

Synthesis and Characterization of Particle-filled Glass/Glass-Ceramic Composites for Microelectronic Packaging (I)

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Abstract : For microelectronic packaging application, the crystallizable glass powder in $\text{CaO-Al}_2\text{O}_3\text{-SiO}_2\text{-B}_2\text{O}_3$ system was mixed with various amounts of alumina inclusions ($\approx 4\mu\text{m}$), and its sintering behavior, crystallization behavior, and dielectric constant were examined in terms of vol% of alumina and the reaction between the alumina and the glass. Sintering of the CASB glass powder alone at 900°C resulted in full densification (99.5%). Sintering of alumina-filled composite at 900°C also resulted in a substantial densification higher than 97% of theoretical density. In this case, the maximum volume percent of alumina should be less than 40%. XRD analysis revealed that there was a partial dissolution of alumina into the glass. This alumina dissolution, however, did not show the particle growth and shape accommodation. Therefore, the sintering of both the pure glass and the alumina-filled composite was mainly achieved by the viscous flow and the redistribution of the glass. Alumina dissolution accelerated the crystallization initiation time at 1000°C and hindered the densification of the glass. Dielectric constants of both the alumina-filled glass and the glass-ceramic composites were increased with increasing alumina content and followed rule of mixture. In case of the glass-ceramic matrix composites showed relatively lower dielectric constant than the glass matrix composite. Furthermore, as alumina content increased, crystallization behavior of the glass was changed due to the reaction between the glass and the alumina. As alumina reacted with the glass matrix, the major crystallized phase was shifted from wollastonite to gehlenite. In this system, alumina dissolution strongly depended on the particle size: When the particle size of alumina was increased to $15\mu\text{m}$, no sign of dissolution was observed and the major crystallized phase was wollastonite.

1. Introduction

The recent trend in electronic devices is toward small, light weight, multi-function, high-performance, and lower cost products. To meet these requirements, large-scale integration(LSI) circuit chips have become more compact and operated at higher speed. Therefore, multilayered alumina used conventionally for substrate material is no longer feasible because alumina has a high dielectric constant thus signal delay is high. Also, due to higher processing temperatures of alumina (greater than 1550°C), the refractory metals such as W and Mo are used for the metallization which give relatively poor electrical conductivity.

Therefore, low dielectric constant and low temperature sintering materials have been de-

veloped for use with high conductive metals (e. g. Ag, Au, and Cu) in order to render higher signal propagation speed and conductivity. Three types of low temperature cofired ceramics(LTCC) which are compatible with gold, silver and copper, have been extensively developed; These materials are (a) ceramic inclusions incorporated glass or glass-ceramic composites,¹⁻⁸⁾ (b) crystallizable glasses^{9,10)}, and (c) crystalline ceramics.¹¹⁻¹²⁾

Among the above categories, ceramic inclusion-filled glass or glass-ceramic composites can offer considerable improvement in various properties such as dielectric constant, coefficient of thermal expansion, and mechanical strength compared to single phase ceramics. The potential for improved properties is, however, offset by the increased difficulties in

forming composites with required high density and controlled microstructure by conventional, pressureless sintering. According to theoretical predictions of the rule of mixture, densification rate of a composite is linearly decreased with increasing the volume fraction of the inclusions.¹³⁾ Experimental data indicates, however, the drastic deviation from the predictions of the rule of mixtures at a certain critical inclusion volume fraction. According to De Jonghe et. al. 's observation¹⁴⁾, the critical inclusion volume fraction is ~ 0.2 for the soda-lime glass matrix and ~ 0.05 for the ZnO polycrystalline matrix. The explanations put forward for the drastic decrement in sinterability of the composite include (a) high viscoelastic backstresses due to mismatch in the shrinkage rates between the matrix and the inclusions,¹⁵⁻¹⁶⁾ (b) the formation of a rigid, continuous network of touching inclusion particles,¹⁷⁾ and (c) packing inhomogeneity of the matrix due to the presence of the inclusion.¹⁸⁾

