



FABRICATION OF BaTiO₃ THICK-FILM LEAD-FREE PIEZOELECTRIC CERAMIC BY USING SCREEN PRINTING METHOD

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ABSTRACT

This paper describes the fabrication process of thick-film lead free piezoelectric ceramics, consisting of barium titanate (BaTiO₃), borosilicate, terpineol, on alumina (Al₂O₃) substrate. The BaTiO₃ thick-films piezoelectric are prepared by screen printing. Suitable ratio for the paste composition is crucial to ensure printability and functionality of lead-free piezoelectric ceramic. Ag/Pd is deposited as the lower and upper electrodes to form a unimorph piezoelectric device. The thick-film which contained 50wt. % BaTiO₃, 10wt.% borosilicate, and 20wt.% terpineol demonstrated the optimal piezoelectric properties with the maximum piezoelectric charge coefficient of 17 pC/N. The thick-film was also inspected under SEM to investigate the quality of the films.

Keywords: thick-film technology, lead-free piezoceramic, piezoelectric charge coefficient d_{33} .

INTRODUCTION

The piezoelectric materials can be categorized into two classification, one of them is Pb-based piezoelectric and the other is non Pb-based piezoelectric. Potential piezoelectric material option exist in either natural crystal (e.g. quartz) or polycrystalline ceramic (e.g. PZT) or polymer form (e.g. PVDF). Lead-based piezoelectric ceramic such as lead zirconatetitanate (PZT) have been widely used as piezoelectric sensor and piezoelectric actuator because of their excellent piezoelectric properties (Chen *et al.*, 2013). However, due to the environmental and health related hazards concern, legal restriction are being taken to reduce or eliminate the use of lead in electronic products (Alkoy & Dursun, 2012). The lead-based piezoelectric ceramics containing PbO more than 60 wt% can cause serious environment pollution and human health problems because of its high toxicity (Cui *et al.*, 2012). Nowadays, advanced materials have been widely studied for consumer, healthcare and military applications, where it enable the design of garments incorporating distributed non-obtrusive sensors (Pons *et al.*, 2007). Therefore, extensive studies and investigation have been conducted on Pb-free piezoelectric ceramic materials for environment friendly and the replacement in electronic products (Cheng *et al.*, 2014) (Byeon and Yoo, 2014).

Barium Titanate (BaTiO₃) is perovskite family with an inorganic compounds and the empirical formula, ABO₃. It is a good candidate for non Pb-based material among the lead-free substances (such as Potassium Niobate (Byeon and Yoo, 2014), and Polyvinylidene Fluoride (Arlt and Wegener, 2010)) for a variety of applications that avoid toxicity of lead-based materials. BaTiO₃ has been studied widely for its structural transition (Cheng *et al.*, 2014) and applications (Ali *et al.*, 2013). Therefore, development of alternative method that can produce a smart material integrated for electronic functions play an important role in lead-free piezoelectric devices. BaTiO₃ is considered as one of the most promising candidate materials for the replacement of Pb-based piezoelectric materials because of its exhibit piezoelectric

properties (Jiang *et al.*, 2014). Recent reports of BaTiO₃ materials remains a major scientific challenge in requirements to develop high piezoelectric sensitivity and composition of lead-free piezoelectric ceramics (Ali *et al.*, 2013). It was reported by (Bai *et al.*, 2012), the lead-free piezoelectric ceramics shows high piezoelectric properties but it limited on bulk ceramic devices.

Ceramic thick-film technology refers to the technology involved deposition of cement based pastes screen printed onto a flat base material (substrate) through the use of a fine woven mesh with desired geometry and then fired at temperatures in the region of 800 – 1000 °C (Kok *et al.*, 2009) (Wu *et al.*, 2011) (Wei *et al.*, 2012). Screen printing technique is the oldest yet simple forms of graphic art reproduction. Fabrication piezoelectric materials usually consist of at least two physical and chemically different materials as states by (Stojanovic *et al.*, 2007) (Fu *et al.*, 2014) (Vijatović *et al.*, 2010) (Zhang and Hao, 2014) (Park and Park, 2011), there are active material, binder and solvents.

