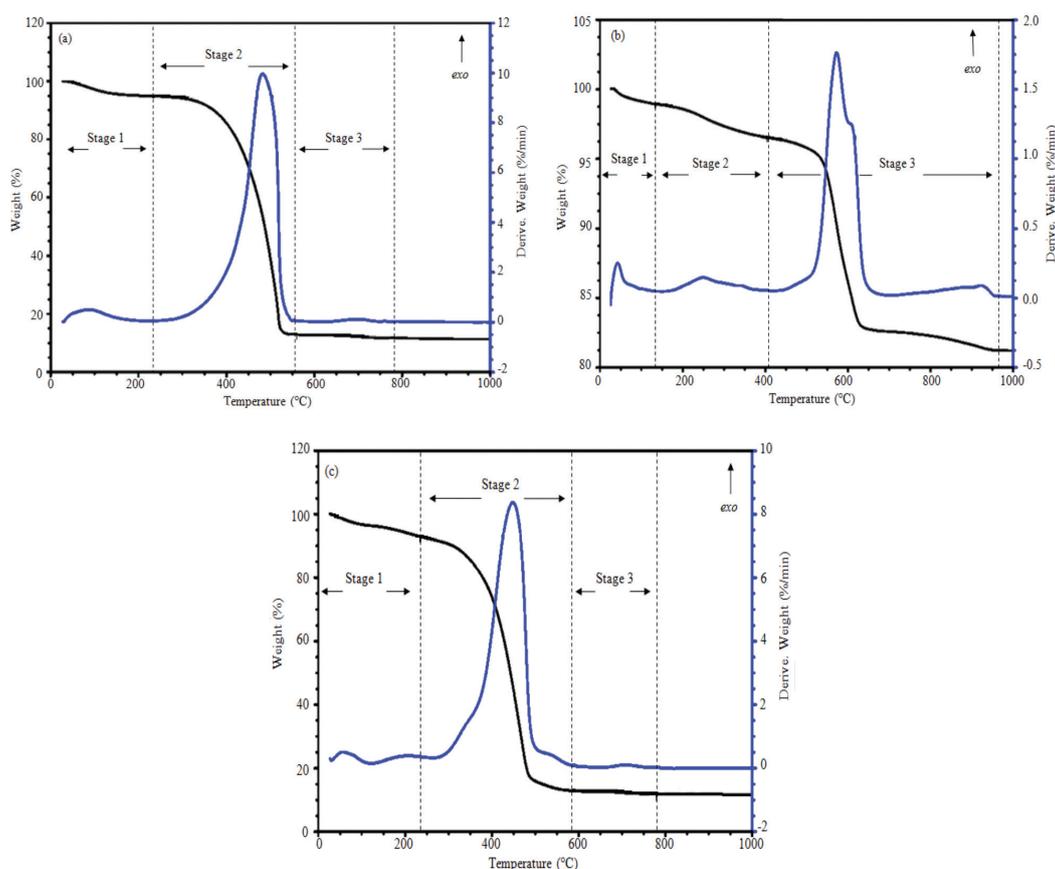


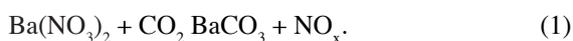
FIGURE 1. TG/DTG curves of BCZY as-synthesis dried powders (a) S1, (b) S2 and (c) S3

TABLE 1. The summarization of thermal characteristics for dried powders ( $T \leq 350^\circ\text{C}$ )

Sample	Stage	Temperature range (°C)	Mass loss (%)	Total mass loss (%)
S1	1	27 – 210	8	94
	2	210 – 600	85	
	3	600 – 800	1	
S2	1	25 – 133	4	21
	2	133 – 410	15	
	3	410 – 970	2	
S3	1	23 – 235	7	88
	2	235 – 590	80	
	3	590 – 780	1	

