Microwave dielectric properties of mineral sillimanite obtained by conventional and cold sintering process

I.J.Induja, M.T.Sebastian

Abstract

The sillimanite (Al2SiO5) mineral has been sintered by conventional ceramic route and by cold sintering methods. The mineral has very poor sinterability and transformed to mullite on sintering above 1525°C.The dielectric properties of sillimanite mineral (Al_2SiO_5) are investigated at radio and microwave frequency ranges. The mineral sintered at 1525°C has low ε_r of 4.71 and tanδ of 0.002 at 1 MHz and at microwave frequency ε_r = 4.43, Q_u× f = 41,800 GHz with τ_f = −17 ppm/°C. The sintering aid used for cold sintering Al_2SiO_5 is sodium chloride (NaCl). The Al_2SiO_5NaCl composite was cold sintered at 120°C. XRD analysis of the composite revealed that there is no additional phase apart from Al_2SiO_5 and NaCl. The densification of the Al_2SiO_5NaCl composite was confirmed by using microstructure analysis. The Al_2SiO_5NaCl composite has ε_r of 5.37 and tanδ of 0.005 at 1 MHz whereas at microwave frequency it has ε_r = 4.52, Qu× f = 22,350 GHz with τ_f = −24 ppm/°C. The cold sintered NaCl has ε_r = 5.2, Qu× f = 12,000 GHz with τ_f = −36 ppm/°C.

1. Introduction

Microwave dielectric materials conquer today's wireless communication industry by providing a wide variety of applications such as broadcasting satellites, cellular phone, global positioning systems etc. [1]. New microwave substrate materials with low relative permittivity and high performance are desired for high speed communication devices [2]. For microwave substrate applications the dielectric materials should possess low dielectric constant (ε_r), low loss(tanδ), low temperature coefficient of permittivity (τ_ε), high thermal conductivity and low coefficient of thermal expansion [3]. Low ε_r materials offer the advantage of fast signal propagation [3,4]. Sillimanite (Al_2SiO_5) is a naturally occurring alumino silicate polymorph which is obtained as a by product during the extraction of rare earths from beach sand minerals [5,6]. The most well-known alumino-silicate polymorphsare sillimanite (Al_2SiO_5), andalusite (Al_2SiO_5) and kyanite (Al_2SiO_5) [7,8]. In 1928, Taylor first resolved the crystal structure of sillimanite while the cell dimensions and space group was determined by Mark and Rosband in 1926 [9,10].Sillimanite decomposes into mullite (Al6Si2O13) and cristobalite (SiO2) on heating between 1500°Cand 1650°C [11,12]. The decomposition temperature may differ depending on the region of its occurrence. Sillimanite and mullite are compounds of alumino silicates with Si-Al tetrahedral chains[13]. The main applications of Al_2SiO_5 are in the fabrication of high tension insulators, spark plugs, glass furnaces, combustion chambers etc [14].

Recently Kahari et al. reported [15,16] a novel method toprepare Li_2MoO_4 ceramics at room temperature by moistening the water soluble Li_2MoO_4 powder using deionized water and pressing the sample at about 130 MPa. The samples were then dried at room temperature or at 120°C. There was no appreciable difference in the sintered density of the samples dried at room temperature, dried at 120°C or sintered at 540°C. The amount of water in the pressed samples was about 2–3 wt% before drying or sintering. It is believed that the densification of the samples occurred during pressing. X-ray diffraction study showed that the crystal structure remains the same and water did not react to form any hydrates. The room temperature dried samples showed a Q_u × f value slightly less than that prepared by conventional sintering method and is attributed to the presence of small amount of residual water. It was found [16] that ceramic powder particle size has great influence on preparation; densification and dielectric properties. Larger particles are

