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Microwave dielectric properties of SnO-SnF_2-P_2O_5 glass and its composite with alumina for ULTCC applications

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Abstract

Ultralow-temperature sinterable alumina-$45SnF_2$:$25SnO$:$30P_2O_5$ glass (Al_2O_3-SSP glass) composite has been developed for microelectronic applications. The $45SnF_2$:$25SnO$:$30P_2O_5$ glass prepared by melt quenching from 450°C has a low T_g of about 93°C. The SSP glass has ε_r and tanδ of 20 and 0.007, respectively, at 1 MHz. In the microwave frequency range, it has ε_r=16 and $Q_u \times f$=990 GHz with τ_f=−290 ppm/°C at 6.2 GHz with coefficient of thermal expansion (CTE) value of 17.8 ppm/°C. A 30 wt.% Al_2O_3-70 wt.% SSP composite was prepared by sintering at different temperatures from 150°C to 400°C. The crystalline phases and dielectric properties vary with sintering temperature. The alumina-SSP composite sintered at 200°C has ε_r=5.41 with a tanδ of 0.01 (1 MHz) and at microwave frequencies it has ε_r=5.20 at 11 GHz with $Q_u \times f$=5500 GHz with temperature coefficient of resonant frequency (τ_f)=−18 ppm/°C. The CTE and room-temperature thermal conductivity of the composite sintered at 200°C are 8.7 ppm/°C and 0.47 W/m/K, respectively. The new composite has a low sintering temperature and is a possible candidate for ultralow-temperature cofired ceramics applications.

KEYWORDS

composites, dielectric materials/properties, glass-ceramics, LTCC, microwaves

1 | INTRODUCTION

There is an increased demand for new materials with attractive dielectric properties in the areas of wireless communication, satellite communication, electronic memory devices, and so on.[1,2] Low-temperature cofired ceramic technology (LTCC) is a well-established multilayer technology having numerous applications.[3] At present, extensive research is being carried out to develop new materials having sintering temperature less than 700°C.[2] Several glass-ceramics, molybdates, vanadates, tungstates, tellurates based ceramics are reported in the literature as ultralow-temperature cofired ceramics (ULTCC) materials.[2] Energy saving, reduction in processing time, as well as cost effectiveness, and cofiring with conductors such as nanosilver ink are the attractive features of ULTCC technology.[2,4] For future applications, there is an urgent demand for the development of new ULTCC materials.[5]

Over the past several years, considerable efforts have been made by researchers to develop new glasses having low glass-transition temperature. Glasses having low glass-transition temperature (T_g) offers a wide variety of applications such as glass-metal sealing, IC packaging, and so on.[6] Among them, fluorophosphate glasses are of significant interest. In general fluorophosphate glasses have low T_g compared to silicate and borate glasses.[7] The optical properties of fluorophosphate glasses are comparable with that of fluoride glasses, and mechanical properties that of oxide glasses.[8] Besides this, another attracting property of phosphate glasses is its ability to associate fluoride compounds without losing the tendency to form glass.[9] Due to the nonlinear refractive index change, fluorophosphate glasses can be qualified as suitable candidates for high-power lasers.[10] Fluorine-containing glasses find application in the field of laser windows, lenses, and filters.[11] The introduction of network modifying cation tin (Sn) lowers the T_g of fluorophosphate glasses.[12]

Alumina (Al_2O_3) is considered as the gemstone of the electronic industry due to its excellent electrical and physical properties compared to other ceramics.[13] The sintering temperature of Al_2O_3 is relatively high at about 1600°C. The main challenge behind making Al_2O_3 as LTCC/ULTCC material is its high sintering temperature.[14,15] Low melting and low-loss glasses are known to be effective sintering aids for reducing the sintering temperature of various ceramics without much affecting the microwave properties.[3] The traditional method to lower the sintering temperature of the Al_2O_3 is by glass addition. Several authors reported the effective role of glass in reducing the sintering temperature of Al_2O_3.[16-26]

Several research groups have reported the preparation of ternary glass $SnO\text{-}SnF_2\text{-}P_2O_5$. The structure of SSP glass was studied by spectroscopic methods.[6,27,28] Liu et al. prepared different compositions of SSP glass with $x SnF_2\text{-}(70-x)$ $SnO\text{-}30P_2O_5$ glasses (x=40, 45, 50, 55, 60), and employed Fourier Transform Infrared and Xray photoelectron spectroscopy (XPS) techniques to understand the SSP glass structure for the different compositions.[6] From XPS analysis, they found that the change in glass network dimension was responsible for the variation in glass properties for different compositions. They reported that the T_g value decreases and the coefficient of thermal expansion (CTE) value increases with increase in SnF_2 content.[6] Although extensive work has been reported on the structure and properties of SSP glass, no attempt was done to study the dielectric properties. In this study, we report the radiofrequency and microwave dielectric properties of SSP glass and its composite with alumina.