## FOURIER TRANSFORM INFRARED (FTIR) SPECTROSCOPY

FTIR spectra for all the samples after treated at  $T = 300^\circ\text{C}$  and  $1100^\circ\text{C}$ , respectively, are shown in Figure 2. At low synthesizing temperature, the spectra in Figure 2(a) showed the existing of various organic compounds. Transmittance bands around  $\approx 1650$ - $1644$  and  $951\text{ cm}^{-1}$  are related to the characteristic asymmetrical split stretching of carbonate compound from  $\text{CO}_2$  reacted with  $\text{Ba}(\text{NO}_3)_2$  that generate  $\text{BaCO}_3$  (Wang et al. 2011). The possible reaction is shown in (1). The formation of metal complexes that derived from chelation and polymerisation process for S1 and S3 are observed at  $1448$  and  $1461\text{ cm}^{-1}$ , respectively. Similar characteristics are also observed for S2 at  $1553$  and  $1380\text{ cm}^{-1}$  which represents the nitrate group contains in each metal salts (C-N) (Nityanand et al. 2011). The transmittance band at  $566$ - $447\text{ cm}^{-1}$  indicates the presence of metal oxide (M-O) in the samples.



In Figure 2(b), clearly seen low transmittance intensity of carbonate residue which are at  $1644$ ,  $1642$  and  $867\text{ cm}^{-1}$ , respectively, appeared for all samples. The traces of carbonate residue are influenced by the high firing temperature and incomplete combustion. This is due to the adsorption of  $\text{CO}_2$  on the powder surface which is transformed to  $\text{CO}_3$  during calcination process. In S1 and S3, the chelating agent used does not contained any  $-\text{COOH}$  groups, however a small intensities of carbonate residue exists at  $1644$  and  $1642\text{ cm}^{-1}$  suggest that carbonate ion from the incomplete combustion reacted with  $\text{Ba}(\text{NO}_3)_2$  or  $\text{Ba}^{2+}$  to become  $\text{BaCO}_3$ . Since carbonate compound is stable in air, a high firing temperature as high as  $1400^\circ\text{C}$  is needed to eliminate this impurity (Chakroborty et al. 2002). Trace amount of nitrate ions also detected by the appearance of peak at  $\sim 1456$ ,  $1430$  and  $1466\text{ cm}^{-1}$  for every respective samples. The metal-oxide bands at  $459$ - $550\text{ cm}^{-1}$  become

stronger as the calcination temperature increased for all the samples.

## X-RAY DIFFRACTION (XRD)

Figure 3 shows the XRD pattern of cerate-zirconate powder after calcined at  $T = 1100^\circ\text{C}$ . XRD patterns for S2 and S3 showed a mixture of respective  $\text{BaZrO}_3$ ,  $\text{BaCeO}_3$ ,  $\text{CeO}_2$  and  $\text{BaCO}_3$ . For example a peak for  $\text{BaCeO}_3$  at  $2\theta = 28.5^\circ$  (JCPDS card- 220074) and  $\text{BaZrO}_3$  at  $2\theta = 30^\circ$  (JCPDS card- 60039) are clearly seen in the spectrum. S1 however, showed a well develop single-phase perovskite-type oxide of BCZY compound. The pronounced peaks can be indexed to (110), (111), (200), (210), (211), (220) and (310), respectively, according to JCPDS card- 892485. High intensity of perovskite phase using TETA as chelating agents suggest the complete substitution of Zr at Ce sites at  $T = 1100^\circ\text{C}$  (Abdullah et al. 2012). But the appearance of small residue of carbonates is still detected at  $2\theta \approx 26^\circ$  which is in agreement with FTIR result as discussed earlier.

## SCANNING ELECTRON MICROSCOPE (SEM)

The particle morphology of the calcined powder ( $T = 1100^\circ\text{C}$ ) that was observed by SEM shows S1, S2 and S3 in spherical shapes, cubic formation and mixture of rod-like and spherical shape, respectively (Figure 4). The high pH value of chelating agent (TETA  $\approx \text{pH} 11$ ) in both S1 and S3 preserve the molecular level mixing of metal ions besides enhance the homogenous dispersion of metal nitrate. This result is also in-line with the work by Patra et al. (2011). They reported that the sample with high pH value produces more number of spherical particles. S2 cubic shape was observed from the high pressure SC medium in the absence of TETA. Clearly seen the powder is partially agglomerated. The formation of cubic structure can also be related to the fragmentation of spherical particle into small cubes upon calcination according to the studies by Byrappa et al. (2008). Besides that, Cabanas et al. (2007) also managed