In glass or glass-ceramic composites, the situation becomes more complicate if there is a reaction between matrix and inclusions. A reaction between the matrix and the inclusion changes the composition and the viscosity of the matrix, and affects the densification behavior, crystallization behavior and consequently the final properties of the composite. This reaction may be either beneficial or unfavorable to a composite system for substrate application. In present study, a crystallizable glass composition was selected in CaO-Al₂O₃-SiO₂-B₂O₃ system as the matrix. Alumina powders having different average particle sizes were chosen as the rigid inclusions. Research works were carried out to provide an assessment of how the reaction between the matrix and the inclusion affects the crystallization kinetics, densification behavior

and dielectric constant of the composite.

2. Experimental Procedure

A crystallizable glass composition (designated as CASB) was selected in the CaO-Al₂O₃-SiO₂-B₂O₃ system as the matrix. CaO-Al₂O₃-SiO₂-B₂O₃ system was frequently used in developing the substrate material.¹⁹⁻²¹⁾ However, the compositions used by other researchers were in wollastonite-anorthite-silica compatibility triangle. In present study, however, the selected CASB glass composition as listed in Table 1 is located in the wollastonite-anorthite-gehlenite compatibility triangle. B₂O₃ was added for controlling the

Table 1. Composition of As-prepared CASB Glass (in wt%)

CaO	Al ₂ O ₃	SiO ₂	B ₂ O ₃
33.0	17.0	40.0	10.0

viscosity of the glass. The addition of B₂O₃ did not form any borate phases with CaO, Al₂O₃ and SiO₂ during the crystallization. The CASB glass was produced by melting the constituents in a platinum crucible at 1620°C for 2 h, followed by quenching into water. The obtained glass fragments were further ground by using an alumina pestle and mortar, and finally milled for 24 h by using zirconia balls as the grinding media and reagent 2-propyl alcohol. After milling, the average particle size of the glass powder was $\sim 6.49\mu\text{m}$.

Two kinds of alumina having different particle size were selected as the rigid inclusions. A fine grained alumina, average particle size $\approx 4\mu\text{m}$, was obtained from Sumitomo(AM-21). An alumina having larger average particle size $\approx 15\mu\text{m}$ was obtained from High Purity Chemical, Co. Purity of both aluminas is above 99%.

The CASB glass powder and alumina inclusions were mixed with various volume fraction ratios and the mixed powder was uniaxially pressed at 75 MPa to make pellets in 15 mm in diameter and 5 mm in thickness. Samples were isothermally sintered at 900°C and 1000°C with various times in a horizontal tube furnace. They were pushed into the heating zone after binder burn out. A period of 1 min was allowed for the samples to equilibrate at the sintering temperature. The sintered density was measured by using Archimedes method. The theoretical densities, 2.68g/cm³ for the CASB glass and 3.98g/cm³ for alumina were used to estimate the theoretical densities of the composites by using the rule of mixtures. The reported densities are the average of three samples. The densification results were qualitatively confirmed by microstructural observations by using a SEM. Sintering at 1000°C converted the glass to the glass-ceramic form. The precipitated crystal phases during sintering was analyzed by using a XRD. The phase distribution change due to the reaction between the alumina inclusions and the glass powder was also monitored by

XRD phase analysis. The dielectric constants of both glass and glass-ceramic matrix composites containing various vol % of alumina inclusions were measured by using a LC impedance meter.

3. Results and Discussion

3.1. Pure CASB glass powder

One of the main reasons adapting glasses and glass-ceramics as the substrate material is their low sintering temperature, thus they are cofirable with high conductive metals such as Ag, Au, and Cu. However, if the densification temperature of the glass powder is too low, then it is difficult to remove all solvents, binders, and plasticizers, completely. Any residual carbon that may form during binder decomposition, if trapped in the substrate material (higher than 300 ppm), would adversely increase the dielectric constant.²²⁾ Therefore, sintering temperature of the glass must be high enough to complete burn-off the organic additives and also low enough to cofire (<1050°C) with Cu.