2 | EXPERIMENTAL PROCEDURE

2.1 | Preparation of $45SnF_2{:}25SnO{:}30P_2O_5$ glass

Tin-based fluorophosphate ternary glass having the composition $45SnF_2{:}25SnO{:}30P_2O_5$ (SSP) was prepared by conventional melt quenching technique. A two-step melting process was adopted to minimize fluorine loss. Initially ammonium dihydrogen phosphate ($NH_4H_2PO_4$, ≥98%, Sigma Aldrich, St. Louis, MO, USA) was first melted at 450°C for 30 minutes. Then stoichiometrically weighed tin oxide (SnO, 99%, Alfa Aaesar, Tewksbury, MA, USA) and tin fluoride (SnF_2, 99%, Sigma Aldrich) were added one by one to the melt and again heated in the furnace at 450°C for 30 minutes. The whole melt was quenched in distilled water. The molten glass was poured into moulds having different dimensions for dielectric and thermal studies.

2.2 | Al_2O_3-SSP glass composite preparation

The aim of our work was to develop alumina-SSP glass composite suitable for ULTCC applications. Different weight % of alumina and SSP glasses had been prepared. Only 30 wt.% alumina and 70 wt.% SSP glass compositions can be sintered below 400°C with good dielectric properties suitable for ULTCC applications. The sintering temperature was found to increase to above the ULTCC range with increase in alumina content. Only the composition which contains 30 wt.% Al_2O_3-70 wt.% SSP glass yields the required ULTCC temperature range. The suitable composition between Al_2O_3 and $45SnF_2{:}25SnO{:}30P_2O_5$ glass having low sintering temperature was selected for further studies. The composite with 30 wt.% Al_2O_3-70 wt.% $SnO\text{-}SnF_2\text{-}P_2O_5$ glass was prepared by mixing the raw materials using acetone for 10 hours. The slurry was dried and ground well. The Al_2O_3-SSP composite was sintered at different temperatures from 150°C to 400°C.

2.3 | Characterization techniques

The amorphous nature of the as-prepared glass as well as the phase composition of the 30 wt.% Al_2O_3-70 wt.% $SnO\text{-}SnF_2\text{-}P_2O_5$ glass composite was studied using XRay diffractometer (XRD) (CuK_α radiation, PANanalytical X'Pert PRO diffractometer, Netherlands). The spectra of the powder glass were recorded using PerkinElmer Series FT-IR Spectrometer, USA. The glass-transition temperature of the powder glass sample was determined using differential scanning calorimeter (DSC) analysis (TA Instruments Q2000 DSC with Refrigerated Cooling System (RCS), New Castle, DE, USA). For DSC analysis, the glass powder was heated from 30°C to 300°C with a heating rate of 10°/min. Thermo gravimetric/differential thermal analysis (TG/DTA) analysis was done for the glass powder (STA 7300, Thermal Analysis System, Hitachi, Japan) from 30°C to 700°C with a heating rate of 10°/min under nitrogen atmosphere. The density of the SSP glass was measured using Archimedes method using distilled water medium and the density of Al_2O_3-SSP glass composite was determined using dimensional method with the help of a digital screw gauge and weight measured using a semi-micron weighing balance (Shimdazu, AUW220D, Japan). The microstructure of the composite was studied using JOEL-JSM 5600 LV, Tokyo, Japan. The radio (low)-frequency dielectric properties of the glass powder having dimensions 8 mm diameter and 2 mm thickness were determined by parallel plate capacitor method using an inductance capacitance resistance (LCR) meter (Hioki 3532-50LCR Hi Tester, Japan). For the radiofrequency measurements of the bulk Al_2O_3-SSP composite, samples having 11 mm diameter and 1.5 mm thickness, electrode with copper on both sides were used. The thermal conductivity of the composite was measured using a laser flash thermal properties analyzer (Flash-Line2000, Anter Corporation, Pittsburgh, PA, USA). The

thermal expansions of the glass pellet as well as the Al_2O_3-SSP glass composite were measured using thermo mechanical analyzer (Exstar-TMA/SS7300, SII Nano Technology, Inc., Tokyo, Japan). The microwave dielectric properties were measured using a network analyzer (Agilent E5071C ENA series, Agilent Technologies, Santa Clara, CA, USA) operating in the frequency region 300 kHz to 20 GHz. SSP glass samples having 14.2 mm diameter and 6.7 mm thickness were used for microwave measurements. In the case of Al_2O_3-SSP composites, samples having 11 mm diameter and 5.4 mm thickness were used. The dielectric constant was measured by Hakki-Coleman method.[9] The unloaded quality factor was determined using resonant cavity method in the $TE_{01\delta}$ mode.[9] The temperature coefficient of resonant frequency (τ_f) of the SSP glass and the Al_2O_3-SSP glass composite was measured in the temperature range 25°C-60°C using the cavity method.[9] τ_f was determined using the formula:

