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Barium titanate based dielectric sintered with a two-stage process

Chi-Shiung Hsi[a], Yi-Chun Chen[b,*], Heli Jantunen[c],
Min-Je Wu[a], Tzu-Chin Lin[a]

[a] *Department of Material Science and Engineering, National United University, Miao-Li 36003, Taiwan*
[b] *Department of Physics, National Cheng Kung University, Tainan, 70101, Taiwan*
[c] *Microelectronics and Materials Physics Laboratories and EMPART Research Group of Infotech Oulu,*
University of Oulu, P.O. Box 4500, FIN-90014 Oulu, Finland

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Abstract

A two-stage process was employed in this study to prepare dielectric material with high relative permittivity and low sintering temperature. In the first stage, the dielectric barium titanate ($BaTiO_3$) powders were uniformly coated with different amounts of glass via a high temperature melting process. The glass coated powders were then mixed with different amounts of LiF and sintered at temperatures from 850 to 950 °C using different dwell times in the second stage. The addition of LiF assisted the liquid phase formation of the glass, which lowered the sintering temperature and improved the densification of the material during the sintering process. Moreover, the pre-coated glass minimized the diffusion of LiF into the $BaTiO_3$ powders and thus dielectric properties of the sintered dielectrics were consistent with the samples without glass and LiF additions. Very dense sintered ceramics could be obtained using this two-stage process with a low sintering temperature, $\leq$900 °C. The relative permittivity achieved with 2 wt.% glass and 4 wt.% LiF additions sintered at 850 °C for 60 or 120 min was higher than 1700, which was only about 15% lower than pure $BaTiO_3$ sintered at 1450 °C for 4 h. The dielectric loss of the sintered dielectric was less than 0.01 being somewhat higher than with the pure $BaTiO_3$.

Keywords: Dielectric properties; $BaTiO_3$; Glass; Secondary phases; Sintering

1. Introduction

Barium titanate ($BaTiO_3$) is one of the most widely used dielectric materials. It has been utilized as primary material for multi-layer ceramic capacitors (MLCC), electro-optic devices and thermistors because of its good dielectric characteristics[1] In the conventional MLCC process, the green dielectric $BaTiO_3$ layer and silver–palladium electrodes are co-sintered at temperatures above 1000 °C.[2] These expensive electrode materials are always needed because of the high temperatures for dielectric sintering, and so the production cost of multi-layer ceramics is greatly increased. Sintering at lower temperatures with inexpensive electrodes is one of the most promising solutions. Numerous kinds of glasses have been used to introduce liquid phase into $BaTiO_3$ ceramic to lower the sintering temperature.[3–5] Alternatively, additives, such as lithium fluoride is also used as liquid phase generator for promoting the sintering of $BaTiO_3$ ceramic.[6,7]

However, when a large amount of glass is added to dielectric, it also decreases the relative permittivity since secondary phases are usually formed. The addition of Bi_2O_3–B_2O_3[8] and ZnO–B_2O_3–SiO_2[9] glass systems decreased the sintering temperature of $BaTiO_3$ to 850 °C, but at the same time the relative permittivity was reduced to below 1000. Additionally, BaO–B_2O_3–SiO_2 glasses were used to decrease the sintering temperature of Ba–Nd–Sm–Bi–Ti–O microwave dielectric.[10,11] Lee et al. found that $BaTi_4O_9$ dielectrics sintered at 1050 °C with B_2O_3 addition level of $\leq$5 wt.% maintained consistent relative permittivity values similar to the un-doped samples. By contrast, B_2O_3 addition larger than 5 wt.% greatly decreased the relative permittivity.[12]

In this research, both calcium borosilicate (CBS) glass flux and LiF were added to $BaTiO_3$ to accelerate the densification and lower the sintering temperature. To minimize the dielectric-

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[*] Corresponding author. Tel.: +886 6 2757575x65263; fax: +886 6 2747995.
E-mail address: ycchen93@mail.ncku.edu.tw (Y.-C. Chen).

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degradation effect from glass and LiF, a two-stage process was used to produce samples with high relative density and good electrical properties. In the first stage, a thin glass layer was pre-coated on the dielectric powder by a high temperature glass fusion process. In the second stage, the glass coated dielectric powders were mixed with different amounts of LiF and sintered at temperatures lower than 950 °C. The effects of the glass and LiF additions on the microstructure and electrical properties of the samples were studied.

2. Experimental procedure

A calcium borosilicate glass was prepared using reagent grade chemicals with a composition of 46.2 wt.% CaO, 27.4 wt.% B_2O_3, 20 wt.% SiO_2, 2.5 wt.% ZnO, 1.7 wt.% Al_2O_3, and 1.5 wt.% BaO. The chemicals were mixed and melted in a platinum crucible at 1500 °C for 2 h and quenched in water to form amorphous glass that was subsequently dried and crushed into a powder form. Some of the melt glass was also poured into a mold to make a glass bar to measure the temperatures of the glass transition and softening, which were found to be 585 and 650 °C, respectively. The crushed powder was ball milled in a PVC jar using yttrium stabilized zirconia balls. A commercial $BaTiO_3$ (BT) powder (BTP-HT, Prosperity Dielectric, Taiwan) with an average particle size of 1 μm was used as the dielectric material. The BT material had the relative permittivity and loss factor of 2029 and 0.011 at 1 kHz, respectively, when the powder was pressed and sintered at 1450 °C for 4 h.