Table 2 shows measured basic physical prop-

Table 2. The Basic Properties of As-prepared CASB glass

Glass Transition Temperature	762°C
Dilatometric Deformation Temperature	819°C
Crystallization Temperature	1st Peak: onset temp. = 999°C max. temp = 1044°C 2nd Peak: onset temp. = 1072°C max. temp. = 1088°C
Onset Shrinkage Temperature	765°C

erties of the as-prepared CASB glass. The dilatometric onset shrinkage temperature of the glass compact is 765°C which is corresponding to its glass transition point. However, the isothermal sintering of the glass

powder at 765°C exhibited poor densification and revealed that higher temperature is required to obtain a substantial densification. The glass powder compacts were isothermally sintered at the temperature range of

800–1000°C, and change of the relative density as a function of time is shown in Fig. 1. When firing was conducted at 800°C, slightly above the onset shrinkage temperature, the density change of the glass was negligible during 40 min soaking. This result indicates that complete binder burn-off is possible in this

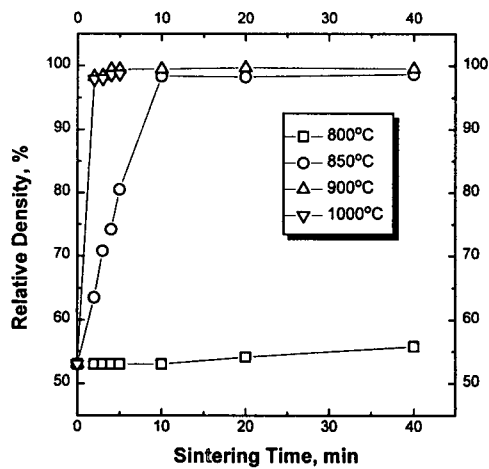


Fig. 1. Relative density as a function of time at temperatures from 800°C to 1000°C.

glass system. Binders used in tape casting process of glass material normally decompose completely at about 500°C. According to Tummala,²²⁾ the removal of the last traces of binder, however, extends to 800°C when the binder is absorbed on the glass surface. After 40 min at 800°C, the density of the CASB glass powder is slightly higher than the green density of the as-pressed glass compact. Therefore, glass compact still has lots of open tunnels for binder removal. Isothermal sintering at 850°C showed that the glass densified continuously. Glass powder reached 98% of theoretical density after 10 min soaking and its density was maintained as the sintering continued. At 900°C, the glass compact instantly reached a density value close to its theoretical density (99.5%) after 4 min soaking. Sintering at 1000°C produced a slightly

decreased densification due to bloating. Also after 5 min soaking at 1000°C, crystallization occurred.

From this isothermal densification study, it is found that the synthesis of the alumina-filled CASB glass composite should conduct at 900°C in order to fully utilize the viscous flow of the glass. Subsequent heat treatment above 1000°C, the glass converts to the glass-ceramic form, hence the alumina-filled glass-ceramic composite can be produced. According to DTA result listed in table 2, the CASB glass exhibits two exothermic peaks related to crystallization. Therefore, glass-ceramic having various crystalline phase may be produced by controlling crystallization temperature. In present study, however, crystallization heat treatment higher than 1000°C did not performed because heat treatment higher than 1000°C is not suitable for cofiring with Cu.

3.2. Alumina-filled glass/ glass-ceramic composites

In densification of a glass/glass-ceramic +rigid ceramic inclusions mixture, the viscous flow and the redistribution of the glass must be fully utilized. However, the densification via the viscous flow and the redistribution of the glass is inhibited by the incorporated inclusions as mentioned earlier in introduction. Experimentally observed that the threshold volume fraction of the inclusions triggering the drastic reduction in the densification rate in the glass matrix composite is about 0.2.¹⁴⁾ This volume fraction is, however, only applicable to a certain system. Therefore, this critical volume fraction is questionable if the system is changed. Therefore, the maximum volume fraction of the inclusions giving a required density must be verified in this alumina +CASB system.