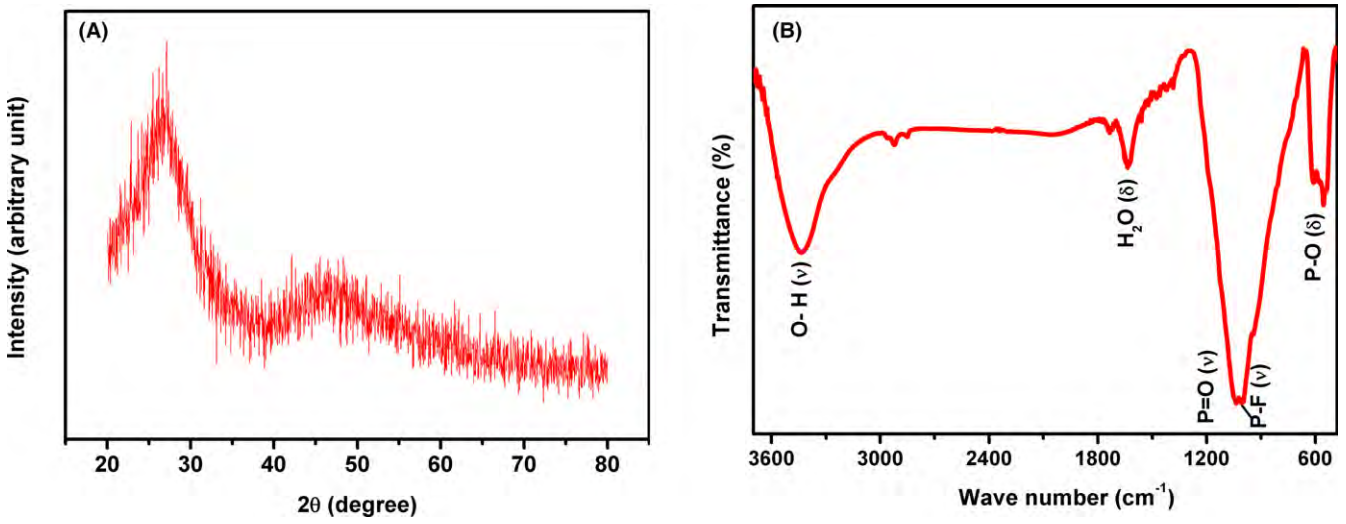

FIGURE 1 XRD pattern (A) and room-temperature FT-IR spectrum (B) of $45SnF_2$:25SnO:$30P_2O_5$ glass

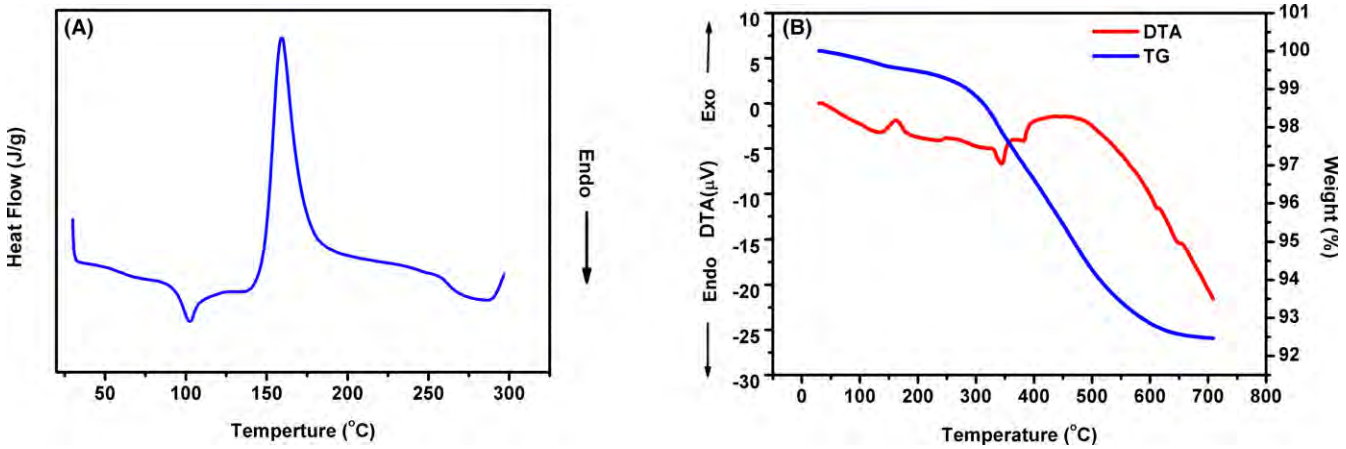

FIGURE 2 DSC and TG/DTA curve of SSP glass powder

TABLE 1 Dielectric properties of SSP glass having density 4.38 g cm^{-3}

Dielectric properties at 1 MHz		Dielectric properties at 5 GHz using split post dielectric resonator		Dielectric properties at using ;Hakki-Coleman method		
ε_r	tanδ	ε_r	tanδ	ε_r (6.2 GHz) (Hakki)	$Q_u \times f$ (GHz) (Cavity)	τ_f (ppm/°C)
20	0.02	17	0.006	16	990	−290

SPDR, split post dielectric resonator.

$$f = \frac{1\Delta f}{f\Delta T} \quad (1)$$

where f is the resonant frequency at room temperature and Δf is the variation in resonant frequency for a change in ΔT.

