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Dielectric properties of new glass-ceramics for LTCC applied to microwave or millimeter-wave frequencies

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Abstract

A new glass-ceramics material called new-glass-ceramics (NGC) that consists of $MgAl_2O_4$ crystals (spinel) and highly crystallized Li–Mg–Zn–B–Si–O glass has been developed for microwave or millimeter-wave frequency applications. NGC can be sintered at temperatures below 1000 °C and co-fired with internal copper electrodes that have high electrical conductivity. Its dielectric constant is 7.4 and its Q value is higher than 2000 at 24 GHz. NGC specimens were investigated using X-ray diffraction analysis (XRD), scanning electron microscopy (SEM), and energy dispersive X-ray spectrometry (EDX). NGC mainly consists of $MgAl_2O_4$, $Mg_3B_2O_6$, and Li_2MgSiO_4 crystal phases, which have high Q values. Using NGC, we could make band pass filter (BPF) with the size 3.2 mm × 2.5 mm × 1.3 mm for fixed wireless access (F.W.A.) system at 26 GHz. This BPF can be mounted on circuit board by solder and shows good characteristics.

Keywords: Sintering; Microstructure; Dielectric properties; Glass ceramics; Functional applications

1. Introduction

Rapid progress continues to be made towards achieving high-speed and high-frequency processing of electronic devices, requiring the electronic components and devices to have ever-higher processing speed and higher integration density. To meet these requirements, low-temperature co-fired ceramic (LTCC) materials are being used in multilayered circuit boards and passive devices (e.g., filters) operating at microwave frequencies.

At microwave or millimeter-wave frequencies (especially over 10 GHz), the dielectric loss is comparable to the electrode loss. Hence, low dielectric loss is required (i.e., $\tan\delta < 0.001$) at over 10 GHz. On the other hand, the wavelength in the dielectric material is proportional to a reciprocal number of a square root of dielectric constant. The materials that have high dielectric constant can decrease device sizes. However, the wavelength became comparable to accuracy of conductor patterning which we can achieve at over 10 GHz. Using high k material requires high accuracy of processing. As a result, it is difficult to form accurate conductor pattern in LTCC for millimetre-wave applications without advanced printing techniques, so that the materials that has high dielectric constant are not required for them.

In response to this problem, LTCC materials that have a high Q value at millimeter-wave frequencies have been developed.[1–3]

To enable the use of multilayered devices at microwave or millimeter-wave frequencies, we developed a new glass-ceramics material called NGC, which has a low dielectric constant and a high Q value at these frequencies.

In this paper, we discuss our investigation of the relationships among the dielectric properties, crystal phases, microstructures, plating system and we introduce a new application, band pass filter (BPF) for fixed wireless access (F.W.A.) system at 26 GHz, using the NGC.

2. Experimental

2.1. Specimen preparation

Specimens of NGC were prepared from $MgAl_2O_4$ and Li–Mg–Zn–B–Si–O glass. $MgAl_2O_4$ powder has high purity (>99%) with a mean particle diameter of 1.0 μm. Mg–Zn–B–Si–O glass also has a mean particle diameter of 1.0 μm. Compounds of $MgAl_2O_4$ and Mg–Zn–B–Si–O glass were mixed to make slurry containing zirconia balls, organic solvent, and organic binder. The slurry was cast into green sheets using doctor blade method. The sheets were cut and laminated to

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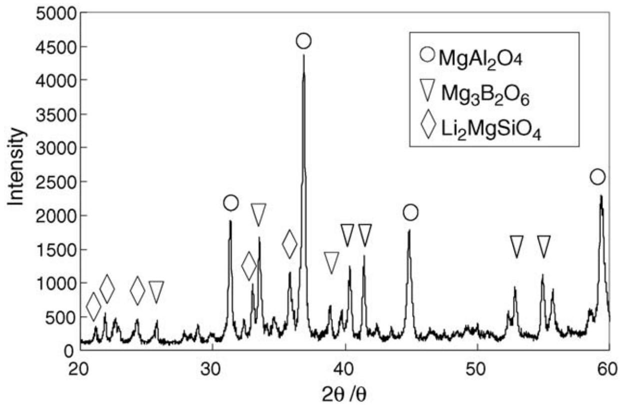

Fig. 1. XRD (Cu Kα) chart of NGC.

make blocks. The blocks were then fired in a low-oxygen partial pressure atmosphere at temperature below 1000 °C.