The sintering of the BT dielectric at low temperatures was enabled by using a two-stage process. In the first stage, the commercial $BaTiO_3$ powder was mixed with different amounts of glass powder and heat-treated at 1050 °C for 30 min followed by the ball milling again in isopropanol (IPA) solution to an average size of 2 μm. Being heat-treated at first stage, the dielectric powders were supposed to be coated with a uniform layer of glass shell. The glass coated powders were then mixed with different amounts of LiF and dry pressed into 20 mm diameter pellets. In the second stage, the pressed pellets were finally sintered at temperatures of 850, 900, and 950 °C with different dwell times. For comparison, the BT dielectric was also prepared by the conventional one-stage process, at which the BT dielectric powder, glass powder, and LiF additive were mixed and sintered with various conditions.

The densities were determined with a He pycnometer (AccuPyc 1330, Micromeritics, U.S.A.) using the powder prepared by grinding the sintered compacts. Differential thermal (DTA, PerkinElmer, U.S.A.) and thermal-mechanical (TMA7, PerkinElmer, U.S.A.) analysis experiments were conducted using a heating rate of 10 °C/min. Surface and cross-section microstructures of the sintered samples were observed by

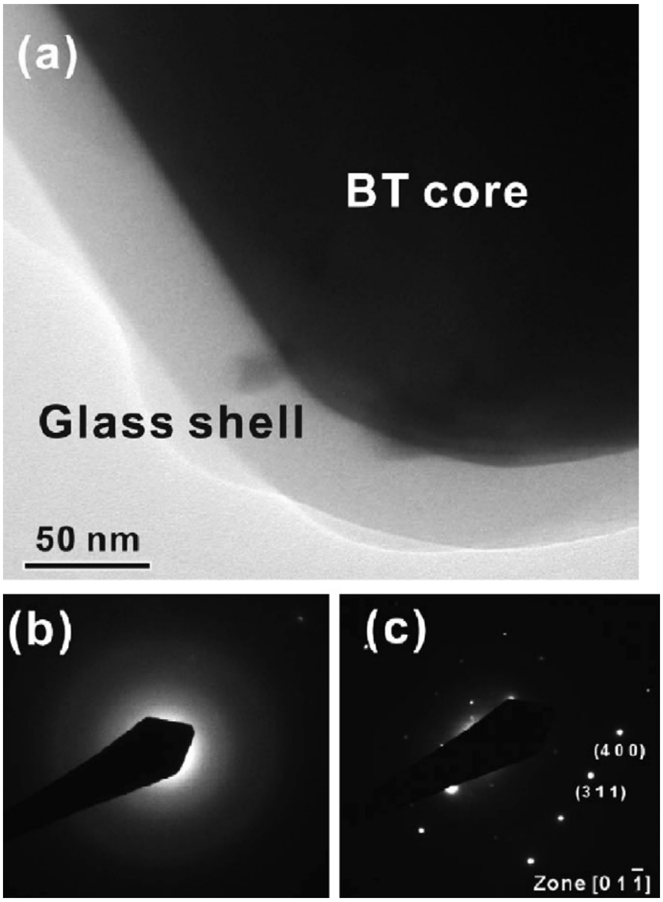

Fig. 1. Transmission electron microscopy (TEM) (a) bright field image of glass coated BT particle, (b) electron diffraction pattern of glass shell, and (c) electron diffraction pattern of the BT particle.

scanning electron microscopy (FE-SEM, JSM6700-F, JEOL, Japan). The sample compositions were examined using energy dispersive X-ray spectroscopy (EDS, Oxford-6587, England) equipped with SEM. The crystalline phases of the sintered dielectrics were determined by X-ray diffractometry (XRD, D/MAX-2200, Rigaku, Japan). The dielectric properties of the sintered compacts were measured using an LCR meter (Agilent E4980A) at 1 kHz. Local dielectric distributions with nano-scale resolution were investigated using an electrostatic force microscope (EFM, Digital Instruments CP-II system). During the non-contact surface scanning, an ac signal (20 kHz, 5 V_{rms}) was applied between the bottom of the sample and the PtIr coated scanning tip (force constant ~2.8 N/m). Under ac voltage, the first harmonic signal of the cantilever oscillation, which was related to the local capacitive properties, was measured through a lock-in amplifier.

3. Results and discussion

Fig. 1(a) shows the TEM (transmission electron microscopy) micrographic of glass coated BT dielectric powder after the first stage preparation process, the core particle was coated with a uniform glass shell with a thickness about 30 nm. The shell had an amorphous glass structure (Fig. 1(b)), while the core of glass coated powder showed $BaTiO_3$ crystal structure (Fig. 1(c)). Ti atom was not detected in the glass shell through EDS analysis. The results of TEM revealed there was few inter-diffusions between the BT core and glass shell occurring at the first stage, which can thus be treated as a pure glass coating process.