Many studies have been reported on the preparation piezoelectric thick films by screen printing, tape casting, composite sol-gel, aerosol spray deposition, electrophoretic deposition, sputter coating, polymeric precursor and reactive-template grain growth (RTGG) method (Bai *et al.*, 2012) (Wu *et al.*, 2011) (Zhang *et al.*, 2010) (Hu *et al.*, 2014). Recently, researchers used a thick film technology in fabricating the BaTiO₃ lead-free piezoelectric ceramic by a screen printing method. However, the thickness range is the primary reason for their piezoelectric properties notable lack of availability (Stojanovic *et al.*, 2002).

The majority of this research is the fabrication of thick-film lead-free piezoelectric ceramic by using screen-printing technique. This process is useful to fulfill the demands for miniaturization, multilayer assemblies and circuit complexity. Thus, this paper reports screen printing unimorph design on alumina substrate with a lead free material piezoelectric layers with the difficulties on the preparation of BaTiO₃ thick-films. The aim of this work is



to characterize electrical properties BaTiO₃ based thick-films piezoelectric ceramics via thick-film technology. The microstructure quality and the electrical properties of the thick-film were analyzed.

Material and preparation of BaTiO₃ piezoelectric films

The lead-free piezoelectric ceramic powders with ratio of 5:2:1 were prepared by thick-films technology. The powders of all chemicals, BaTiO₃ (<3μm, 99%), BaTiO₃ (<2μm, 99.9%), BaTiO₃ (<100nm, 99.9%), borosilicate glass (7056) and terpeneols (99%) were obtained from Sigma-Aldrich. The raw powders were accurately weighed with different amounts according to ratio as shown in Table-1. The first step in fabrication the lead-free thick-film piezoelectric ceramic is the formulation of the powder into pastes. Thus, the mixtures for the lead free piezoelectric pastes are made from the combination of functional powder material, solvent and permanent binder.

Table-1. Material weight ratio 5:2:1 of the lead free piezoelectric ceramic powder.

Material	Function	Weight, %
BaTiO ₃ (<3μm, 99%)	Active material	30
BaTiO ₃ (<2μm, 99.9%)	Active material	15
BaTiO ₃ (100nm, 99.9%)	Active material	5
Borosilicate glass (7056)	Binder	10
Terpineol (alpha)	Solvent	20

The primary powders were fully mixed through dry-milling process with mortar and pestle. Then, the primary powders were ball-milled with the addition of borosilicate glass powder for 30 min. The mixtures of the raw powder materials were homogenized together through a triple roller millin wet-milling process with alpha-terpineols solvent for 1 hour, where it formed a printable paste. The viscosity of the prepared screen printable pastes were measured by viscometer and adjusted in the range 20 to 60 Pa.

Screen printing method of BaTiO₃ thick film

Screen-printing is a printing technique that uses a woven mesh to support an inks or pastes medium blocking stencil, as shown in Figure-1. The equipment for fabrication via thick-films technology required inks, which are the piezoelectric pastes, squeegee, image unimorph design, photoemulsion (woven mesh) and screen, mesh frame. The screen printing process was carried out using a fine woven mesh that was mounted on a metal frame. The screen printable pastes were transported over the screen by a squeegee. A squeegee was moved across the screen stencil, forcing the pastes past the threads of the woven mesh.

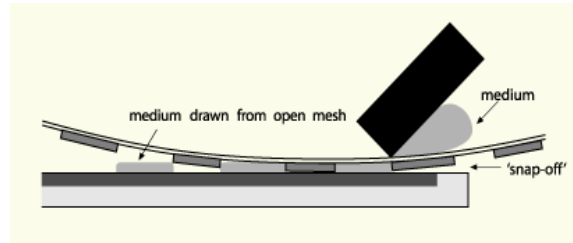


Figure-1. Screen printing method of thick-film lead-free piezoelectric ceramics.