2.2. Measurements

The dielectric properties at microwave frequencies were measured using the circular cavity resonator method[4] using a cavity with hollow resonant frequency of 35 GHz. The thickness of the measured specimens was about 500 μm. The dielectric constant was approximately 7.4, and the resonance frequency was approximately 24 GHz. The temperature coefficient of the resonance frequency (τf) was measured at 15 GHz in the temperature range −55 to 85 °C.

To clarify the relationship between the firing process and chemical stability, the specimens were pressure-cooker tested for 100 h at 120 °C and 95% RH. Before and after the test, the dielectric properties of the specimens were measured using the circular cavity resonator method to estimate the degradation in the *Q* values.

The specimens were investigated using scanning electron microscopy (SEM), energy dispersive X-ray spectrometry (EDX), and X-ray diffraction analysis (XRD). The mechanical strength was measured using a three-point bending test, the thermal expansion coefficient was measured using thermal mechanical analysis, and the thermal conductivity was measured using the razor-flash method.

3. Results and discussion

3.1. Dielectric properties of NGC

The results showed that dielectric constant and the *Q* value of the NGC are 7.4 and 2200 at 24 GHz, respectively, when fired at 940 °C in reducing atmosphere. It showed also chemical stability in good level.

Fig. 1 shows the XRD pattern of NGC. The main crystal phases detected were those of $MgAl_2O_4$, $Mg_3B_2O_6$ (Kotoite), and Li_2MgSiO_4, but small amounts of $Mg_2B_2O_5$ and Mg_2SiO_4 crystal phases were also detected. No hollows indicating the presence of glass phase were found, leading us to conclude that the amount of glass phase causing degradation of *Q* value is small. Table 1 shows the dielectric properties of the main crystals in NGC. The *Q* values of the $MgAl_2O_4$ and $Mg_3B_2O_6$ crystal phases were very high. Also, Li_2MgSiO_4 had a high *Q* value. Anticipated *Q* value of NGC from those of crystal phases is higher than that of practical NGC's *Q* value. This means that just a small amount of phase causing a low *Q* value is present. To attain a high *Q* value at over 10 GHz we must thus control not only the amount of phases having high loss, but also chemical composition of these.

Table 1
Dielectric properties of NGC materials

Material	$MgAl_2O_4$	$Mg_3B_2O_6$	Li_2MgSiO_4
Dielectric constant	8.5	7.2	8.5
Q value	8400 at 14 GHz	9400 at 16 GHz	2000 at 15 GHz

3.2. Sintering behavior

Fig. 2 shows the shrinkage and DSC curves of NGC. The exothermic peak from 250 to 400 °C was due to combustion of the organic binders. The NGC showed two-step shrinkage during sintering. Shrinkage in the first step, which occurred at about 600 °C, was due to softening of the glass, and this shrinkage ended with crystallization of the glass. Based on high-temperature XRD, these crystals were $Mg_3B_2O_6$ and Li_2MgSiO_4 and grew on a plateau from 650 to 850 °C. Shrinkage in the second step, which occurred at about 900 °C, was caused by melting of the Li_2MgSiO_4 crystal phase that was crystallized from glass. The molten Li_2MgSiO_4 was re-crystallized during the cooling step.

3.3. NGC microstructure

Fig. 3 shows a SEM image of NGC (cross-section of ceramics). XRD and EDX analyses revealed the presence of $MgAl_2O_4$ and $Mg_3B_2O_6$ crystals. The $MgAl_2O_4$ crystals were about 1 μm in diameter and did not show any grain growth. The $Mg_3B_2O_6$ crystals, which were generated by the crystallization of glass, were 2–3 μm in diameter. There were also domains we