To further investigate the possible interaction between the CBS glass and BT powder during the glass coating process, a mixture of 50 wt.% CBS glass and 50 wt.% BT dielectric powder was analyzed using DTA. The onset temperature at 615 °C (Fig. 2) in the DTA analysis was the glass transition temperature of the glass powder. The measurements showed only one exothermic peak at a temperature of 725 °C. According to the X-ray diffraction analysis, a very low amount of $CaTiO_3$ phase formed in the samples when more than 10 wt.% glass was used to coat the BT dielectric powder at 1050 °C (pattern (b), of Fig. 3). When a larger amount of CBS was used (20 wt.%), the $CaTiO_3$ phase became more evident as shown in pattern (c) of Fig. 3. By contrast, there were no secondary phases found from the X-ray diffraction pattern of 5 wt.% glass coated BT powder (pattern (a), Fig. 3). Therefore, the exothermic peak at 725 °C in the DTA curve should correspond to the $CaTiO_3$ formation during the heat treatment formed by the interaction between the coating glass and the BT powder. The endothermic peak at 851 °C corresponds to the melting of residual glass when large glass addition (50 wt.%) was used. The glass started to melt at around 800 °C.

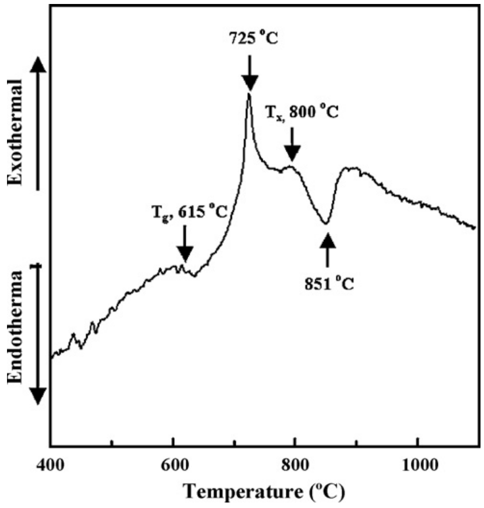

Fig. 2. DTA analysis of 50 wt.% CBS glass + 50 wt.% BT dielectric powder.

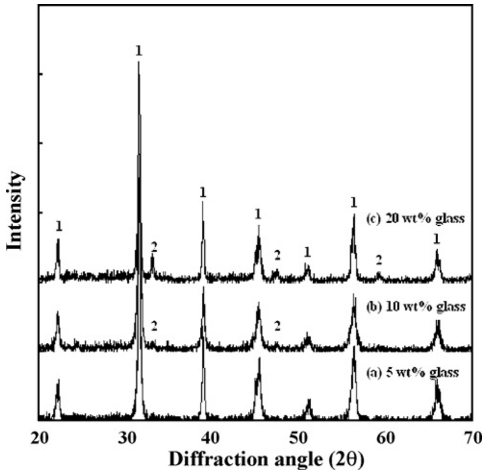

Fig. 3. X-ray diffraction patterns of the BT powder with (a) 5 wt.%, (b) 10 wt.%, and (c) 20 wt.% glass coating. 1: $BaTiO_3$ (74–1962), 2: $CaTiO_3$ (JCPDS 81–0562).

When the BT was mixed with 5 wt.% glass and fired at 1050 °C for 30 min, the melted glass covered the dielectric powders. The powder exhibited round shaped particles, adhering to each others with a thin layer of glass film, as shown in Fig. 4(a). During the high temperature glass coating process at 1050 °C, the glass constituents diffused into the dielectric powders. Fig. 4(b) shows a cross-section of 10 wt.% glass coated dielectric powder after treatment by HF acid etching. Calcia (CaO) content across the grain from A point to B point was in the range of 0.8–1.0 wt.%, while silicon oxide (SiO_2) content was in the range of 0.5–1.0 wt.% (Fig. 4(c)). There was no obvious calcium concentration gradient formed across the dielectric particles. This indicates that a core-shell structure was not evident in this heat treatment process. No secondary phases were found from the X-ray diffraction pattern of 5 wt.% glass coated dielectric powder. Grains with high calcium content (>15 wt.%), were found in the 10 wt.% glass coated powder, as indicated by the arrow in Fig. 4(d). The X-ray powder analysis shows that this might represent particles of $CaTiO_3$ phase.

The TMA result of a pure glass powder compact exhibits onset temperature at 620 °C, as shown by curve (a) in Fig. 5, corresponding to the glass softening temperature. There were two onset temperatures on the TMA curve for the 20 wt.% glass coated dielectric comp act, which suggests the existence of the second stage process. The first onset temperature was located at 570 °C and the second onset temperature at 785 °C, as shown in