Initially, the bottom electrode was deposited with silver/palladium (Ag/Pd) slurry directly on 96% alumina (Al₂O₃) substrate (40 mm x 10m x 0.5 mm) substrate in the predefined geometric pattern. After printing, the films were dried off with 60°C exposure of infrared light for 15 min. Then, the BaTiO₃ piezoelectric layers were screen-printed with a 280 mesh screen mask to the underlying bottom electrode. The BaTiO₃films were left at room temperature one minute in order to level out and dry. The thick film was dried offwith 60°C exposure of infrared light for 1 hour. This process was repeated twice with the second layer of BaTiO₃thick film. Before deposition of top electrode, a heat treatment for firing condition was carried out in the thick films according to the temperature profile in Figure-2.

Table-2. Summary of printed layer drying process parameters.

Layer	Drying conditions	Duration
Bottom electrode	Infrared, 60°C	15 min
Active layer	Infrared, 60°C	1 hours
Top electrode	Infrared, 60°C	15 min

The top electrode layer was screen printed similarly with the bottom electrode through desired pattern on the layer of BaTiO₃thick film. The BaTiO₃thick film was sandwiched in between bottom electrode and top electrode. The drying process was performed in air at 60°C using a homemade box with infrared light distance 20 cm fromBaTiO₃thick film. The printed layer was dried for certain duration according to the material that deposited on the Al₂O₃ substrate as stated in Table-2. The final heat treatment of firing condition was carried out for whole BaTiO₃thick-film piezoelectric ceramic cantileverin air. The variable temperature ranging from 850°C to 1050°C.

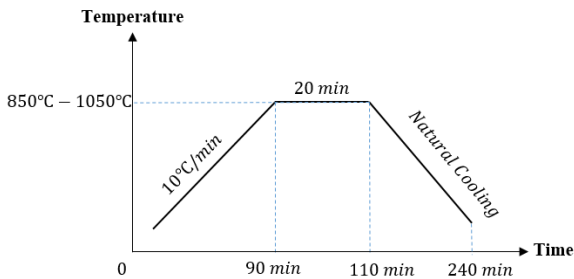


Figure-2. Temperature profile of the BaTiO₃ thick-film piezoelectric ceramics enabled by firing condition.

The thick-film usually consists of several layers of material in order to obtain the desired thickness. The number of printed layers of film was shown in Table-3. The bottom and top electrode of thick film consisted of one screen-printed layer which was approximately 10-15 μm . The specimen dimension for the BaTiO₃ thick-film piezoelectric ceramics measurements were shown in Table-4 with 0.5 mm of and 0.2 mm thickness for the alumina substrate and sandwich layer of barium titanate (BaTiO₃) ceramics, respectively. The films dimensions depend on the geometric design on the woven mesh of the screen.

Table-3. Material layers of the lead free piezoelectric ceramic membrane structures.

Layer	Role	Prints
Al ₂ O ₃	Substrate	-
Ag/Pd	Bottom electrode	1
BaTiO ₃	Active layer	2
Ag/Pd	Top electrode	1

Electrical poling was performed by heated on the hot plates with applied external electric fields. The BaTiO₃ thick-film piezoelectric ceramics were polarized in direct contact. For short-term 15 minutes poling duration, the BaTiO₃ thick-film piezoelectric ceramics were poled with ranging temperature 60°C to 160°C and the applied electric fields at 20 kV/cm. The static piezoelectric charge coefficients (d_{33}) were characterized by a direct method based on ZJ-6B Quasi-static d_{33} meter using the Berlincourt Method. Scanning electron microscopy (SEM) investigations were performed in order to record the microstructure of the BaTiO₃ thick-film piezoelectric ceramic and to analyse the particle size of the BaTiO₃ pastes deposited on the bottom electrode. In addition, the film-thickness was determined using ZEISS EVO 50 XVP.

Structure of BaTiO₃ thick-film piezoelectric ceramics cantilever

The thick film piezoelectric ceramic printed on alumina substrate is intended to be operated in the form of cantilever structure. The structure can provide a more

comfortable approach for mounting the device on the body of vibrational machinery for harvesting kinetic energy. The sandwiched cantilever structure is shown in Figure-3 (a) and (b), consisting of four layers which are substrate, Ag/Pd electrode for bottom layer, piezoelectric layers, and Ag/Pd electrode for top layer.

