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Microwave dielectric properties of **cold sintered Al_2O_3-NaCl composite**

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Abstract

Alumina-sodium chloride (Al_2O_3-NaCl) composite (1:1 ratio in weight percentage (wt %)) is cold sintered at 120°C. **The cold sintered composite with a densification of 96% has ε_r and tanδ values 6.53 and 0.007 respectively at 1 MHz.** The microwave dielectric properties of Al_2O_3-NaCl composite at 5 GHz are ε_r = 6.04 and tanδ 0.002 with τ_ε value +32 ppm/°C. **The energy saving cold sintering process (CSP) indicates the possibility to integrate with polymers** as well as metals for the fabrication of electronic products.

Keywords: Electroceramics, Cold sintering, Dielectrics, Ceramic composites

1. Introduction

Dielectric materials having attractive properties are of great importance in the field of wireless communication. Low dielectric constant (ε_r) and low loss (tanδ) materials can **increase** the signal propagation speed in communication systems. Cold sintering is a very useful technique for the development of next generation microelectronic devices. The main advantage of cold sintering process (CSP) is that it provides fast densification of ceramic materials at an extremely low temperature [1]. In CSP, densification is aided with the help of liquid solvent by applying sufficient amount of temperature and pressure [2]. Dissolution-precipitation is the key process in CSP method [2]. CSP is an **inexpensive** and energy saving method to fabricate dense ceramics.The low temperature sintering enables the incorporation of ceramics into polymers [3].CSP method has been successfully applied to a wide variety of materials including $LiMoO_4$, Y_2O_3-ZrO_2,$Na_2Mo_2O_7$, $K_2Mo_2O_7$, (1-x)Li_2MoO_4-xPTFE, $Li_{1.5}Al_{0.5}Ge_{1.5}(PO_4)_3$ (LAGP), V_2O_5, ZnO, sillimanite, etc[4, 5, 6, 7, 8, 9, 10]. Recently Funahashi et al. reported the possibility of integrating n-type and p-type thermoelectric oxides and a separating insulating layer to demonstrate functional multilayer thermoelectric generator devices through the CSP method [9].

Alumina is a well-known dielectric material having amazing dielectric, thermal as well as mechanical properties [11, 12,13]. Molla et al. studied the effect of Mg doping on thedielectric properties of Al_2O_3[14]. The ε_r, Q ×f value and τ_f value of nano α-Al_2O_3 ceramics without sintering aid at 1550°C for 4 h were 10, 521000 (at 14 GHz) and 48.9 ppm/°C respectively [13]. The effect of porosity and grain size on the microwave dielectric properties of Al_2O_3 was investigated in detail by Penn et al. and they concluded that when the grain size of Al_2O_3 exceeds 3-4µm, the tanδ value is found to increase but the ε_r value remains as a constant with grain size [15]. Ohsato et al. reported that use of high purity alumina powder can give a Q×f of 680,000 GHz with ε_r of 10.1 and temperature coefficient of resonant frequency (τ_f) of -60ppm/°C on sintering at 1550°C [16].

In the present paper we report the microwave dielectric properties of cold sintered **alumina-sodium chloride composite where NaCl act** as a the sintering aid.

2. Experimental

The alumina (Al_2O_3) (99.9 % ,<10 µm, Sigma Aldrich) and sodium chloride (NaCl) (**99%**, S D Fine Chem Ltd (SDFCL)) was used for the present study. A 1:1 ratio (in weight percentage (wt %)) was maintained between NaCl and Al_2O_3 for the preparation of dense NaCl-Al_2O_3composite.The Al_2O_3-NaCl composite was prepared as follows: first NaCl was moistened with 4 wt % deionized water and then Al_2O_3 was added to it in order to make a **semi-solid** paste. TheAl_2O_3-NaCl composite was then hot pressed using a die set at a temperature of about 120^{o}C (50 min) and pressure 200 MPa. **The thermocouple attached with the press recorded the temperature. The sintering temperature of the cold sintered Al_2O_3-NaCl composite is optimized after several trial and error methods and also by finding the density and comparing with the theoretical density of the composite. It may be possible to apply higher pressure with a further decrease in sintering temperature.**