3 | RESULTS AND DISCUSSIONS

3.1 | Structural, dielectric, and thermal properties of SSP glass

Figure 1A shows the XRD pattern of the SSP glass indicating the amorphous nature. The absence of crystalline phase in the XRD confirms the formation of $45SnF_2$:25SnO:$30P_2O_5$ glass prepared by quenching method. Figure 1B shows the room-temperature FT-IR spectrum of $45SnF_2$:25SnO:$30P_2O_5$ glass powder in the range 4000 to 400 cm^{-1}. The spectra indicate the presence of OH which might be due to the moisture absorption during the preparation of the sample using KBr. The band at 3438 cm^{-1} can be attributed to the stretching vibration of O–H bond, 1630 cm^{-1} due to the bending vibration of H–OH bond. The bands corresponding to 1100 to 930 cm^{-1} is due to the stretching vibration of P=O bond. The band around 630 cm^{-1} is attributed to the P–O bending vibration. The band at 1028 cm^{-1} corresponds to the P–F stretching vibration.[6]

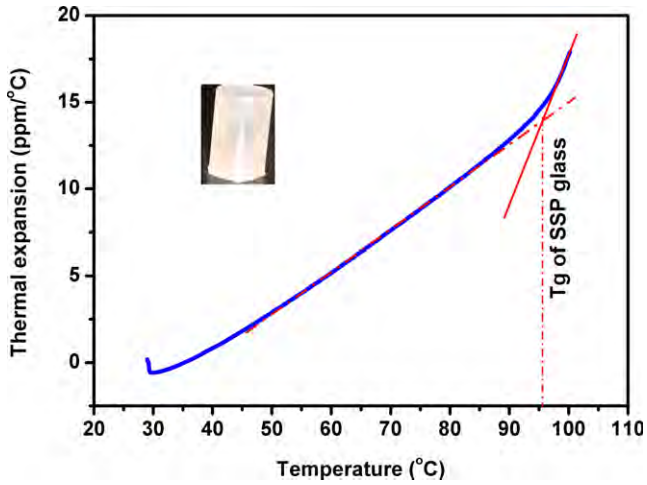

FIGURE 3 Variation in thermal expansion with temperature for SSP glass

The glass-transition temperature of $45SnF_2$:25SnO:$30P_2O_5$ glass as determined by the DSC is found to be about ~93°C (see Figure 2A). Liu et al. reported that the glass-transition temperature of $45SnF_2$:25SnO:$30P_2O_5$ at about 135°C.[6] The difference in the T_g value in the present study as compared to the reported value may be due to the difference in melting time duration, origin and purity of chemicals, furnace heating conditions, and so on. The TG/DTA of SSP glass is shown in Figure 2B. The exothermic peak in the DTA curve at 163°C is attributed to crystallization of SnF_2 and SnO_2 formed by the oxidation of SnO_2 and the endothermic peak at ~343°C in the DTA curve corresponds to the onset of the melting of the individual components in the glass, particularly SnF_2 as evident from XRD (Figure 5). The weight loss found in the TG curve can be attributed to the volatilization loss of fluorine and hydroxyl by forming hydrogen Fluoride during melting.[6]

Table 1 gives the dielectric properties of SSP glass. In general, the density of the glasses depends on the atomic weight of the individual components used in the glass as

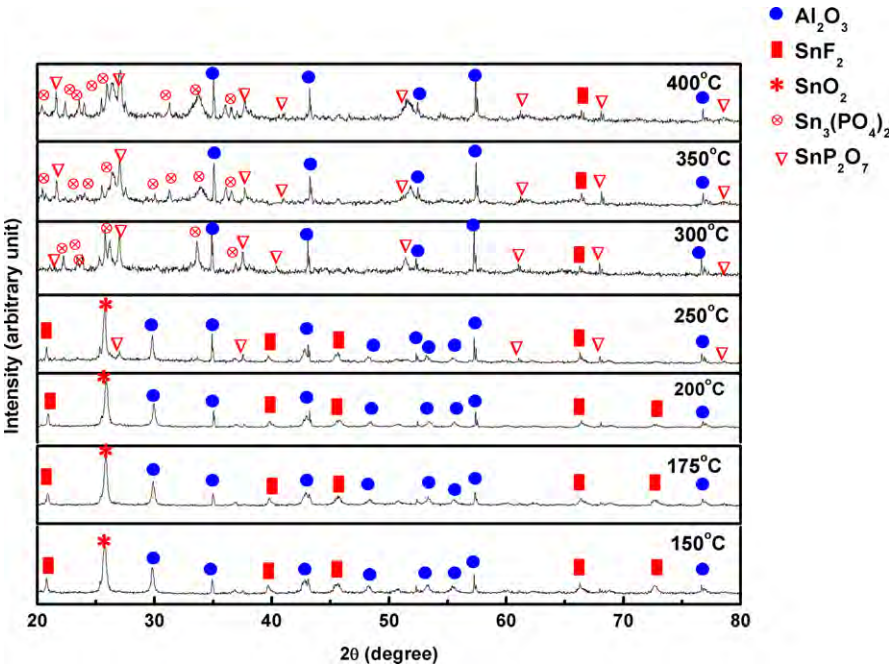

FIGURE 4 XRD pattern of Al_2O_3-SSP glass composite sintered at different temperatures (150°C-400°C)