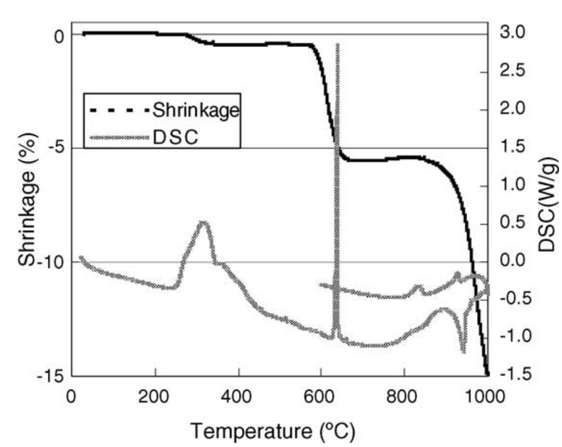

Fig. 2. Shrinkage and DSC curves of NGC.

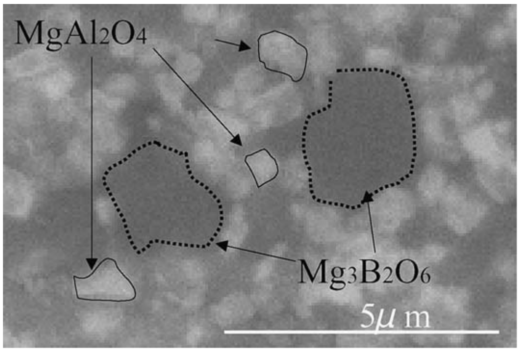

Fig. 3. SEM image of NGC.

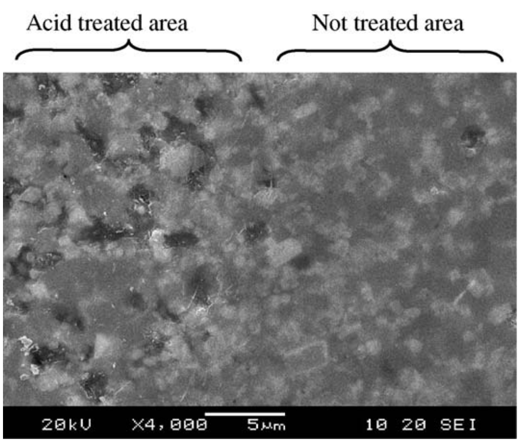

Fig. 4. SEM image of NGC with half of image was soaked part in weak acid.

considered to be Li_2MgSiO_4 crystal phase, but the crystal shape in Fig. 4 does not indicate the existence of any such crystal phase.

Fig. 4 shows SEM image of NGC (cross-section of ceramics) soaked in weak acid (pH 4.5), left half was the soaked part in this acid. The soaked part of NGC has many pores that were eroded by weak acid. Fig. 5 shows the XRD pattern of soaked NGC and non-soaked one. Soaked NGC has fewer Li_2MgSiO_4 than non-soaked NGC. Also, critical difference shows between the soaked parts and other parts at silicon content by the analysis of EDX. The content of silicon at soaked parts is lower than that of non-soaked parts. So we consider that Li_2MgSiO_4 was eroded by weak acid.

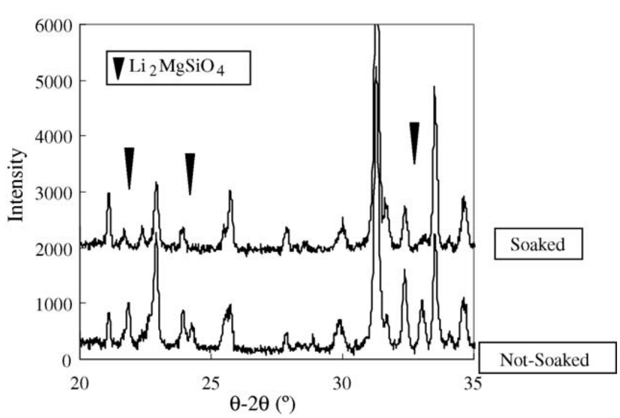

Fig. 5. XRD (Cu Kα) chart of NGC soaked part in weak acid.

Table 2
Representative properties of NGC

	NGC
Dielectric constant	7.4
Q value at 24 GHz	2200
Temperature coefficient of resonance frequency (ppm °C^{-1})	−90
Mechanical strength (MPa)	250
Thermal expansion coefficient (ppm °C^{-1})	10.6
Thermal conductivity (W m^{-1} K^{-1})	5.59

Generally plating bath shows weak acidity to stabilize metal ion. To plate copper electrodes on NGC, we must pay attention to the conditions of plating bath system in spite of high crystallization of NGC.