The moisture present in the composite was removed by keeping the cold sintered Al_2O_3-NaCl composite in hot air oven at 120^{o}C for 24 h. The phase composition of Al_2O_3-NaCl was studied using XRD (CuKα radiation, PANalyticalX'Pert PRO diffractometer, Netherlands). The microstructure of the fractured surface of cold sintered Al_2O_3-NaCl sintered at 120^{o}C was recorded using Scanning Electron Microscope(JOEL-JSM 5600 LV, Tokyo, Japan and Zeiss, Germany). The dimensions of the cold sintered Al_2O_3-NaCl composite were determined using a digital screw gauge and weight using a semi-micron weighing balances (Shimdazu, AUW220D, Japan). **The cold sintered Al_2O_3-NaCl** composite having 11 mm diameter and 1.5 mm thickness coated with silver paste on both sides were used **for the measurement of radio frequency dielectric properties. Thin sheets having dimensions 40 mm × 40 mm and thickness 0.6 mm were used for microwave measurements of Al_2O_3-NaCl composite by the Split Post Dielectric Resonator method (SPDR) at 5 GHz.** The temperature coefficient of dielectric constant (τ_ε) was determined by keeping the samples inside a 5 GHz SPDR in the temperature range 25°C-60°C. **The dielectric measurements were carried in air atmosphere in an AC room.**

3.Results and discussion

Fig. 1 shows the Tg curve of Al_2O_3-NaCl composite.The weight loss is almost constant within the limits of experimental error up to 790°C. It appears that there is no moisture escape within this temperature range.The weight loss after 790°C is due to the decomposition of NaCl. The microstructure of the fractured surface of Al_2O_3-NaCl composite cold sintered at 120°C is shown as Fig.2(a). The backscattered image of the fractured surface (Fig.2 (b)) confirms the presence of Al_2O_3 and NaCl in the composite. The presence of any impurity phase is not visible in the microstructure which indicates the feasibility of the CSP method for the room temperature sintering of the Al_2O_3 material which is useful for the fabrication of substrates needed for the electronic industry.

The theoretical density of Al_2O_3-NaCl composite, calculated using equation (1) is obtained as 2.93 g/cm^3.

$$D = \frac{W_1 + W_2}{\frac{W_1}{\rho_1} + \frac{W_2}{\rho_2}} \quad (1)$$

where W_1 and W_2 are the weights of Al_2O_3 and NaCl respectively and ρ_1 and ρ_2 are the corresponding densities.

The experimentally obtained density for the composite is 2.82 g/cm^3. Thus CSP method helps to achieve 96 % densification on cold sintering at 120°C for Al_2O_3-NaCl composite.The ε_r and tanδ at 1 MHz for NaCl are 5.88 and 0.009 respectively [10]. The dielectric properties of Al_2O_3-NaCl composite are also measured in the frequency range 10 kHz - 1 MHz and is shown in Fig.3. The dielectric constant and loss tangent both decreased with increase in frequency. The Al_2O_3-NaCl composite has ε_r=6.53 with tanδ = 0.007 at 1 MHz. **The error associated with dielectric constant and dielectric loss is about1 %.**

The sintering aid NaCl has ε_r of 5.22 (~15.5 GHz), $Q_u \times f$ = 12,000 GHz and τ_f = -36ppm/°C [10]. The microwave dielectric properties of Al_2O_3-NaCl composite at 5 GHz are ε_r = 6.04 and tanδ = 0.002. The stability of dielectric constant with temperature of the composite is very important for practical applications [17]. The variation of dielectric constant with temperature is shown in Fig.4 and it is found that there is only a marginal change of ε_r with temperature. The τ_ε value for Al_2O_3-NaCl composite is +32 ppm/°C.**The error associated with SPDR measurements is about 1 % [12].**

4. Conclusion

Al_2O_3-NaCl **composite** is successfully cold sintered at 120^{o}C and its structural and micro structural studies are carried out. The composite has 96% densification. The low frequency ε_r = 6.53 and tanδ =0.007. Al_2O_3-NaCl composite shows good microwave dielectric properties with ε_r = 6.04 and tanδ 0.002 with τ_ε value +32 ppm/°C at 5 GHz. The low temperature sintering of Al_2O_3 using NaCl sintering **aid can be a** potential candidate for future substrate applications.