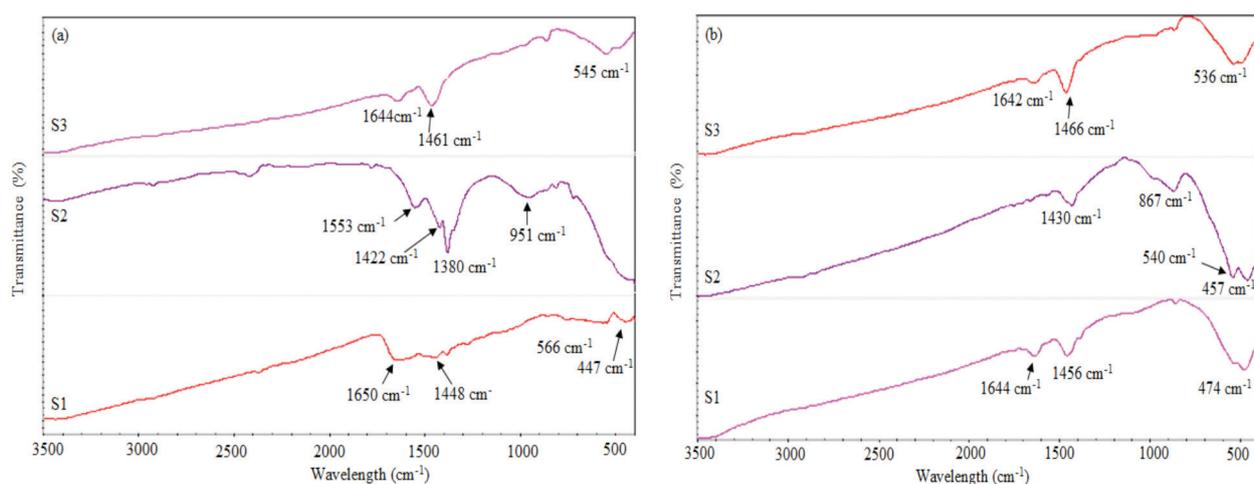


FIGURE 2. FT-IR spectra for (a) dried powder ( $T = 300^\circ\text{C}$ ) and (b) calcined powder ( $1100^\circ\text{C}$ )

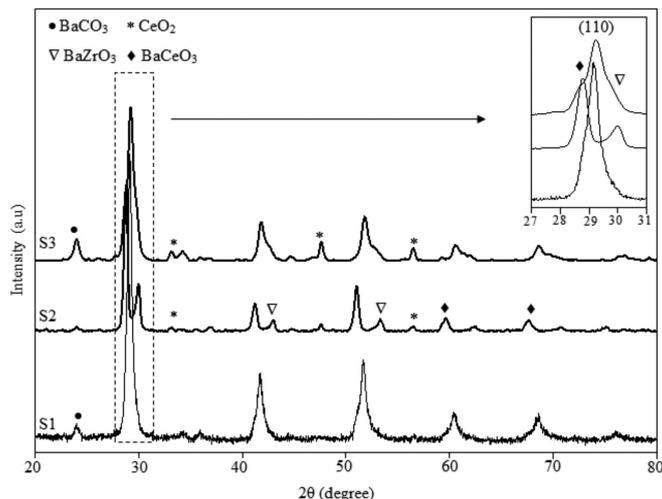


FIGURE 3. XRD patterns for BCZY powder after calcined at  $T=1100^{\circ}\text{C}$