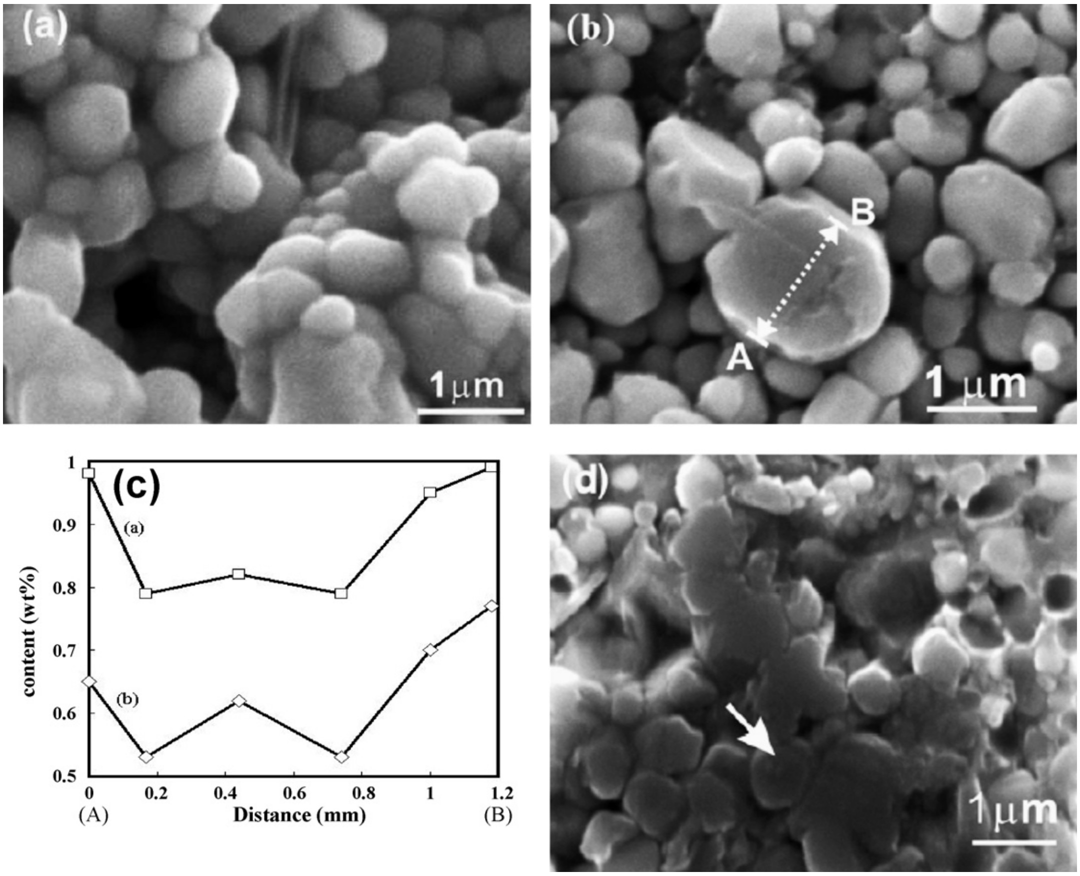

Fig. 4. SEM image showing the microstructure of the BT powder after heat treatment at 1050 °C with (a) 5 wt.% glass coating, (b) 10 wt.% glass coating and dilute HF acid etching, and (d) 10 wt.% glass coating. (c) CaO and SiO_2 content profile along line AB of (b).

curve (b) of Fig. 5. There was a plateau in the TMA curve (b) around 700 °C, which was due to formation of secondary phases between coating glass and $BaTiO_3$ dielectric, as indicated by the DTA and XRD results (Fig. 1 and Table 1). The first shrinkage related to the softening of coated glass and the second shrinkage related to the softening of residual coating glass.

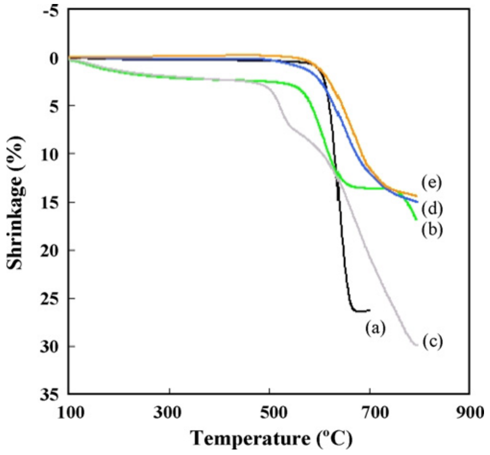

Fig. 5. Thermal-mechanical analysis of (a) CBS glass, (b) 20 wt.% glass + 80 wt.% BT, (c) 20 wt.% glass + 80 wt.% BT + 2 wt.% LiF, (d) 5 wt.% glass + 95 wt.% BT + 2 wt.% LiF, and (e) 2 wt.% glass + 98 wt.% BT + 2 wt.% LiF powder compacts.

When 2 wt.% LiF was added to the 20 wt.% glass coated BT, the TMA result shows lower onset temperature. The second shrinkage became obscure at 625 °C, as indicated by curve (c) in Fig. 5. In this sample, the added LiF lowered the softening temperatures of the coating and residual glass, and the first and second onset temperatures moved toward each other. The first and second onset temperature increased with decreasing content of the coating glass. When the coating glass decreased to 5 wt.% and 2 wt.% with 2 wt.% amount of LiF, the first onset temperature was at about 590 °C being only 20 °C lower than that of the compact without the LiF addition. The second onset temperatures of those samples were higher than 800 °C.