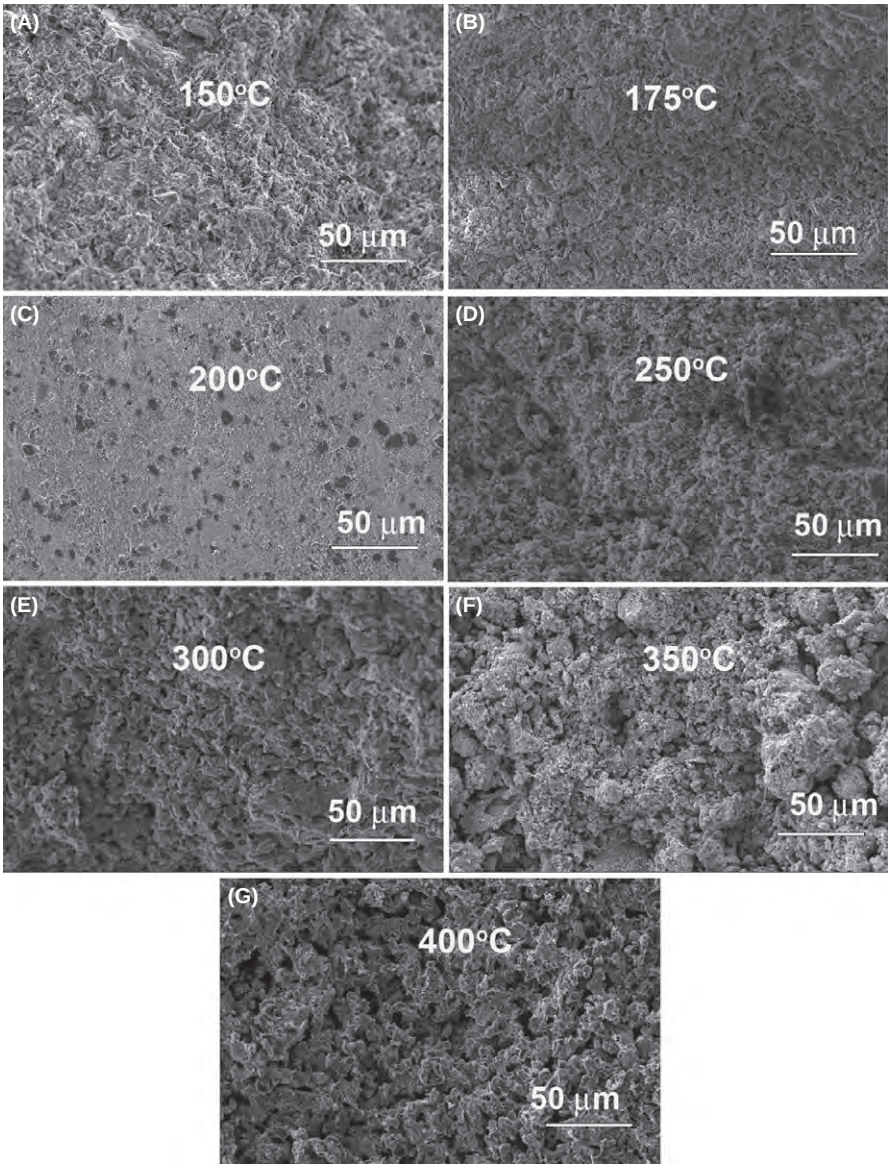

FIGURE 5 Microstructure of fractured surfaces of Al_2O_3-SSP glass composite sintered at (A) 150°C, (B) 175°C, (C) 200°C, (D) 250°C, (E) 300°C, (F) 350°C, and (G) 400°C

well as the compactness of the structure units in the glass.[29] The density of the SSP glass is obtained as 4.38 g cm^{-3}. For most of the glasses, the dielectric constant depends on ionic and electronic polarization, while the dielectric loss depends on relaxations.[4] Dielectric loss in the glass is mainly due to conductance, ion relaxation, deformation, and vibration.[11] At low frequencies, ion relaxation and conductance losses are predominant.[5] The dielectric constant and dielectric loss at 1 MHz are 20 and 0.02, respectively. The dielectric constant for $45SnF_2$:25SnO:$30P_2O_5$ glass at microwave frequency is found to be 16 (6.2 GHz). The quality factor ($Q_u \times f$) is obtained as 990 GHz. The ε_r value for the thin sheet of $45SnF_2$:25SnO:$30P_2O_5$ glass measured at 5 GHz is 17 with tanδ 0.006. The small variation in ε_r of SSP glass in two methods (Hakki and split post dielectric resonator [SPDR]) is due to the difference in frequency used for measurement and the use of thin sheet and large bulk samples with slight variations in density. The density reported for the bulk SSP glass in the present study is 4.38 g cm^{-3}. However, the sample used for SPDR measurement is very thin compared to the bulk and has a lower density of 4.30 g cm^{-3}. The difference in density of the samples used for determining the dielectric constant using Hakki and SPDR method is the possible reason for the difference in dielectric constant. In the microwave frequency region, only ionic and electronic polarization contributes to dielectric constant.[12] For practical applications, the resonant frequency should be independent of temperature variations. τ_f value of SSP glass is found to be -290 ppm/°C measured in the temperature range 25°C-60°C.