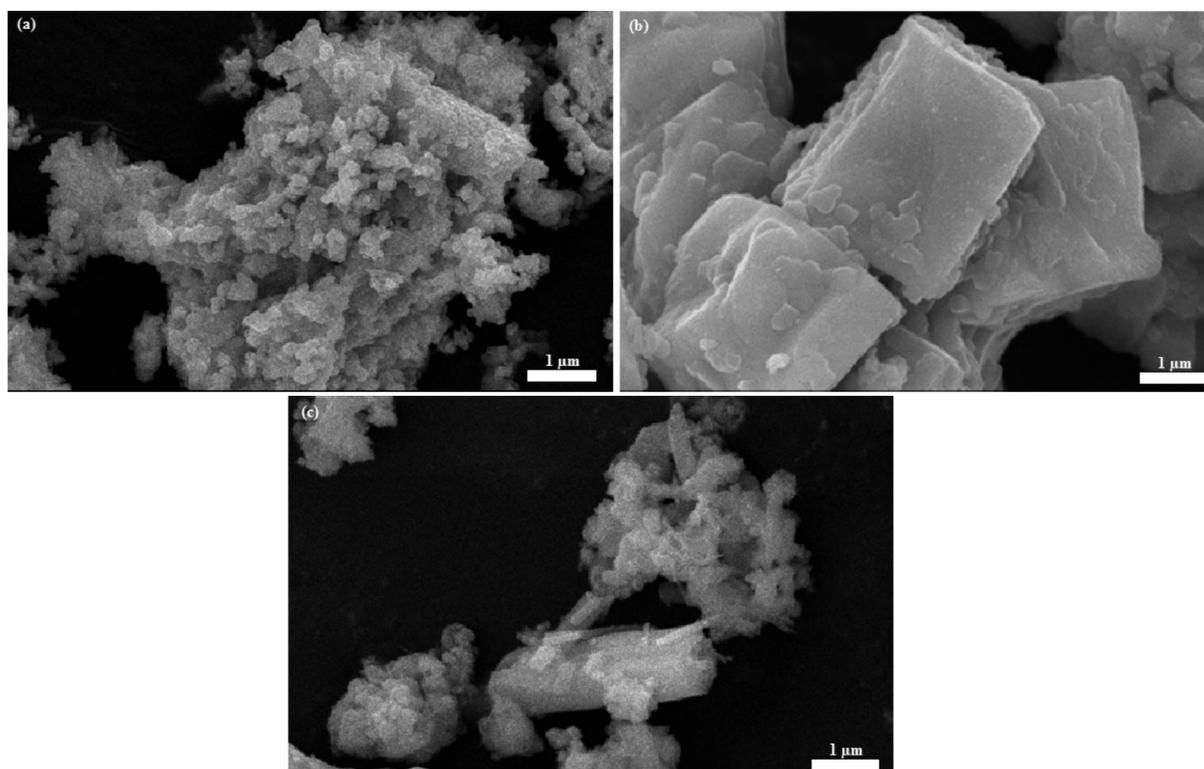


FIGURE 4. SEM image of BCZY powder after calcined at  $1100^{\circ}\text{C}$  for (a) S1, (b) S2 and (c) S3

to compose highly cube-like YAG particles in the highest Sc EtOH content. On the other hand, S3 shows a mixture of spherical and rod-like BCZY powder. The alteration of certain number of spherical powder into rod-like shape might be due to the supersaturation and nucleation or particle growth during supercritical fluid synthesis. The interaction of particles with liquid solvent at high pressure can result in fusion of small particles to create an irregular shape. The partial agglomerate of S3 probably because of Ostwald ripening which the small particle grows due to high solubility of solvent. In addition, the high calcination

temperature used also fuses the small particle to produce large agglomerate powder (Singh et al. 2009).

#### CONCLUSION

Sample of  $\text{BaCe}_{0.54}\text{Zr}_{0.36}\text{Y}_{0.1}\text{O}_{2.95}$  (BCZY) was prepared via a sol-gel method, supercritical ethanol and supercritical ethanol assisted sol-gel method. At calcination temperature of  $1100^{\circ}\text{C}$ , the small amount of carbonate impurities still present in the samples as proven by FTIR and XRD results. The used of TETA for S1 effectively result in a single-phase

of BCZY perovskite type-oxide. SEM analysis showed S1 powder is in spherical shape, S2 in cubic shape and S3 consists of a mixture between spherical and rod-like structure. Further study on the electrical properties of the BCZY prepared with TETA is still in progress and will be reported elsewhere.

#### ACKNOWLEDGEMENTS

The authors would like to thank the Ministry of Education (MOE), Malaysia for the Exploratory Research Grant Scheme (ERGS/2013) and Universiti Teknologi MARA for the Internal Grant (PSI/2013).

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Received: 15 May 2013

Accepted: 1 May 2014