3.4. Other properties

Table 2 shows representative properties of NGC. Mechanical strength of 250 MPa was obtained, which is sufficient for devices and multilayer substrates used at microwave or millimeter-wave frequencies. We also obtained high thermal conductivity of 5.59 W m^{-1} K^{-1}. The mechanical strength and thermal conductivity values of NGC are higher than those of conventional LTCC materials; this is due to the high crystallization of NGC. The thermal expansion coefficient of NGC from room temperature to 600 °C was 10.6 ppm °C^{-1}.

The temperature coefficient of the resonance frequency was −90 ppm °C^{-1}. However, this is insufficient for filters used at microwave frequencies, since at those frequencies, filters need higher thermal stability. Further study is to be done to fulfil this demand.

3.5. Application

Using the NGC, we made a multi-layer BPF for F.W.A. system at 26 GHz. Fig. 6 shows the BPF whose size is 3.2 mm × 2.5 mm × 1.3 mm. Inner and outer copper electrodes were co-fired. Using electroless nickel plating bath system without lead, nickel films were plated on copper electrodes. Gold thin films were deposited on the nickel films.

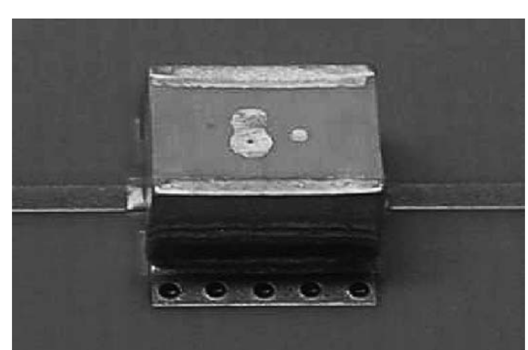

Fig. 6. Outlook of the BPF.

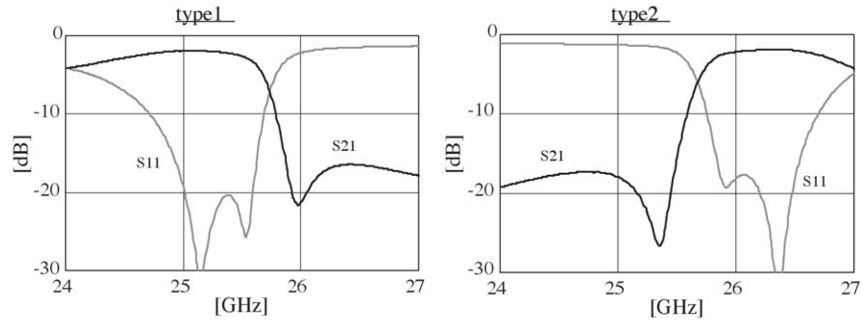

Fig. 7. Frequency characteristics of BPF on the test board (I.L. = 0.9 dB).

Fig. 7 shows frequency characteristics of the BPF from 24 to 27 GHz. This BPF had a low insertion loss which is less than below the 2.0 dB on the test board whose insertion loss is 0.9 dB at 26 GHz. The BPF were mounted on test board and connected by solder. Shielding electronic-magnetic field by the ground electrodes reduces interferences with surrounding of the devise.

4. Conclusion

The effect of firing profiles on crystal phases, microstructures, and dielectric properties of a newly developed glass-ceramics NGC were investigated. We obtained a Q value of 2200 at 24 GHz. The mechanical strength was 250 MPa, and the temperature coefficient of the resonance frequency was $-90\ \text{ppm}\ ^{\circ}\text{C}^{-1}$.

Thus, except for the temperature coefficient of resonance frequency, NGC was found to have good properties. To apply NGC in devices that require high thermal stability, it will be necessary to improve the temperature coefficient of its resonance frequency.

To realize high Q, we should control a small amount of residual phases after crystallized.

We made BPF for F.W.A. system at 26 GHz. This BPF has good properties whose insertion loss is below the 2.0 dB on the test board at 26 GHz.

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