advantageous in fabricating Li_2MoO_4 ceramics by moistening and pressing method. Smaller particles lead to clay like clusters leading to non-uniform densification, warpage and cracking. Kahari et al. [17] tailored the dielectric properties by adding TiO_2 and $BaTiO_3$ in Li_2MoO_4 with optimized room temperature preparation method. More recently Randall and co-workers have done extensive work [18–28] on room temperature or sintering below 200°C by the same technique and introduced the term cold sintering process (CSP). CSP is primarily based on the solid particle rearrangement with the help of liquid phase followed by densification by dissolution-precipitation [19]. The temperature, pressure, the particle size of the ceramic powder, amount of solvent added are the key factors that control the CSP process [19]. CSP technique has been utilized for achieving dense ceramic, ceramic-polymer composites at temperature less than 200°C. Simplicity as well as energy saving are the attractive features of CSP process [15,23]. Owing to the low temperature sintering, CSP method offers the advantage of integrating polymers with ceramics. The CSP method was successfully applied to NaCl, alkali molybdates, KH_2PO_4, $NaNO_2$, zirconia, ZnO, $BaTiO_3$ and V_2O_5 [22,25–28]. The possibility of incorporating ceramics with polymers in single step sintering using the emerging CSP method for the study of Li_2MoO_4–(-CF_2-)n(PTFE), electrolyte $Li_{1.5}Al_{0.5}Ge_{1.5}(PO_4)_3$(-$CH_2CF_2$)x[$CH_2CF(CF_3)$y(PVDF-HFP) and semiconductor V_2O_5–poly(3,4-ethylenedioxythiophene)polystyrene sulfonate(PEDOT:PSS) composites was also exploited [20]. Baker et al. fabricated monolithic capacitor using lithium molybdenum oxide ceramic material by CSP method [24]. In the present paper we report the microwave dielectric properties of sillimanite mineral sintered by the conventional solid state method and by cold sintering process using NaCl as sintering aid.

2. Experimental

The mineral sillimanite obtained from Indian Rare Earths Lim-ited (IRE) was planetary ball milled for 6 h using distilled water medium in order to reduce its particle size. The average particle size of the Al_2SiO_5 after ball milling was found out using dynamic lights cattering instrument (Malvern Zetasizer, Nano-ZS, UK). The slurrywas dried at 100°C. The dried Al_2SiO_5 powder was ground well and polyvinyl alcohol (PVA) was added prior to pelletizing the bulk sample. The bulk Al_2SiO_5 was sintered at 1525°C and the structural, micro structural analysis and dielectric studies were carried out. Sodium chloride (NaCl) from SDFCL, Mumbai, India was used for the present study. NaCl was moistened using deionized water and then transferred into suitable die set and hot pressed at 120°C by applying a pressure of 200 MPa. After several trial and errors, a 1:1 ratio (in weight% (wt%)) was maintained between Al_2SiO_5 and NaCl for cold sintering Al_2SiO_5. For preparing the composite containing Al_2SiO_5 − NaCl, first NaCl was moistened with deionized water and Al_2SiO_5 was added into it. The mixture was thoroughly mixed using deionized water to make a paste. In the present work, 4 wt% deionized water was added to make the paste of Al_2SiO_5– NaCl depending upon the weight of the composite taken. The paste was in semi solid form. The Al_2SiO_5− NaCl composite was then hot pressed using die set at a temperature of about 120°C (50 min) and pressure 200 MPa. In order to remove the moisture content, the cold sintered NaCl and Al_2SiO_5−NaCl composite was kept in hot air oven at 120°C for 24 h. The phase composition of bulk Al_2SiO_5, Al_2SiO_5−NaCl was studied using XRD (CuKα radiation, PANalyticalX'Pert PRO diffractometer, Netherlands). The room temperature FT-IR spectrum of cold sintered Al_2SiO_5NaCl was recorded using Agilent Technologies, 600ATR, UK using the KBr pellet method. The microstructure of the fractured bulk Al_2SiO_5sintered using the conventional high temperature sintering method and Al_2SiO_5NaCl cold sintered at120°C was recorded using scanning electron microscopy (JOEL-JSM5600LV, Tokyo, Japan and Zeiss, Germany). The density of the bulk Al_2SiO_5, cold sintered NaCl and Al_2SiO_5− NaCl 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 radio frequency dielectric properties were measured by parallel plate capacitor method using an LCR meter (Hioki 3532-50 LCR Hi Tester, Japan). For radio frequency measurements of bulk Al_2SiO_5, cold sintered NaCl and the Al_2SiO_5− NaCl composite samples having 11 mm diameter and 1.5 mm thickness coated with silver paste on both sides were used. The microwave dielectric properties of bulk Al_2SiO_5, cold sintered NaCl and Al_2SiO_5− NaCl composite having

diameter 11 mm and 5.4 mm thickness was analyzed using Hakki − Coleman method using a vector network analyzer (Agilent E5071C ENA series, AgilentTechnologies, Santa Clara, CA) in the operating range 300 kHz–20 GHz. The temperature coefficient of resonant frequency (τ_f) of bulk Al_2SiO_5, cold sintered NaCl and Al_2SiO_5−NaCl composite were measured using the cavity setup in the temperature range 25°C–60°C for every 5° rises in temperature.