Fig. 6 shows the linear shrinkage in the one-stage and two-stage process of the BT compacts with different glass and LiF contents, where all the samples were sintered at 900 °C for 60 min. The flat dash line (curve (e), Fig. 6) at value of 17.6% was the reference linear shrinkage of $BaTiO_3$ powder doped with 2 wt.% LiF without glass. The two-stage process prepared samples had higher linear shrinkages than those prepared by the one-stage process. In the one-stage process, the dielectric powders were mixed with 2, 5, 10, and 20 wt.% glasses, and the shrinkage of the compact disks increased linearly with the glass content (curve (a), Fig. 6). The shrinkage with 10 wt.% coating glass addition in one-stage process was about 7.5%, and the sintered sample had very porous microstructure (Fig. 7(a)) similar to the microstructure of the powder after glass coat-

Table 1
Phase formation in the two-stage sintering of the BT samples with different glass coatings, LiF additions and sintering conditions

Glass content (wt.%)	LiF (wt.%)	Sintering		Phase formed
		Temp. (°C)	Time (min)	
5	0	850	60	$BaTiO_3$
5	4	850	60	$BaTiO_3$, $LiTiO_2$
5	0	950	60	$BaTiO_3$
5	4	950	60	$BaTiO_3$, $LiTiO_2$, Ca (TiO_3)
10	0	850	120	$BaTiO_3$, Ca (TiO_3), $Ba_2TiSi_2O_8$
10	4	850	120	$BaTiO_3$, $Ba_2TiSi_2O_8$
10	0	950	120	$BaTiO_3$, Ca (TiO_3), $Ba_2TiSi_2O_8$
10	4	950	120	$BaTiO_3$, $Ba_2TiSi_2O_8$
20	0	850	120	$BaTiO_3$, Ca (TiO_3), $Ba_2TiSi_2O_8$
20	4	850	120	$BaTiO_3$, Ca(TiO_3), $Ba_2TiSi_2O_8$
20	0	950	120	$BaTiO_3$, Ca (TiO_3), $Ba_2TiSi_2O_8$
20	4	950	120	$BaTiO_3$, Ca (TiO_3), $Ba_2TiSi_2O_8$

ing at 1050 °C (Fig. 4(a)). During the one-stage process, the glass melted and wetted the dielectric particles. The melt glass grouped the dielectric together and formed large glass/dielectric agglomerates due to the surface tension of the melting glass. A porous microstructure was thus produced.

When the dielectric powders were pre-coated with 2 and 5 wt.% glasses followed by sintered at 900 °C for 60 min in the two-stage process, the powder compact shrinkage was about 1.5% higher than those in the one-stage process (curve (b), Fig. 6). There were even more than 10% shrinkage difference between the one-stage and two-stage prepared powder compacted for more than 10 wt.% addition of glass. The microstructures of the samples with 10 wt.% addition of glass prepared by the two-stage process were also relatively dense (Fig. 7(b)). During the sintering stage in the two-stage process, the glass on the dielectric particles surface melted and formed viscous liquid over the particles, and thus the compacts were sintered via liquid phase improvements. For more than 10 wt.% CBS glass pre-coated, there were enough liquid phases covering the dielectric particles, and thus the linear shrinkage of the powder compacts became larger.

With 2 wt.% LiF additive and sintered by one-stage process at 900 °C for 60 min, the linear shrinkage of the 2, 5, 10, and 20 wt.% glass content samples were 15.3, 14.2, 18.3, and 21.2%, respectively (curve (c), Fig. 6). In low glass content samples

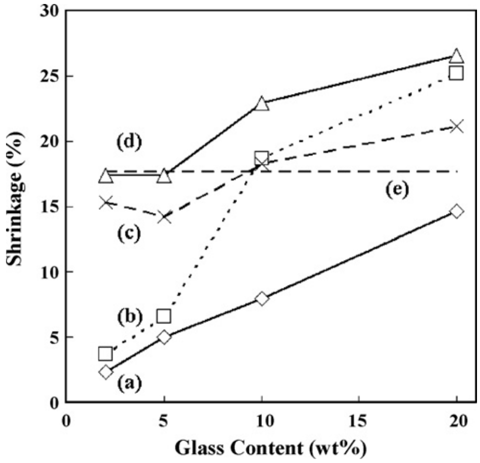

Fig. 6. Linear shrinkage in the one-stage and two-stage processes of the BT compacts with different glass and LiF additions (sintered at 900 °C for 60 min). (a) One-stage without LiF, (b) two-stage without LiF, (c) one-stage with 2 wt.% LiF, (d) two-stage with 2 wt.% LiF, and (e) BT + 2 wt.% LiF, without glass addition.

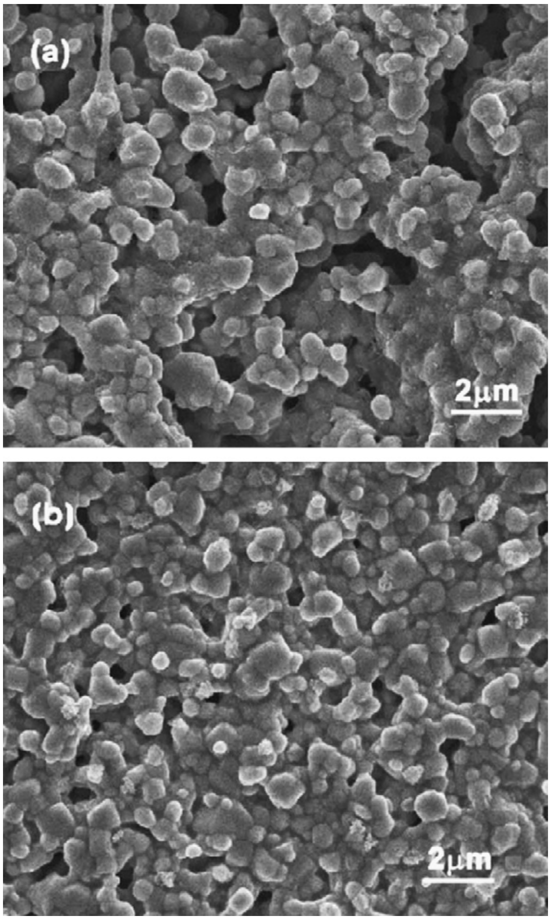

Fig. 7. Microstructure of the BT samples with 10 wt.% addition of coating glass after (a) the one-stage process and (b) two-stage process (sintered at 900 °C for 60 min).