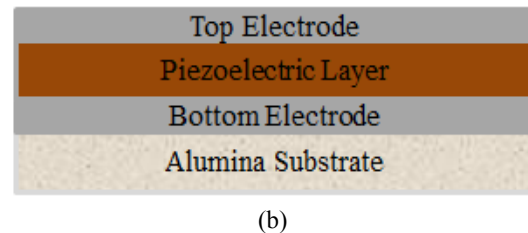
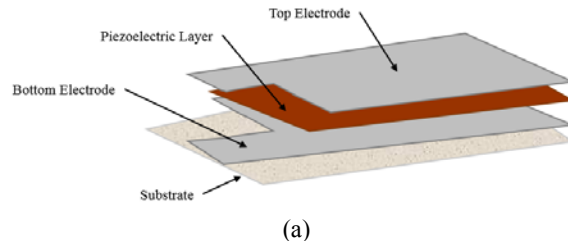


Figure-3. (a) Breakout diagram and (b) cross sectional view of the sandwiched BaTiO₃ thick-film piezoelectric ceramic.

Fabrication of BaTiO₃ thick-films

BaTiO₃ films were deposited on the underlying Ag/Pd bottom electrode. In this experiment, the screen printed electrode was specifically used Ag/Pd paste because to produce strong adhesions for BaTiO₃ films and high stresses from drying shrinkage and thermal expansion while firing. As a result of printed films, the Ag/Pd conductor also serves as glue for the BaTiO₃ thick film piezoelectric ceramic cantilever. The BaTiO₃ thick film piezoelectric was successfully fabricated by using screen printing techniques as shown in Figure-4.

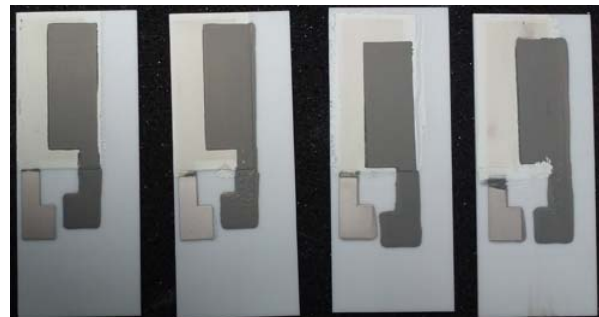


Figure-4. The BaTiO₃ thick film piezoelectric sandwiched between the top electrode and bottom electrode on alumina substrate.

Electrical properties

The fabricated BaTiO₃ thick-film piezoelectric cantilever was characterized both visually and electrically.



The electrical characterization is done by measuring the direct piezoelectric effect. The direct measurement method of BaTiO₃ thick-film piezoelectric ceramic cantilever were characterize with d_{33} meter using Berlincourt method as shown in Figure-5.

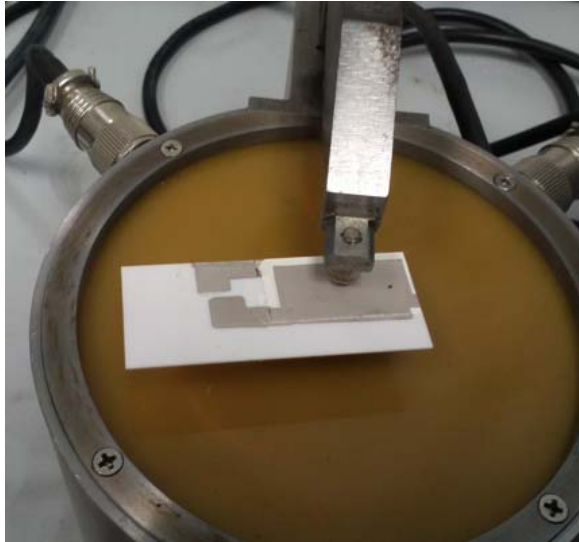


Figure-5. Direct measurement method of the BaTiO₃ thick-film piezoelectric ceramics.