The knowledge about thermal expansion studies of the SSP glass is very crucial for practical applications. Figure 3 shows the linear expansion of SSP glass in the temperature range 30°C-100°C. The CTE value for the $45SnF_2$:25SnO:$30P_2O_5$ is found to be 17.8 ppm/°C which is very close to

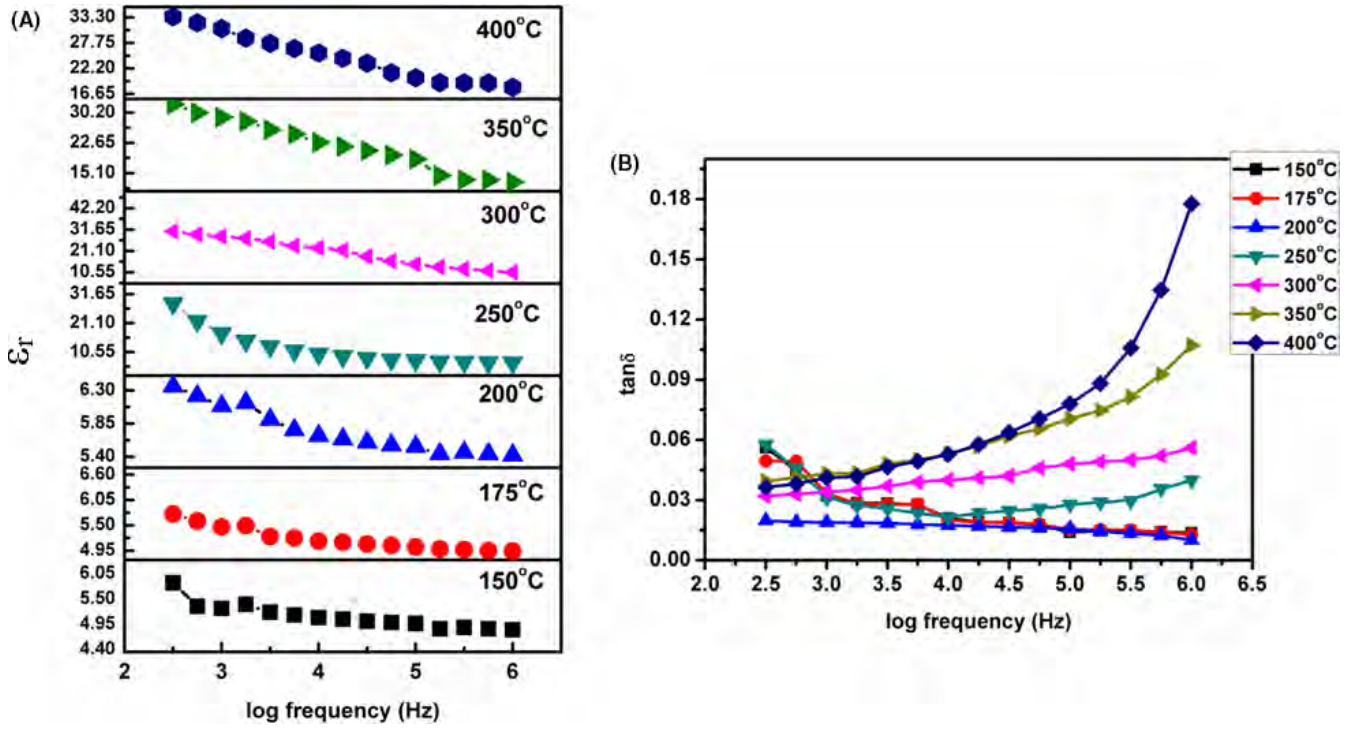

FIGURE 6 Variation in (A) dielectric constant and (B) dielectric loss with radiofrequency for Al_2O_3-SSP glass composites sintered at different temperatures (150°C-400°C)

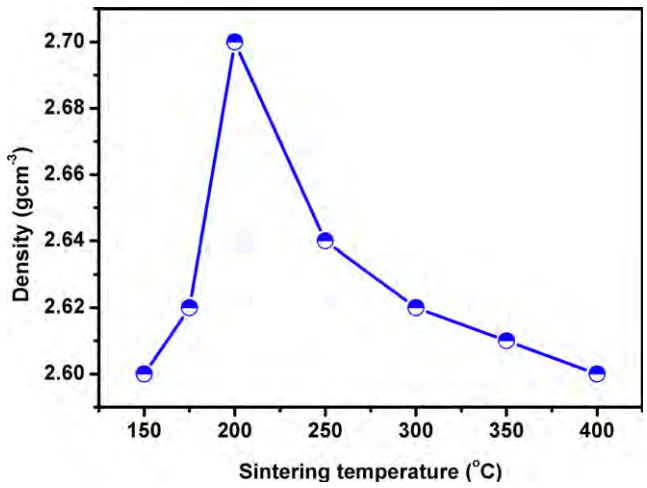

FIGURE 7 Variation in density with sintering temperature for Al_2O_3-SSP glass

that reported by Liu et al. The photograph of SSP glass is shown in the inset of Figure 3. It has been reported that low T_g glasses would have higher CTE value.[6] The T_g value obtained for the SSP glass from CTE measurement is about ~95°C and is in agreement with that obtained from DSC analysis (Figure 2A).