Fig. 2 shows the isothermal densification result of the CASB glass+alumina ($\approx 4\mu\text{m}$) mixture at 900°C with that of the pure CASB glass. Up to 20% alumina additions, the effect of inclusions on the densification was negligible. The densification took place rapidly in the first 3 min, then slow down, and eventually stopped at a constant relative density. After 40 min, the relative density of the composite containing 10% alumina is slightly

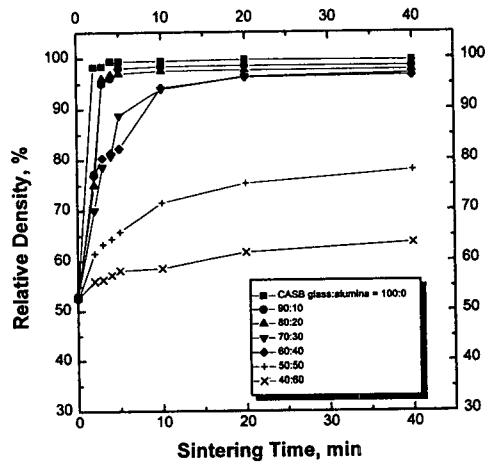


Fig. 2. Relative density as a function of time at 900°C for the samples with various alumina contents.

higher (98.5%) than that of 20% alumina (97.7%) incorporated composite. On the other hand, in case of the composites containing 30-40 vol% of alumina, the densification took place gradually and reached 97% and 96.5% of theoretical density, respectively after 40 min soaking. The composite containing higher than 50 vol% alumina, the densification rate was drastically reduced and the final relative density was below to 78%. This drastic reduction in densification probably resulted from the development of tensile stress which oppose to compressive sintering stress¹⁵⁻¹⁶⁾ and the percolation of the inclusions.¹⁷⁾ Therefore, in case of the CASB glass+alumina composite, less than 40 vol% of alumina incorpor-

ation is required to get a substantial densification ($>97\%$). This maximum vol% of inclusions is higher than that of De Jonghe et. al.'s finding.¹⁴⁾ Fig. 3 and Fig. 4 show XRD analysis and microstructure observation

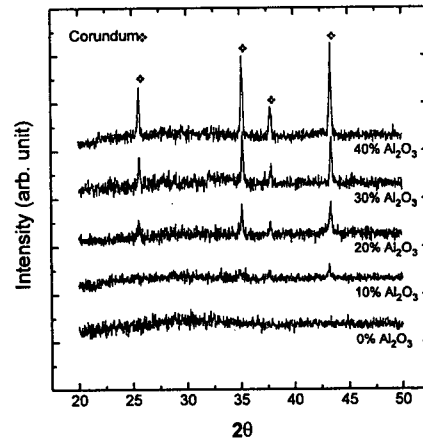


Fig. 3. XRD patterns of the alumina-filled glass composites after sintered at 900°C for 40 min. The average particle size of alumina = $4\mu\text{m}$.

results of the alumina-filled glass composite containing different vol% alumina after sintered at 900°C for 40 min, respectively. As it can be seen, there was no sign of crystallization throughout the matrix. The composites containing less than 40% alumina inclusions shows almost fully densified microstructures. Furthermore, individual alumina particle is surrounded by the glass network. This microstructural observation implies a good redistribution and wetting of the glass on the alumina. The composite containing 50% alumina, however, did not sintered well, resulting in poor density.

Heating the sample at 1000°C resulted in multiphase crystallization throughout the glass. The theoretical density of the crystallized sample was not able to determine because the exact volume fraction of each precipitated crystalline phase was hardly de-