3. Results and discussions

Sillimanite is a naturally occurring mineral with orthorhombic structure having Pbnm space group symmetry. The average particle size of the Al_2SiO_5 powder was about 1 µm. Fig. 1 (a) shows the XRD pattern of sillimanite (Al_2SiO_5) sintered at 1525°C and Fig. 1(b) shows the XRD pattern of the bulk sintered at 1550°C. The peaks of Al_2SiO_5 are identified and indexed using JCPDS file no.22 − 0018, mullite ($Al_6Si_2O_{13}$) using 15 − 0776 and the peaks of cristobalite (SiO_2) is identified and indexed using JCPDS file no. 85 − 0621. Asevident from the XRD; the maximum temperature that the mineral sillimanite can be sintered is limited up to 1525°C. It may be noted that sillimanite mineral often contains a small amount of quartz (SiO_2) which on heat treatment at high temperatures changes into cristobalite form. At 1550°C, sillimanite decomposes into mullite ($Al_6Si_2O_{13}$) and excess silica as glass or cristobalite according to the reaction given in (1) [29]. Even though the crystal structure of sillimanite and mullite are identical (both are orthorhombic) and further more the diffraction patterns of both of them are also similar. However, there are some characteristic diffraction peaks that distinct them [6].

$$3Al_2SiO_5 \rightarrow Al_6Si_2O_{13} + SiO_2 \quad (1)$$

(sillimanite) (mullite) (cristobalite)

The microstructure of the fractured surface of sillimanite (Al_2SiO_5) sintered using the conventional solid state ceramic method at 1525°C is shown in Fig. 1(c). The grains of Al_2SiO_5areclearly visible in the microstructure. The microstructure depicts the fact that sillimanite (Al_2SiO_5) consists of particles of polygonal shape having non-uniform size. The poor densification of the bulk Al_2SiO_5 is also evident from the microstructure. The variation of the radio frequency dielectric properties of bulk Al_2SiO_5 sintered using the conventional ceramic route at 1525°Cis shown in Fig. 2. The ε_r and tanδ at 1 MHz of bulk sillimanite (Al_2SiO_5) sintered at 1525°C is 4.71 and 0.002 respectively. The error in dielectric properties measurement at the radio frequency for ε_r and tanδ is 1%.The mineral Al_2SiO_5 possess low dielectric constant and low loss at 1 MHz. The sillimanite (Al_2SiO_5) mineral is difficult to densify by sintering at 1525°C and further increase in temperature convert the sillimanite to mullite. The sillimanite sintered at 1525°C has a density of 2 g/cm^3. However, the theoretical density of sillimanite is 3.1 g/cm^3 (calculated from the XRD data in the present study). The mineral sillimanite contains small amount of different impurity phases/substitutions and hence there can be differences in the theoretical as well as experimental densities of samples from different localities. In the present case, the evaluation of percentage density calculation is difficult due to presence of quartz (SiO_2) as secondary phase in the raw sillimanite. Hence the sillimanite is densified by cold sintering process using NaCl. The XRD pattern of sillimanite-sodium chloride (Al_2SiO_5– NaCl) composites cold sintered at 120°C is shown in Fig. 3. The peaks of sillimanite (Al_2SiO_5) (JCPDS file no. 22-0018), NaCl (JCPDSfileno.77-2064), quartz (SiO_2) (JCPDS file no.89-8934) are identified and indexed in the XRD. It should be noted that there is no reaction between Al_2SiO_5 and NaCl by the cold sintering. The microstructure of the fractured surface of Al_2SiO_5– NaCl composite cold sintered at 120°C is shown in Fig. 4. The densification of Al_2SiO_5 ceramics with the aid of NaCl is evident from the microstructure. Fig. 4 (a) shows the microstructure of fractured surface and Fig. 4 (b) is the backscattered image of Al_2SiO_5– NaCl composite cold sintered at 120°C. The SEM pictures do not show additional secondary phases other than

Al_2SiO_5 and NaCl. Here the densification of Al_2SiO_5 is promoted by the grain growth with the aid of pressure and temperature.