3.2 | Structural, dielectric, and thermal properties of Al_2O_3-SSP glass composite

The XRD patterns of the Al_2O_3-SSP glass composite sintered at different temperatures (150°C-400°C) are shown in Figure 4. It is found that SSP glass which has a low T_g glass, recrystallizes to form the individual components of the glass and react to form secondary phases at higher temperatures. The XRD pattern shows the peaks of Al_2O_3 (JCPDS file no. 88-0107), SnO_2 (JCPDS file no. 88-0287), SnF_2 (JCPDS file no. 71-2018), and secondary phases of SnP_2O_7 (JCPDS file no. 75-1143) and $Sn_3(PO_4)_2$ (JCPDS file no. 70-0391) are also identified. The DSC and TG/DTA analysis of the SSP glass indicates that crystallization starts at about 150°C. The glass in the composite crystallizes into SnO_2, SnF_2 and is evident in the XRD of the sample sintered at 150°C. Above 200°C, the SnF_2 melts and peaks become very weak indicating that fluorine is escaping. The SnO_2 reacts with P_2O_5 to form SnP_2O_7 and $Sn_3(PO_4)_2$ and is supported by the DSC and TG/DTA curves.

Figure 5 shows the microstructure of the fractured surface of Al_2O_3-SSP glass composite in the temperature range 150°C-400°C. The SEM pictures show the presence of secondary phases. Al_2O_3 do not react with SSP glass in the entire sintering temperature range. The porosity of the composite increases above 200°C due to escape of

fluorine. The deficiency of fluorine lead to the crystallization of SnP_2O_7 and $Sn_3(PO_4)_2$ at temperatures above 200°C.

Figure 6 shows the variation in dielectric properties of the Al_2O_3-SSP glass composite measured in the radiofrequency region for the samples sintered at different temperature range 150°C-400°C. The dielectric constant is found to increase with increase in sintering temperature and is attributed to the formation of secondary phases. The increased amount of secondary phase formation and phase transition of tin-based phosphates may be the possible reason for the increase in dielectric constant at higher temperatures. The formation of secondary phases during sintering at different temperatures and vaporization of fluorine may be responsible for the variations in dielectric properties. The contribution of extrinsic loss is more predominant in Al_2O_3-SSP glass composite. Impurities, microstructural defects, grain boundaries, porosity, and so on, in the composite lead to high dielectric losses.[3,15,30] It has been reported that SnP_2O_7 undergoes phase transitions at 287°C and 557°C[31] which may affect the dielectric properties. The composite sintered at 200°C has ε_r=5.41 with a tanδ of 0.01 at 1 MHz. The composite sintered above 200°C, show an increase in dielectric loss with frequency due to the increased amount of porosity in the composite. At 200°C temperature, the dielectric properties of the composite are found to be good for ULTCC practical applications. The dielectric properties of the composite can be tailored according to the need by sintering at different temperatures. Figure 7 shows the variation of density with sintering temperature for Al_2O_3-SSP glass composite. The density of the composites drops after 200°C which in turn increases the porosity in the composite. The decrease in density after 200°C also supports the escape of fluorine from Al_2O_3-SSP composite. The dielectric properties of the secondary phase's particularly tin phosphates are not known due to the difficulty in getting sintered pellets of these materials. Hibino et al. also observed the same difficulty in sintering SnP_2O_7 ceramic.[32] The precise knowledge about the dielectric properties of these secondary phases are needed to account for the anomalous trend observed in dielectric properties over the range of sintering temperature.

Figure 8 shows the variation of ε_r, $Q_u \times f$, and τ_f for Al_2O_3-SSP glass composite as a function of sintering temperature at microwave frequency. In the microwave frequency, the dielectric constant varies between 4.6 and 17.0. The composite sintered at 200°C has dielectric constant of 5.20 (11 GHz). The microwave dielectric properties of the Al_2O_3-SSP glass composite revealed that with increase in sintering temperature, the $Q_u \times f$ values increases up to 350°C and further increase in sintering temperature decreases the quality factor. However, the dielectric

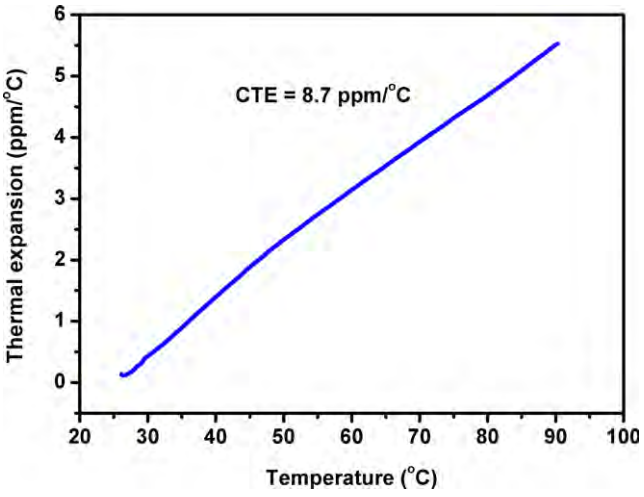

FIGURE 9 Thermal expansion study of Al_2O_3-SSP glass composite sintered at 200°C