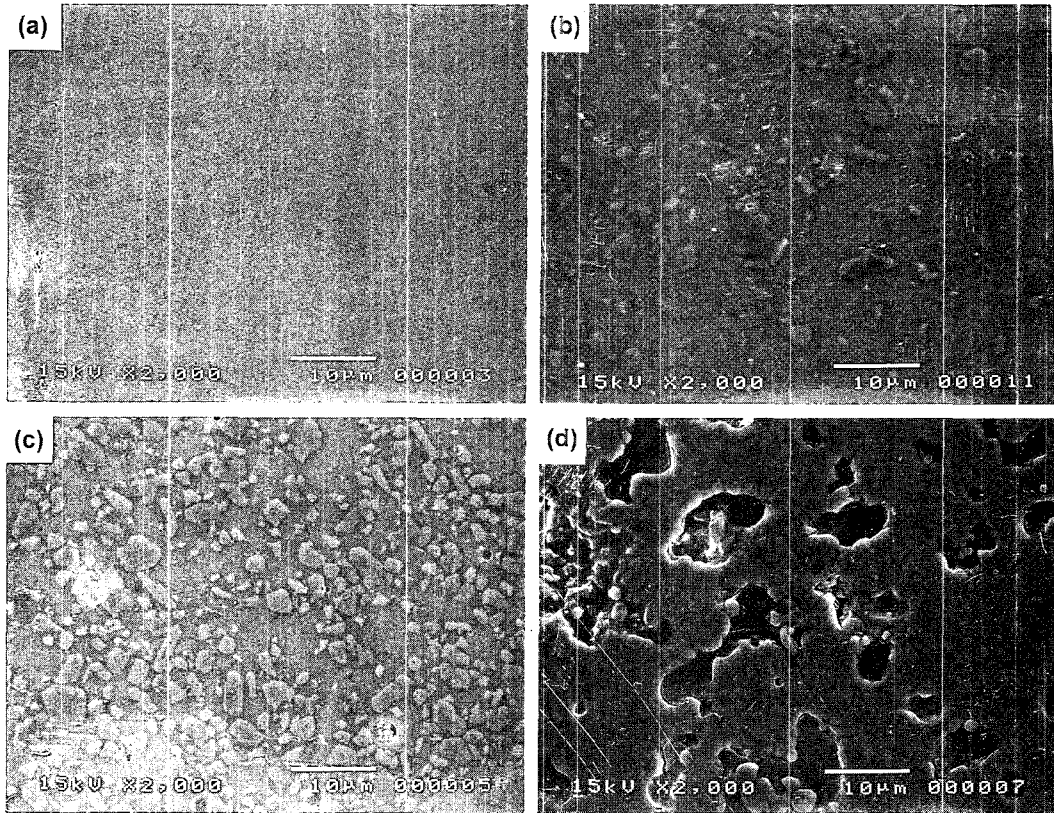


Fig. 4. Microstructures for the sample with (a) 0 vol.%, (b) 30 vol.%, (c) 40 vol.%, and (d) 50 vol.% alumina inclusions, fired at 900°C for 40 min.

terminated. Therefore, at a given volume percent of alumina inclusions, percent of linear shrinkage as a function of time is present in

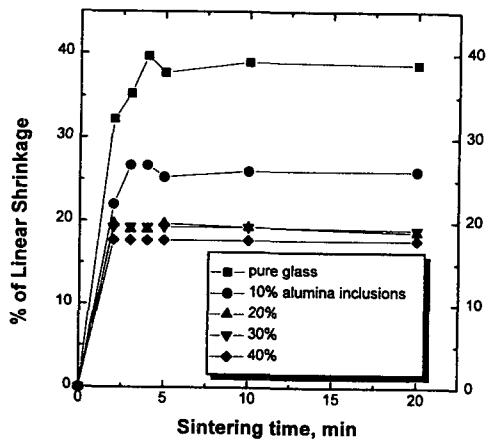


Fig. 5. Percent linear shrinkage as a function of time at 1000°C for the samples with various alumina inclusion contents.

In Fig. 6 and Fig. 7, XRD phase analysis results of the pure CASB glass and 30% alumina incorporated composite sintered at

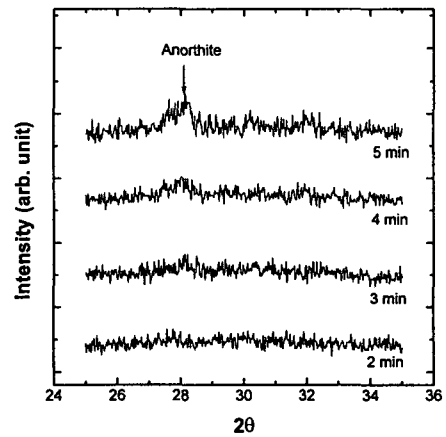


Fig. 6. XRD patterns of CASB glass as a function of sintering time at 1000°C.