The room temperature FT-IR spectrum of Al_2SiO_5– NaCl composite is shown in Fig. 5. No peaks corresponding to water is identified in the FT-IR spectrum, which indicates that there is no residual water in the cold sintered sample. The peak around 1177 cm^{-1} corresponds to the characteristic peak of sillimanite which is assigned to the Al-O stretching vibrations [6]. The band position at 891 cm^{-1} is attributed to the symmetric stretching of Si − O bond, 808 cm^{-1} corresponds to the antisymmetric bending, deformation of Si-O-Si and 697 cm^{-1} anti-symmetric, bending, deformation Si O Si; Al1 translations [30,31]

The variation of dielectric properties of cold sintered NaCl at the radio frequency in the range 10 kHz −1 MHz is shown in Fig. 6(a). The dielectric constant and dielectric loss at 1 MHz for NaCl is 5.88 and 0.009 respectively. Fig. 6 (b) shows the radio frequency dielectric properties of Al_2SiO_5– NaCl composite cold sintered at 120°C.The ε_r and tanδ of the Al_2SiO_5– NaCl composite is 5.37 and 0.005 respectively. The addition of NaCl influences the dielectric properties of the Al_2SiO_5. The water solubility nature of the sintering aid NaCl used for the present study enables dissolution – precipitation which is a pre-requisite condition for the cold sintering process with the help of sufficient temperature and pressure.

The microwave dielectric properties of bulk Al_2SiO_5 sintered at1525°C, NaCl and Al_2SiO_5– NaCl cold sintered at 120°C is given in Table 1. The mineral Al_2SiO_5 possess low dielectric constant with excellent $Q_u \times f$ value (41,800) GHz. The temperature coefficient of resonant frequency of Al_2SiO_5 is obtained as −17 ppm/°C. The dielectric properties of the mineral indicate the possibility of use in microwave applications. The low dielectric constant of sillimanite provides the advantage of fast signal propagation when used for practical applications [32]. The NaCl sintering aid in the present work cold sintered at 120°C has ε_r= 5.22 (∼15.5 GHz) with $Q_u \times f$ = 12, 000 GHz and τ_f= −36 ppm/°C. The Al_2SiO_5– NaCl composite has ε_r of 4.52 with $Q_u \times f$ = 22,350 GHz with τ_f= −24 ppm/°C. Compared to the bulk Al_2SiO_5, the $Q_u \times f$ value is found to be decreased with the addition of NaCl. But the addition of NaCl, the densification of the Al_2SiO_5 is improved by employing the CSP process. The value of τ_f which is a measure of drift of resonant frequency with respect to temperature and low negative value in the present case indicates that the composite can be fruitfully used for microwave applications. Since NaCl is a hygroscopic material, for practical applications the composite can be coated or encapsulated with low loss plastic materials or silicone rubber. According to the rule of mixtures,

$$\varepsilon_c = V_1\varepsilon_1 + V_2\varepsilon_2 \quad (2)$$

where ε_c is the dielectric constant of the Al_2SiO_5– NaCl composite, V_1 and V_2 are the volume fractions of Al_2SiO_5 and NaCl, ε_1 and ε_2 are the corresponding dielectric constants. The dielectric constant obtained from the theoretical calculation of dielectric constant using the rule of mixtures is found to be 4.63 which are very close to the experimental value (see Table 1).

5. Conclusions

The feasibility of the cold sintering process in the densification of the naturally occurring mineral Al_2SiO_5 is successfully demonstrated in the present work. Compared to the conventional ceramic route, Al_2SiO_5 can be easily densified using the CSP. The structural analysis reveals the fact that there is no impurity phase in the composite Al_2SiO_5– NaCl composite. The cold sintered-composite has ε_r = 5.37 and tanδ of the order of 10^{-3} at 1 MHz. The microwave dielectric properties of the composite are ε_r = 4.52, $Q_u \times f$ = 22,350 GHz with τ_f= −24 ppm/°C.

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References

[1] K. Zhang, L. Yuan, Y. Fu, C. Yuan, W. Li, Microwave dielectric properties ofAl2O3ceramics co-doped with MgO and Nb2O5, J. Mater. Sci. Mater. Electron.26 (2015) 6526–6531.

[2] K.M. Manu, C. Karthik, L.C. Leu, K.A. Lazar, R. Ubic, M.T. Sebastian, Crystalstructure and microwave dielectric properties of LiRE9(SiO4)6O2ceramics(RE = La, Pr Nd, Sm, Eu, Gd, and Er), J. Am. Ceram. Soc. 96 (2013) 1504–1511.

[3] M.T. Sebastian, H. Jantunen, Low loss dielectric materials for LTCCapplications: a review, Int. Mater. Rev. 53 (2008) 57–90.

[4] H. Ohsato, M. Terada, I. Kagomiya, K. Kawamura, K.I. Kakimoto, E.S. Kim,Sintering conditions of cordierite for Microwave/Millimeter wave dielectrics,IEEE Trans. Ultrason. Ferroelectr. Freq.Control 55 (2008) 1081–1085.