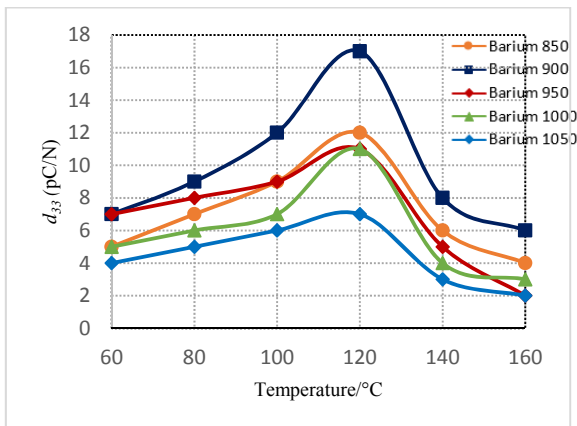


Figure-6. Polarization measurement of BaTiO₃ thick-film piezoelectric ceramic with different sample firing condition.

The compulsory process in the fabrication of BaTiO₃ thick-film piezoelectric ceramics is polarization. There are two important variables in this process, which are the applied electric fields and the Curing temperature. The electrical properties of the BaTiO₃ thick film piezoelectric ceramics samples were investigated on the piezoelectric charge coefficient and polarization behavior. The BaTiO₃ thick-film piezoelectric ceramics were polarized in direct contact with applied external electric fields at 20 kV/cm for 15 minutes poling duration. Too high external electric field could lead the piezoelectric in occurring short circuit. The applied external electric

fields depend on the thickness of the piezoelectric layers between top and bottom electrode. A thick the thickness of the piezoelectric film cantilever need high applied electric field compare to thin films. The response of BaTiO₃ thick-film piezoelectric ceramics were poled with different firing condition ranging temperature 60°C to 160°C were presented in Figure-7.

The measurement results of the highest charge coefficient piezoelectric properties (d_{33}) value was 17 pC/N at 120°C. On the other hand, all the BaTiO₃ thick films were reached at high value of d_{33} piezoelectric properties at 120°C as shown. It was reported that the piezoelectric ceramic was exhibit high value of d_{33} piezoelectric properties as it reaches or near to its Curie temperature (Ali *et al.* 2013) (Stojanovic *et al.* 2007). However, BaTiO₃ exhibit a relatively low Curie Temperature (T_c), which resulting from the tetragonal to paraelectric cubic phase transition at 120°C (Ali *et al.* 2013). From the polarization measurement, the highest peak response was BaTiO₃ with 900°C firing temperature, followed by second peak was BaTiO₃ with 850°C, the third and fourth share the same peak were BaTiO₃ with 950°C and 1000°C, respectively and lastly was BaTiO₃ with 1050°C firing temperature. In summary, the optimum firing condition for BaTiO₃ thick films is 900°C and the poling temperature is 120°C.

The result of the high piezoelectric properties characterization performed as function of temperature between 850°C to 1050°C were presented in Figure-10. Measurements were made with the different firing condition of BaTiO₃ thick film piezoelectric sample. The result shows it increasing to the highest value charge coefficient piezoelectric properties (d_{33}) was recorded at 900°C. The d_{33} values gradually decreasing at 950°C and further decreasing at 1050°C. There are many factors during preparation of thick-films piezoelectric ceramics could effect on the characterization of piezoelectric properties (Stojanovic *et al.* 2007). The observation can be made through BaTiO₃ thick film piezoelectric sample start to peel off at 1050°C but it still have a small d_{33} value piezoelectric was measured and there is no obvious crack. In order to explore more on firing condition, we try to increase the firing temperature to 1100°C for BaTiO₃ thick film piezoelectric sample. Unfortunately, the samples were totally defects and the Ag/Pd electrode peel off from the substrate. In the meantime, there is no d_{33} value was observed in the measured data. This evidence was expected due to the effect of different thermal treatment for the BaTiO₃ thick film piezoelectric ceramics samples. It have been reported by (Kok *et al.* n.d.) That the electrode was peeling off from the ceramic layer as stated.