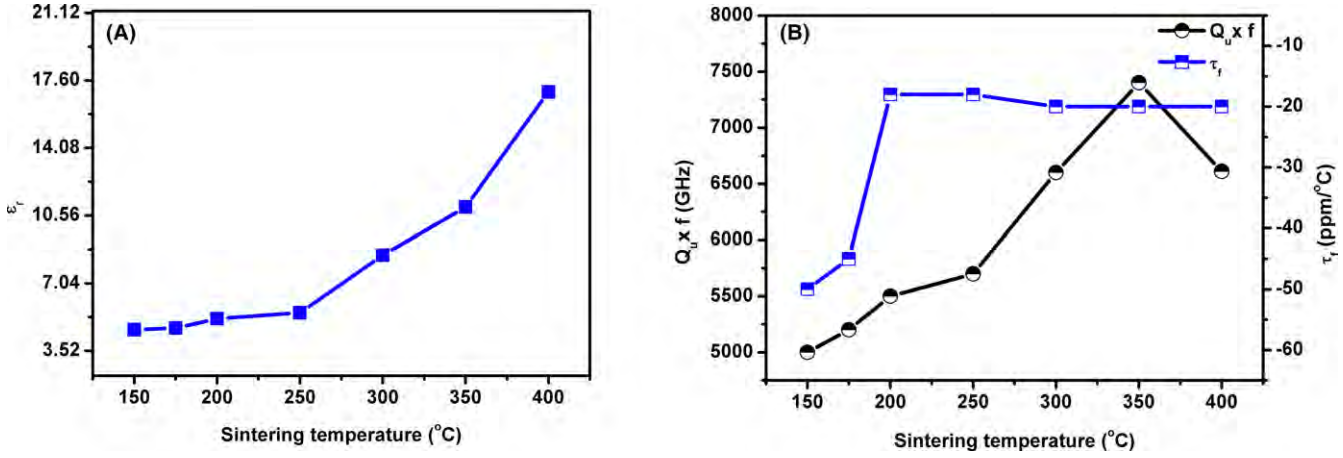

FIGURE 8 Variation in (A) ε_r, (B) $Q_u \times f$, and τ_f for Al_2O_3-SSP glass composite at microwave frequency with sintering temperature (150°C-400°C)

constant increases with increase in sintering temperature. It may be noted that the secondary phases are formed in the composite at different sintering temperatures. The τ_f value is small for the samples sintered at 200°C and 250°C. In the present study, the Al_2O_3-SSP glass composite sintered at 200°C (2 h) has $Q_u \times f$ value 5500 GHz with τ_f of −18 ppm/°C. The possible error in the measurement of ε_r using Hakki-Coleman method is of the order of 0.3%.[30] The increased glass content, secondary phase formation, and poor densification of the samples may be the reason for the relatively low quality factor. However, the material has a very low sintering temperature.

The thermal expansion studies of the Al_2O_3-SSP glass composite is carried out in the temperature range 30°C-90°C and the linear dimensional change with temperature is shown as Figure 9. The bulk composite sintered at 200°C has a CTE value of 8.7 ppm/°C. The thermal conductivity of the composite is very important for electronic applications. The room-temperature thermal conductivity of the Al_2O_3-SSP glass composite is 0.47 W/m/K. It has been reported that phosphate glasses have low thermal conductivity as well as higher thermal expansion as compared to the silicate glasses.[29]

4 | CONCLUSIONS

A novel composition based on Al_2O_3-SSP glass has been developed for ULTCC applications. The $45SnF_2$:$25SnO$:$30P_2O_5$ glass has good dielectric properties both in radio (ε_r of 20 and tanδ of 0.007 at 1 MHz) and microwave frequencies (ε_r of 16 [6.2 GHz], $Q_u \times f$=990 GHz with a τ_f value of −290 ppm/°C). The CTE value of $45SnF_2$:$25SnO$:$30P_2O_5$ glass is found to be 17.8 ppm/°C. The Al_2O_3-SSP glass composite sintered below 200°C indicates the presence of Al_2O_3, SnF_2 and SnO_2 and above 200°C, SnP_2O_7, $Sn_3(PO_4)_2$ are identified from the XRD. The Al_2O_3-SSP glass composite sintered at 200°C (2 h) has ε_r=5.41 with a tanδ of 0.01 at 1 MHz and ε_r=5.20 (11 GHz) with $Q_u \times f$=5500 GHz with temperature coefficient of resonant frequency (τ_f)=−18 ppm/°C at microwave frequency. The room-temperature thermal conductivity of the Al_2O_3-SSP glass composite is 0.47 W/m/K and CTE in the temperature range 30°C-90°C is found to be 8.7 ppm/°C.

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