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List of figures

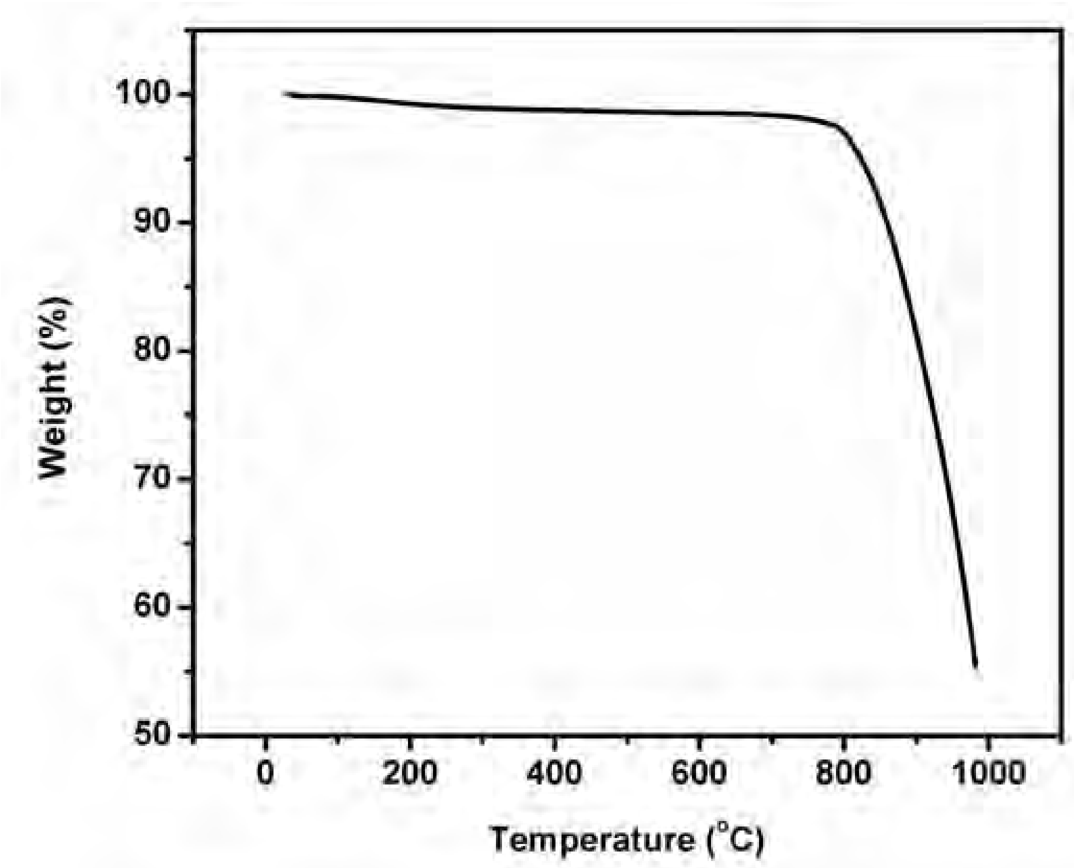

Fig.1. TG curve of Al_2O_3-NaCl composite.