In order to investigate the aging response in Figure-9, the BaTiO₃ films co-fired at 900°C were measured for three constitutive days. The measurements were taken for one hour with continuously applied dynamic force of 1N at 1 kHz. At the beginning of measurement it shows that the value of d_{33} which is due to the residual charges on the film and it reduces exponentially as the force is applied continuously and settle down at around 6 pC/N. From the graph it also



shows the aging response of the piezoelectric thick-film material which decay over time.

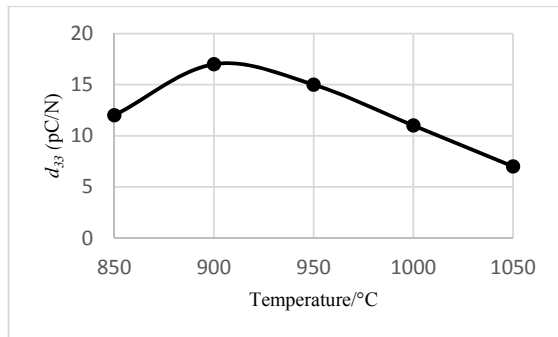


Figure-7. The measurement of charge coefficient properties (d_{33}) of BaTiO₃ thick-film with firing conditions.

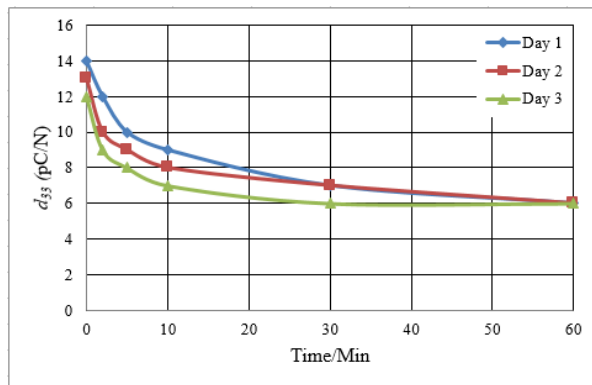


Figure-8. The aging response of BaTiO₃ thick-film piezoelectric ceramic with continuously one hour applied dynamic force for three days.

SEM Analysis

The cross-section of two layers screen-printed BaTiO₃ films is presented in Figure-9. The thickness of the two layers of BaTiO₃ films was approximately 140 μ m. The adhesion of the bottom and the top electrode layer over-printed were good and it could not be peeled off with tape. There is no interdiffusion were observed between Ag/Pd electrode and the BaTiO₃ thick films. The dense of the BaTiO₃ thick films and homogenous microstructure were observed.

The BaTiO₃ thick film piezoelectric prepared by screen printing method show after screen-printed good crystallinity and no evident presence of secondary phases in Figure-13. As it is seen from the results obtained by SEM analysis, the particle size is possible to observe. The highly textured microstructure of BaTiO₃ is visible pattern structure in high magnification shown in Figure-11. The grain size of the thick films seems to be uniform.

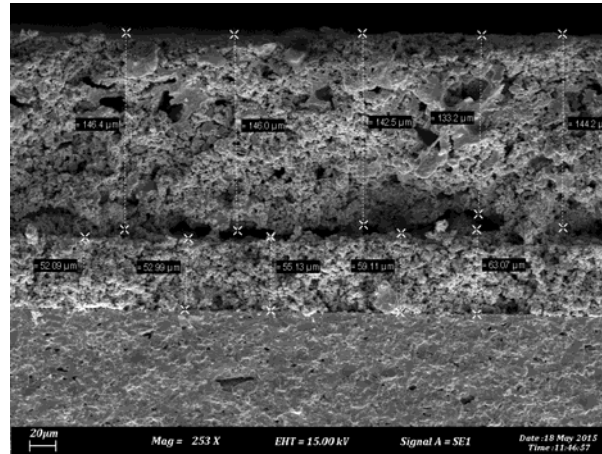


Figure-9. The cross section of the BaTiO₃ thick film piezoelectric sandwiched between the top electrode and bottom electrode on alumina substrate.