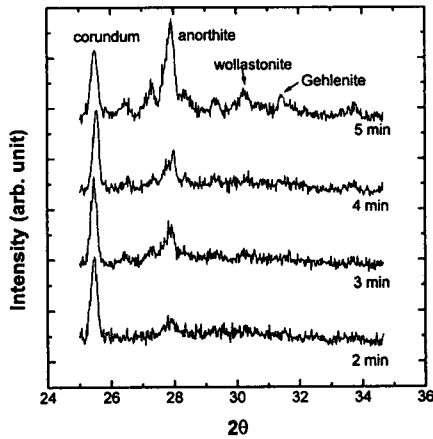


Fig. 7. XRD patterns of 70% CASB glass-30% alumina mixture as a function of sintering time at 1000°C.

1000°C as a function of time are shown. Fig. 5 through 7 explicitly show the sintering process for alumina filled CASB glass-ceramic composite with a 20 min hold time. The pure CASB glass powder exhibited a maximum shrinkage after 4 min soaking at 1000°C. According to DTA results, 1000°C is the onset temperature for crystallization. XRD analysis result (see Fig. 6), however, shows that heat treating at 1000°C for 4 min did not give any substantial crystallization. This is due to the crystallization kinetics, and indicating that there is an incubation time for crystallization at 1000°C. Therefore, first 4 minutes at 1000°C, instead of crystallization, viscous flow of the glass powder was dominant and this resulted in maximum densification. After 5 min soaking, the specimen was slightly expanded. This is probably due to bloating of pores and crystallization of anorthite ($\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$). Anorthite has a lower density ($\rho=2.763\text{g}/\text{cm}^3$) than the glass matrix ($\rho=2.8108\text{g}/\text{cm}^3$), therefore a slight volume expansion during anorthite precipitation is possible. As the sintering prolonged, wollastonite ($\text{CaO} \cdot \text{SiO}_2$, $\rho=2.92\text{g}/\text{cm}^3$) and gehlenite ($2\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot \text{SiO}_2$,

$\rho=3.048\text{g}/\text{cm}^3$) phases were precipitated (see Fig. 10). At this time, the dimensional change of the sample was not detected because the sample already had become a rigid form.

At a glance of Fig. 5, the final firing shrinkage was drastically reduced as the vol % of alumina increased. For example, the shrinkage of the 20% alumina filled composite is almost half of the shrinkage of the glass alone. This shrinkage result, however, does not indicate that the densification of the 20% alumina-filled composite was progressed only half of that of the pure glass. The incorporated rigid alumina inclusions do not shrink during sintering but only glass powder does. As vol % of the rigid inclusions increases, the total area to be shrunken is decreased. Therefore, total shrinkages are not same even though their firing density are same. Microstructural observation supports this fact. Fig. 8 shows microstructures of the composites containing different vol. % of alumina inclusions sintered at 1000°C for 20 min. As you can see, there is no evident of drastic densification difference. However, comparing with the microstructures shown in Fig. 4, some large pores left when the composites were isothermally sintered at 1000°C. This result shows some factors exist to hinder the densification at 1000°C. As you can see in Figs. 6 and 7, the initiation of crystallization at 1000°C became faster when alumina added. This fast crystallization prevents the viscous flow and reduces densification. Therefore, two step heat treatment is required for processing a dense alumina-filled glass-ceramic composite. First heat treatment is for densification of the glass powder at 900°C, and the subsequent heat treatment at 1000°C is for crystallization of the glass matrix.