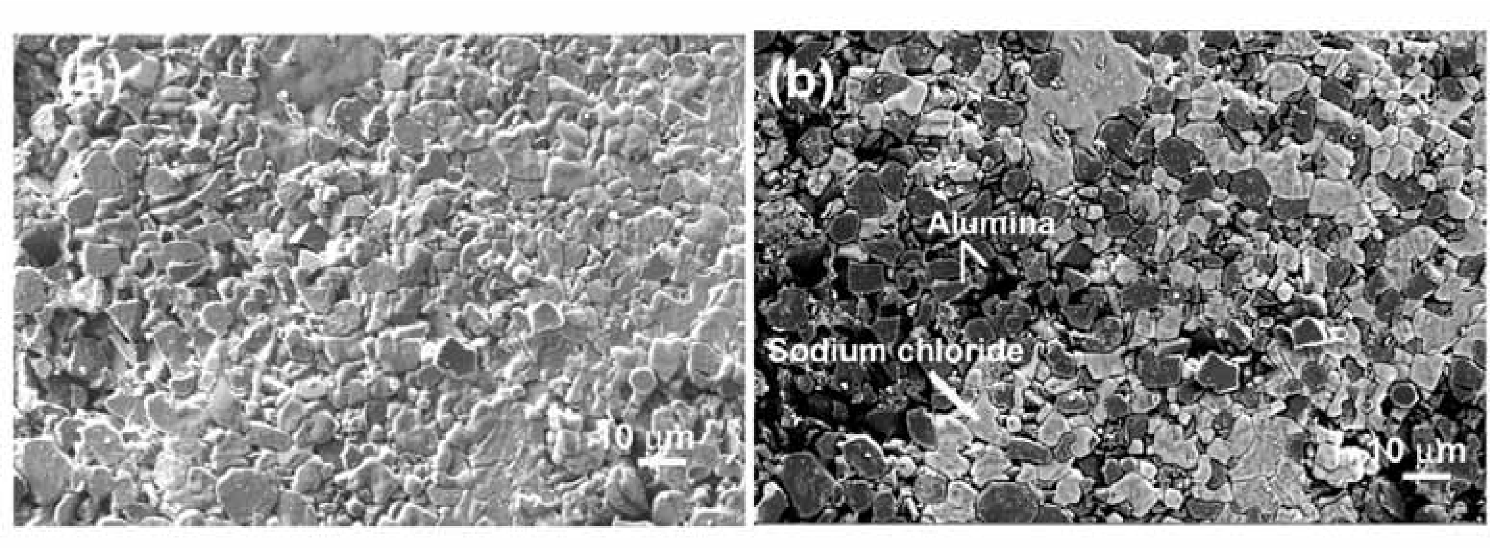

Fig.2 (a) Fractured surface of Al_2O_3-NaCl composite (b) backscattered image of Al_2O_3-NaCl composite cold sintered at 120^oC.

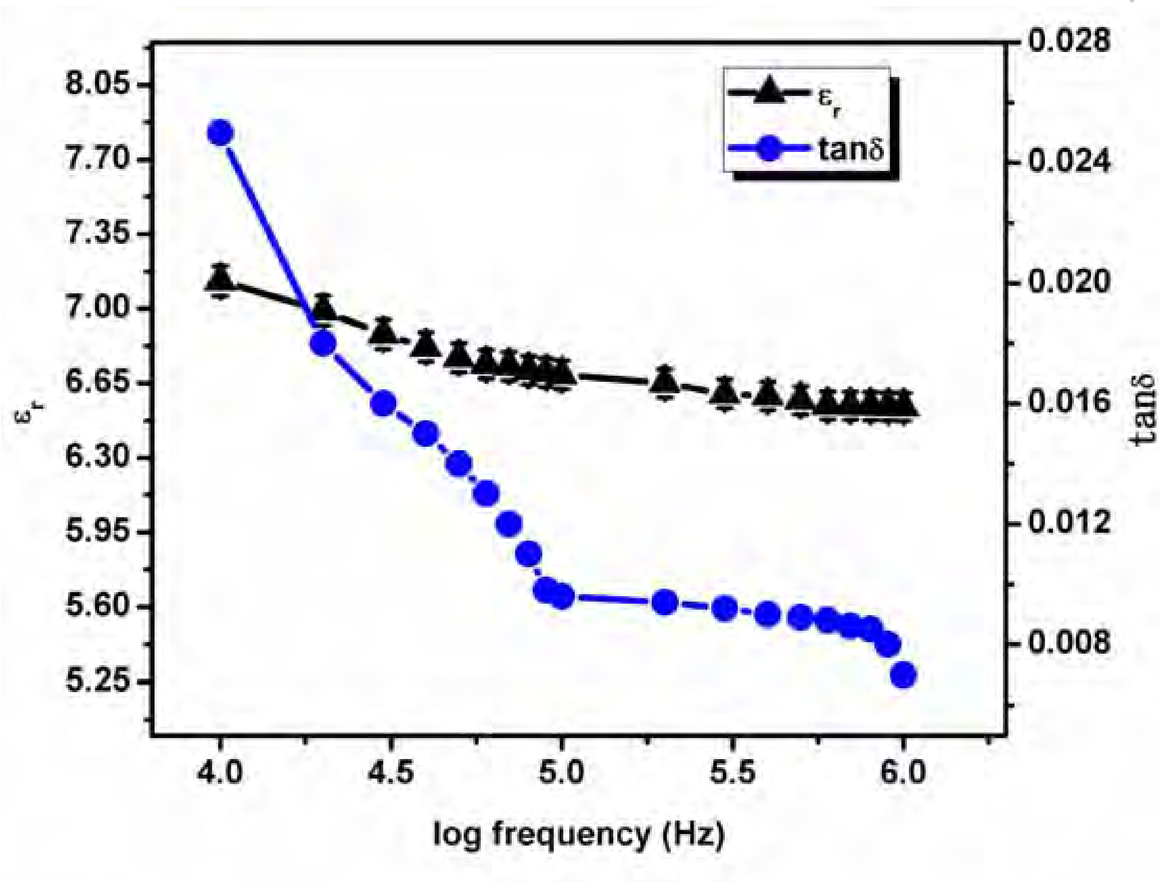

Fig.3. Variation of dielectric properties with radio frequency for Al_2O_3-NaCl composite cold sintered at 120°C.