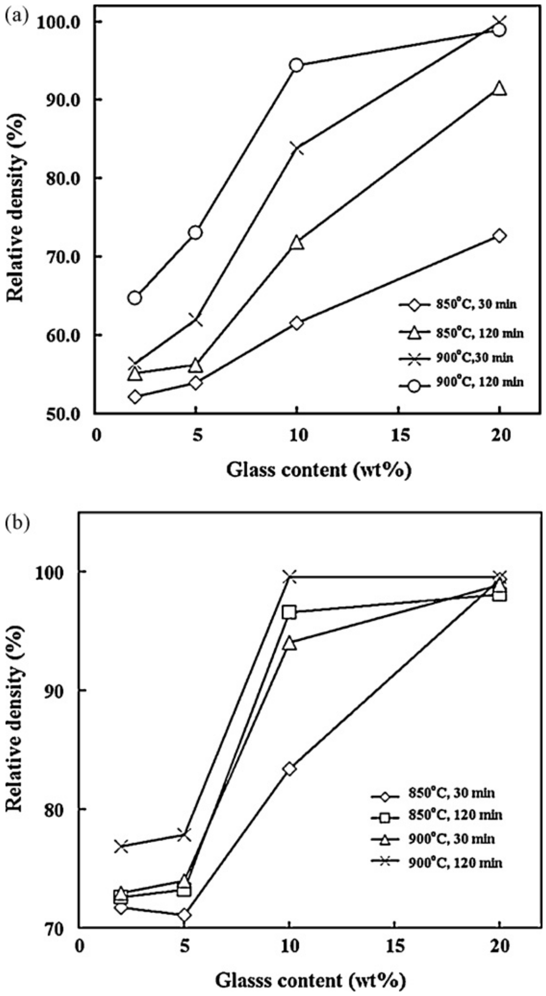

Fig. 8. Relative density of the samples after the two-stage as a function of glass coating addition (a) without (b) with 1 wt.% addition of LiF.

(<5 wt.%), the linear shrinkage difference between one-stage and two-stage process were smaller than 3.2%, while there were more than 9% linear shrinkage differences between the samples with and without LiF additions. Additive of LiF determined the linear shrinkage of the $BaTiO_3$ ceramics with low glass content during sintering. In the high glass content (10 and 20 wt.%), there were more than 10% differences between the linear shrinkage of the one-stage and two-stage prepared samples without LiF additive. With 2 wt.% LiF addition, samples prepared by two-stage process also had larger linear shrinkage than those prepared by one-stage process. Therefore, it suggested that the two-stage process enhanced the densification of BT dielectric ceramics during sintering.

Fig. 8(a) shows the relative density of the sintered samples prepared using different glass coated dielectrics after the two-stage process. The samples were sintered at temperatures of 850 and 900 °C for 30 and 120 min. The results show that the density of the sintered samples increased with the amount of glass coating, sintering temperature and time. When the sintered samples were prepared by 2 wt.% glass coated dielectric powder, their relative densities were lower than 70%. On the contrary, higher relative density (>90%) could be obtained when the samples were with 20 wt.% amount of glass and sintered at 900 °C. Fig. 8(b) shows that 1 wt.% addition of LiF to the glass pre-coated powders increased the relative density when sintered at temperatures of 850 and 900 °C for 30 and 120 min. The main reason is that LiF decreased the melting temperature and the viscosity of the glass. When the sintered disks were prepared using the glass coated dielectric powder with 4 wt.% LIF, the relative densities of sintered pellets were higher than 90% for the same sintering conditions.

Fig. 9 shows the microstructures of the BT dielectric compacts prepared by the two-stage process. Very porous