Fig. 9 shows the dielectric constants of CASB+alumina composites measured at 1

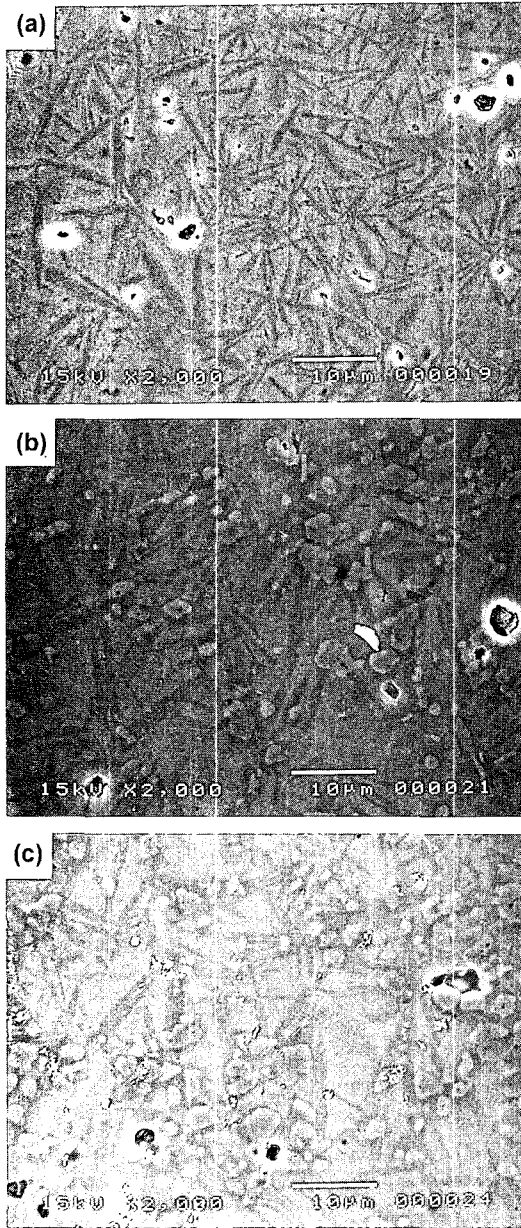


Fig. 8. Microstructures for the sample with (a) 0 vol.%, (b) 20 vol.%, and (c) 30 vol.%, alumina inclusions, fired at 1000°C for 20 min.

MHz. The glass-ceramic matrix composites exhibited lower dielectric constant than glass matrix composites due to the precipitation of crystalline phases having low dielectric constants such as wollastonite ($\epsilon=7\sim 8$) and anorthite ($\epsilon=6$).^{20,23} The dielectric constants of

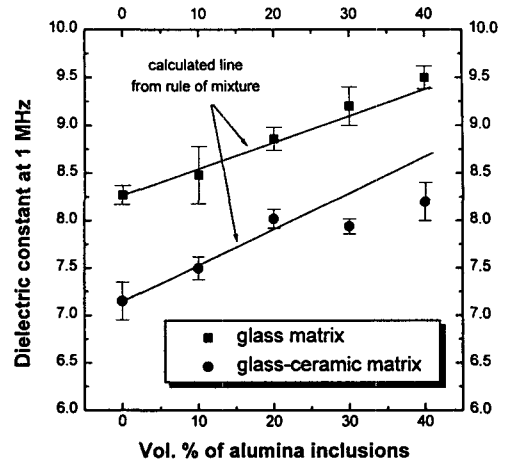


Fig. 9. Dielectric constant of alumina-filled CASB glass-ceramic composite as a function of alumina content.

both the glass and the glass-ceramic matrix composites were linearly increased with increasing alumina vol% due to incorporation of alumina having higher dielectric constant ($\epsilon=11$). In both cases, the dielectric constant values were matched with the theoretically predicted values. XRD analysis revealed that the major crystalline phase was changed as alumina content increased. Fig. 10 shows the XRD patterns of CASB glass containing up to

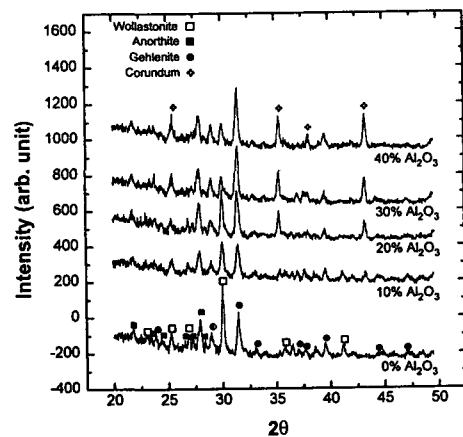


Fig. 10. XRD patterns of alumina-filled CASB glass-ceramic composite after sintered at 1000°C for 20 min. The average particle size of alumina = 4 μm.