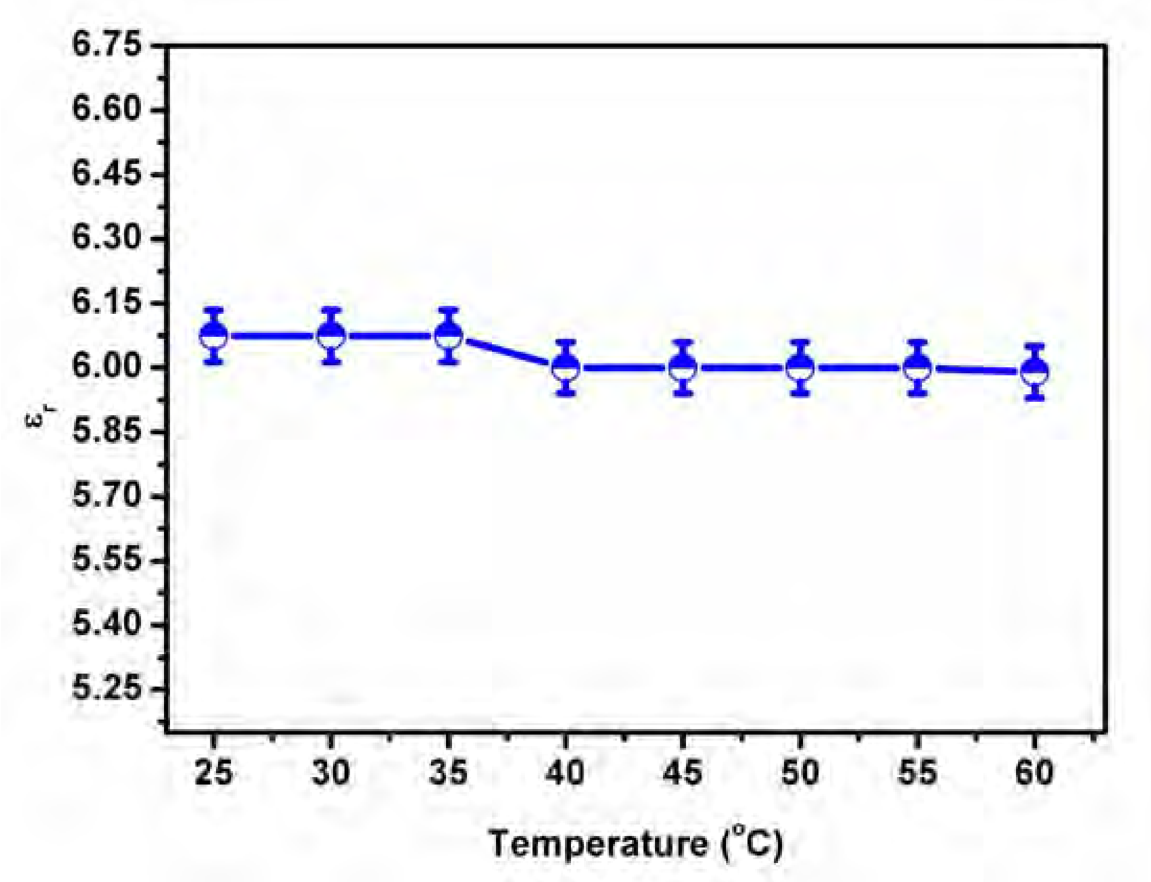

Fig.4. Variation of ε_r with temperature at 5 GHz for Al_2O_3-NaCl composite cold sintered at 120°C.

Highlight of the work

- ✓ Alumina-NaCl prepared by cold sintering processat120^{o}C with 96 % densification.
- ✓ Cold sintering process is cost effective, simple and energy saving
- ✓ The Al_2O_3-NaCl composite has good microwave dielectric properties.
- ✓ Al_2O_3-NaCl composite has low dielectric constant and low dielectric loss.
- ✓ Useful in microwave communication systems with high signal speed.