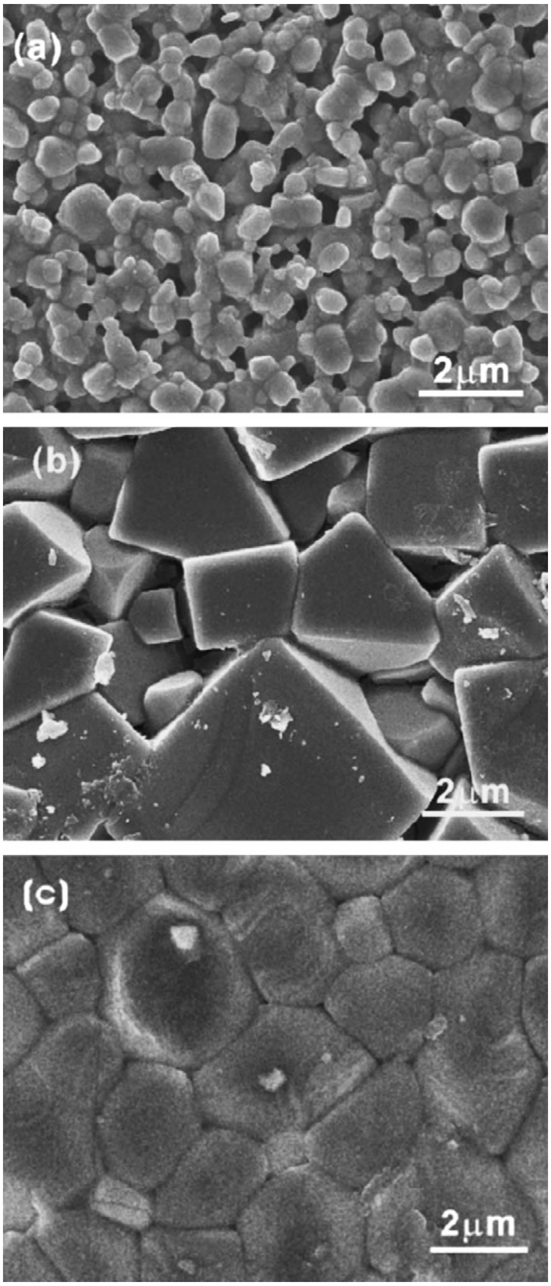

Fig. 9. Microstructures of the BT samples with (a) 2 wt.% addition of coating glass without LiF sintered at 850 °C for 30 min, (b) 2 wt.% addition of coating glass with 4 wt.% of LiF sintered at 850 °C for 30 min, and (c) 2 wt.% addition of coating glass with 4 wt.% of LiF sintered at 900 °C for 30 min.

microstructure (Fig. 9(a)) was observed for the sample of 2 wt.% glass coating BT dielectric powder without doping LiF and sintered at 850 °C for 30 min. The average grain size of the sintered dielectric was in the range of 0.5–1 µm. However, when larger amount of LiF was added (4 wt.%), the densification of the microstructure was improved (Fig. 9(b)). The average grain size in this case was in the range of 1–3 µm. Increasing the sintering temperature of this sample to 900 °C produced very dense microstructure without further increase of the grain size (Fig. 9(c)).

Table 1 lists the phases formed in the two-stage prepared samples with different amounts of glass coating, LiF additions and sintering conditions. When the samples were prepared using 5 wt.% glass coating, there were no secondary phases when sintered at 850 °C and 900 °C. With the 4 wt.% LiF addition and 5 wt.% glass coating, $LiTiO_2$ secondary phase was observed after sintered at 850 °C. With increasing sintering temperature up to 950 °C, $CaTiO_3$ phase was also formed. The samples with more than 10 wt.% glass coating and without LiF addition showed the formation of $CaTiO_3$ and $Ba_2TiSi_2O_8$. However, when 4 wt.% of LiF was added to these samples and sintered at 850 and 950 °C, $Ba_2TiSi_2O_8$ was the only secondary phase. When increasing the amount of coating glass (20 wt.%), on the other hand, two secondary phases, $CaTiO_3$ and $Ba_2TiSi_2O_8$, were shown again. It should be noted that when the samples with LiF addition of 4 wt.% and higher glass coating addition (>10 wt.%) were used, the $LiTiO_2$ phase disappeared and the $Ba_2TiSi_2O_8$ was formed instead. This result revealed that when the amount of glass addition was large enough, it was able to dissolve the Li^+ ions.

The samples made of the pure BT powder sintered at temperature of 1450 °C for 4 h had the relative permittivity and loss ($\tan\delta$) of 2029 and 0.011 at 1 kHz, respectively. Lin and Wu[7] reported that due to the formation of $BaLiF_3$ liquid phase, the BT dielectric doped with LiF can be sintered at temperatures lower than 950 °C. The samples with the LiF additions of 1 and 4 wt.% sintered at 900 °C for 60 min had the relative permittivity value of 4350, but the dielectric loss was higher than 0.037.

The relative permittivity and dielectric loss of the samples after the two-stage sintering were shown in Table 2. The samples with the 2 wt.% addition of the coating glass, 4 wt.% addition of LiF sintered at 850 °C for 60 min to 120 min had the relative permittivity value of about ∼1700 and the dielectric loss <0.01. The increasing amount of the coating glass decreased the permittivity and increased the loss. The lower permittivity of the samples with higher glass contents (>5 wt.%) was due to the formation of secondary phases, such as $Ba_2TiSi_2O_8$ and $Ca(TiO_3)$, with poor dielectric properties. The LiF addition to the glass coated powders promoted the densification of the microstructure and thus generally increased the relative permittivity. This addition also decreased the loss value except when very large amount of coating glass (20 wt.%) was added and high sintering temperature (900 °C) was used. As shown by the XRD analysis, the glass phase crystallizes during heat treatment, which might be the reason for the achieved lower dielectric loss.