40% alumina sintered at 1000°C for 20 min. The pure CASB glass, with no alumina inclusions, exhibited multiphase crystallization and the major crystallized phase was wollastonite. However, as the vol% of alumina inclusions were increased, wollastonite peak was decreased. And when alumina was added higher than 30%, gehlenite phase became the major crystallized phase instead of wollastonite. This result indicated that the crystalline gehlenite was crystallized from the Al_2O_3 -rich glass, which contains more Al_2O_3 than did the original glass; In other words, gehlenite crystallization was attributed to the partial dissolution of alumina. Precipitation of gehlenite as the major phase throughout the matrix did not increase the dielectric constant. And this result indicated that the dielectric constant of gehlenite is probably similar to wollastonite and anorthite. Wollastonite and anorthite phases have been frequently used as microelectronic packaging substrate due to their low dielectric constant.^{19-21,23} In this study, it was found that the glass-ceramic having gehlenite phase also can be used as the substrate material.

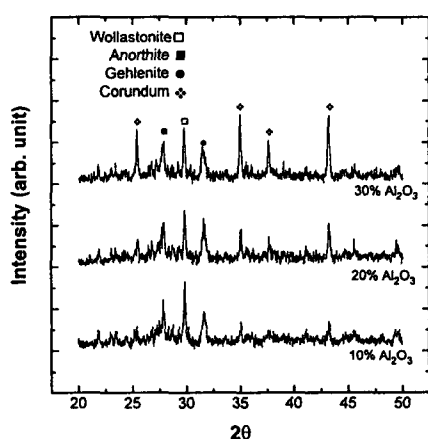


Fig. 11. XRD patterns of the alumina-filled CASB glass-ceramic composite after sintered at 1000°C for 20 min. The average particle size of alumina = 15 μm .

A larger size of alumina powder, classified to a narrow size range about an average of $\approx 15\mu\text{m}$, were mixed with the glass powder and the mixture sintered at 1000°C for 20 min. XRD patterns of the obtained samples are shown in Fig. 11. It is clearly shown that, wollastonite remains as the main crystalline phase even though the alumina content increases because of no dissolution of alumina.

This result indicates the alumina dissolution strongly depends on the particle size of alumina.

4. Conclusions

The crystallizable glass powder in $\text{CaO-Al}_2\text{O}_3\text{-SiO}_2\text{-B}_2\text{O}_3$ system was mixed with various amounts of alumina particle, and their sintering behavior, crystallization behavior, and dielectric property were examined in terms of vol. % of alumina and the reaction between the alumina inclusions and the CASB glass.

Sintering of the pure CASB glass powder alone at 900°C resulted in full densification. Sintering of the mixture of the glass powder and alumina inclusions at 900°C revealed that the amount of incorporated alumina inclusions should be less than 40% in order to obtain a substantial densification (higher than 97% of theoretical density). XRD analysis revealed that there was a partial dissolution of alumina into the glass. This reaction did not greatly affect the densification mechanism since no sign of alumina particle growth and shape accommodation was observed. Therefore, it was concluded that the sintering of both the pure glass and the alumina-filled composite was mainly achieved by the initial stage of liquid phase sintering; viscous flow and redistribution of the glass. The alumina dissolution, however, accelerated the crystallization initiation time at 1000°C and hindered the densification of the glass. Therefore, in processing of the

alumina-filled composite, a rapid heating to 1000°C for the densification and the crystallization is not desirable. In addition, the alumina dissolution changed the crystallization behavior of the glass and the dielectric constant of the composite. As alumina dissolves into the glass matrix, the crystalline gehlenite is crystallized instead of wollastonite from the Al₂O₃-rich glass, which contains more Al₂O₃ than did the original glass. This alumina dissolution strongly depends on the particle size of alumina.

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