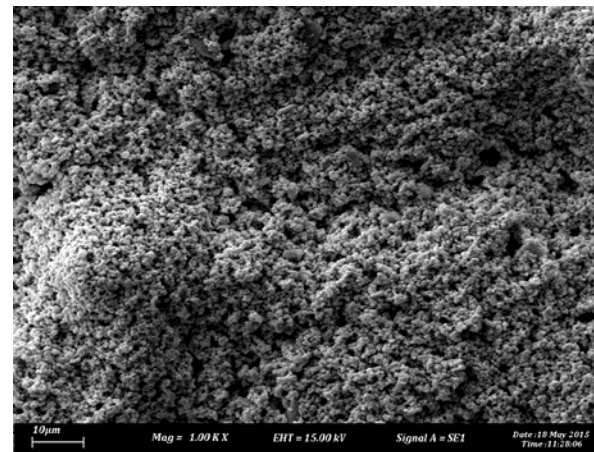


Figure-10. The microstructure of BaTiO₃ layers of the thick-film piezoelectric ceramic on the alumina substrate.

According to (Wolf and Trolier-McKinstry, 2004), temperature dependence on thickness and grain size of piezoelectric materials. Therefore, the Curie temperature of bulk piezoelectric materials and films material may not be identical. From the observation and experimentation, this BaTiO₃ thick film piezoelectric ceramics samples cannot be fired exceed than 1100°C because need to lower in order to obtain full densification of the films. The BaTiO₃ content of material decrease of density by observation made in SEM analysis which is containing little pores is presented in Figure-12. This behavior may be due to an oxygen vacancy because the samples were firing at high temperature (850°C-1050°C) and in an oxygen environment. The grain size of BaTiO₃ thick-films are less than 1 μ m, approximately 800-600 nm and are influenced by the milling process and annealing time that make the compounds good. This evidence were prove and realized by (Ha and Choi, 2014) that particle size control give effect in improvement of piezoelectric properties. Hence, the phenomenon of this



cause can be explained by relationship of printing force in order to obtain a dense structure.

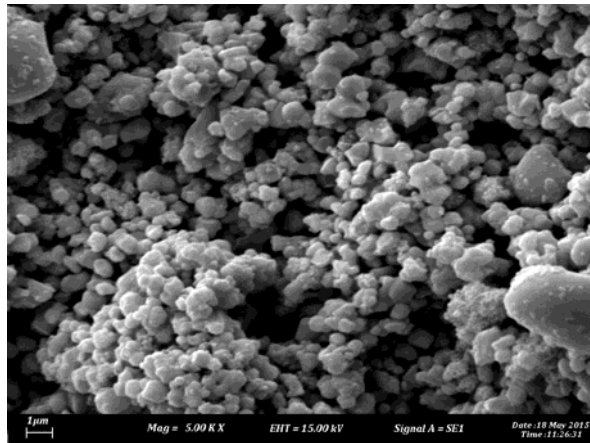


Figure-11. The high magnification of BaTiO₃ thick-film piezoelectric ceramics.

Another factor that can be made from the observation of SEM image was the homogeneity of the BaTiO₃ ceramic. The fact that the powders strong agglomeration is because of the thick-films were prepared from the mixture of barium titanate powders, binder and borosilicate glass and after that deposited on Al₂O₃ substrates. The ratio was the main part of making piezoelectric ceramics. In addition, the combination between the different particles sizes of the BaTiO₃ powders as shown in Table-1. Therefore, the small hole of particle can be filled by the small particle size (100 nm) make the homogeneous BaTiO₃ films. It is difficult to reach an optimal paste composition within reasonable time. Thus by combining with other process will help the increasing piezoelectric properties in the materials.

CONCLUSIONS

Screen printing is one of the simple techniques in fabricating thick-film piezoelectric ceramics. It also suitable be used for printing multilayers films, besides that it is relatively low-cost without involving any complicated process. From the experiment, the results demonstrated that the screen-printed lead-free BaTiO₃ thick-films are potential to be used for non-invasive applications of the sensor devices. The composition of BaTiO₃, borosilicate and temporary binder in the ratio of 5:2:1 has produced an optimum piezoelectric charge coefficient of 17 pC/N when co-fired at 900°C and poling temperature at 120°C.

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