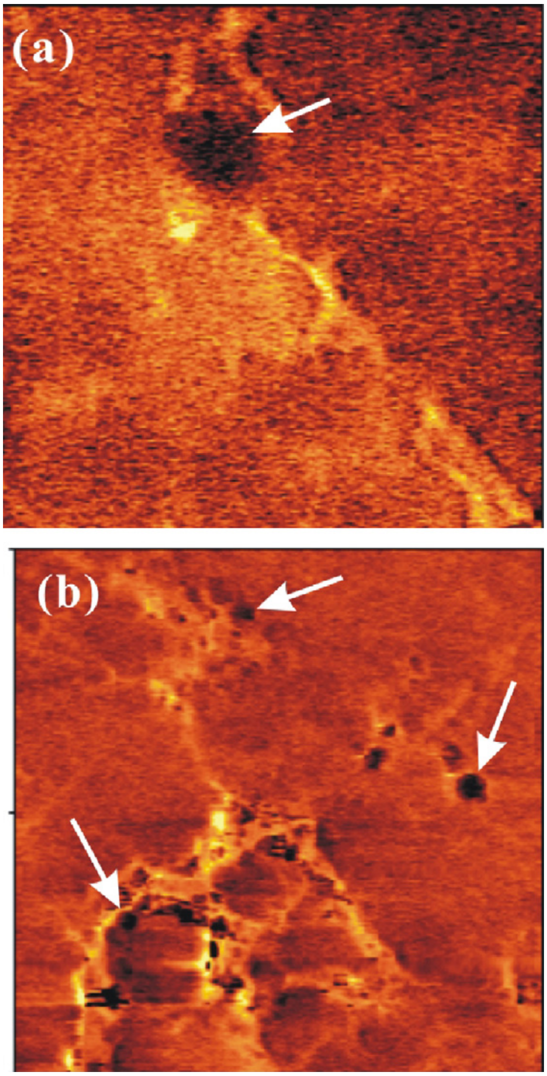

Fig. 10. EFM images of the BT samples prepared using 10 wt.% glass content and (a) 2 wt.% and (b) 4 wt.% addition of LiF. The arrows indicate the regions with low relative permittivity (scanning size: 10 µm × 10 µm).

Fig. 10 shows the EFM mappings of the 10 wt.% glass–$BaTiO_3$ samples with 2 wt.% and 4 wt.% addition of LiF. When an ac voltage ($V_{ac}\cos\omega t$) was applied, the electric force (F_e) between the sample and the scanning tip is created as shown in the following equation:

$$F_e = \frac{1}{4}\frac{dC}{dz}V_{ac}^2 + \frac{\sigma_b}{2\varepsilon_0}CV_{ac}\cos\omega t + \frac{1}{4}\frac{dC}{dz}V_{ac}^2\cos 2\omega t \qquad (1)$$

where C is the tip-to-sample capacitance, z is the tip-to-sample distance, ε_0 is the vacuum permittivity, and σ_b is the bound charge due to electric polarization. Fig. 10 shows the distribution for the amplitude of first harmonic term ($\cos\omega t$), which is proportional to σ_b and C, both related to the local dielectric constant. The EFM images reveal that in the 2 wt.%-LiF sample, the secondary phases, i.e. the low relative permittivity regions, were gathered together, while in the 4 wt.%-LiF sample, the secondary phases were expelled and arranged along the grain boundaries. The low-constant secondary phase in the EFM images confirms the results of X-ray diffraction patterns. Since

Table 2
The relative permittivity and dielectric loss of the BT samples at 1 kHz after the two-stage sintering with different glass coatings, LiF additions and sintering conditions

Sintering temp. (°C)	Glass content (wt.%)	LiF (wt.%)	Dwelling time					
			30 min		60 min		120 min	
			ε_r	tan δ	ε_r	tan δ	ε_r	tan δ
850	2	0	117	0.588	133	0.570	152	0.625
850	2	4	1875	0.154	1725	0.008	1775	0.009
850	5	0	577	0.714	503	0.278	548	0.500
850	5	4	1055	0.012	1205	0.012	1162	0.015
850	10	0	503	0.833	523	0.179	609	0.333
850	10	4	740	0.013	825	0.013	843	0.013
850	20	0	172	0.023	219	0.132	305	0.034
850	20	4	289	0.016	366	0.011	404	0.010
900	2	0	708	0.769	640	0.200	1592	0.526
900	2	4	1628	0.118	2228	0.185	2292	0.222
900	5	0	901	0.476	552	0.500	683	0.588
900	5	4	1051	0.016	1128	0.014	1154	0.014
900	10	0	778	0.200	753	0.256	762	0.164
900	10	4	827	0.014	847	0.014	844	0.014
900	20	0	200	0.005	193	0.004	195	0.005
900	20	4	396	0.011	484	0.012	446	0.011

the distribution of the low-constant phases were mainly in the grain boundaries, the impedance of the electric polarization in the normal grains was minimized, and thus resulted in relative good macroscopic dielectric properties than normal glass- or LiF- additive dielectrics.

4. Conclusions

In this research, a two-stage process to enable sintering BT samples at low temperatures is presented. In the first stage of the process, dielectric powders were pre-coated by glass using a high temperature fusion via melting the glass. Uniform, good-adherence glass layers were thus formed over the powders. In the second stage, LiF was added to assist the glass softening. Dense microstructures (relative density >90%) could be obtained by sintering at low temperature (<900 °C) for short dwell time (<60 min). Second phases of $LiTiO_2$, $CaTiO_3$, and $Ba_2TiSi_2O_8$ were found from the glass and LiF doped dielectrics sintered at temperature higher than 850 °C. All samples containing 2 wt.% of coating glass and 4 wt.% of LiF showed higher relative permittivity >1700 and low loss value ∼0.01 at 1 kHz. The good dielectric performance achieved with the two-stage process was attributed to the high relative density and decreasing the glass content of the sintered dielectric.

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