[5] S. Yugeswaran, M. Vijay, K. Suresh, P.V. Ananthapadmanabhan, Z. Karoly, J.Szepvolgyi, Synthesis of mullite from sillimanite dissociation throughtransferred arc plasma torch, Int. J. Miner. Process. 99 (2011) 54–60.

[6] A. Tomba, M.A. Camerucci, G. Urretavizcaya, A.L. Cavalieri, M.A. Sainz, A.Caballero, Elongatedmullite crystals obtained from high temperaturetransformation of sillimanite, Ceram. Int. 25 (1999) 245–252.

[7] J.K. Winter, S. Ghose, Thermal expansion and high-temperature crystalchemistry of the Al2SiO5polymorphs, Am. Mineral. 64 (1979) 573–586.

[8] H. Yang, R.M. Hazen, L.W. Finger, C.T. Prewitt, R.T. Downs, Compressibility andcrystal structure of sillimanite Al2SiO5at high pressure, Phys. Chem. Miner.25 (1997) 39–47.

[9] W.H. Taylor, The structure of sillimanite and mullite, Z. Kristallogr 71 (1928)205–218.

[10] W.A. Deer, R.A. Howie, J. Zussman, Rock-Forming Minerals: Orthosilicates,Vol. 1A, The Geological Society, 1997.

[11] S. Tripathi, G. Banerjee, Synthesis and mechanical properties of mullite frombeach sand sillimanite: effect of TiO2, J. Eur. Ceram. Soc. 18 (1998) 2081–2087.

[12] S.S. Srikant, S.K. Singh, P.S. Mukherjee, R.B. Rao, Value addition to redsediment placer sillimanite using microwave energy and in depth structuraland morphological characterization of mullite, J. Miner. Mater. Charact. Eng.11 (2012) 1055–1062.

[13] J.P. Hirth, L. Kubin, Dislocations in Solids, The 30th Anniversary Volume,Elsevier, UK, 2010.

[14] S. Aryal, P. Rulis, W.Y. Ching, Density functional calculations of the electronicstructure and optical properties of aluminosilicate polymorphs (Al2SiO5), Am.Miner. 93 (2008) 114–123.

[15] H. Kahari, M. Teirikangas, J. Juuti, H. Jantunen, Dielectric properties of lithiummolybdate ceramic fabricated at room temperature, J. Am. Ceram. Soc. 97(2014) 3378–3379.

[16] H. Kahari, M. Teirikangas, J. Juuti, H. Jantunen, Improvements andmodifications to room temperature fabrication method for dielectricLi2MoO4 ceramics, J. Am. Ceram. Soc. 98 (2015) 687–689.

[17] H. Kahari, M. Teirikangas, J. Juuti, H. Jantunen, Room-temperature fabricationof microwave dielectric Li2MoO4-TiO2composite ceramics, Ceram. Int. 42(2016) 11442–11446.

[18] C.A. Randall, J. Guo, H. Guo, A. Baker, M.T. Lanagan, Cold Sintering Ceramicsand Composites, US Provisional Patent Application, (2015) 62/234 389.

[19] H. Guo, J. Guo, A. Baker, C.A. Randall, Hydrothermal-assisted cold sinteringprocess: a new guidance for low-temperature ceramic sintering, ACS Appl.Mater. Interfaces 8 (2016) 20909–20915.

[20] J. Guo, S.S. Berbano, H. Guo, A.L. Baker, M.T. Lanagan, C.A. Randall, Coldsintering process of composites: bridging the processing temperature gap ofceramic and PolymerMaterials, Adv. Funct. Mater. 26 (2016) 7115–7121.

[21] N.J. Lóh, L. Simão, C.A. Faller, A. De Noni Jr., O.R.K. Montedo, Review article. Areview of two-step sintering for ceramics, Ceram. Int. 42 (2016) 12556–12572.

[22] J. Guo, H. Guo, A.L. Baker, M.T. Lanagan, E.R. Kupp, G.L. Messing, C.A. Randall,Cold sintering: a paradigm shift for processing and integration of ceramics,Angew. Chem. Int. Ed. 55 (2016) 11457–11461.

[23] J. Guo, S.S. Berbano, H. Guo, A.L. Baker, M.T. Lanagan, C.A. Randall, Coldsintering process of composites: bridging the processing temperature gap ofceramic and polymer materials, Adv. Funct. Mater. 26 (2016) 7115–7121.

[24] A. Baker, H. Guo, H.J. Guo, C. Randall, Utilizing the cold sintering process forflexible- printable electroceramic device fabrication, J. Am. Ceram. Soc. 99(2016) 3202–3204.

[25] H. Guo, A. Baker, J. Guo, C.A. Randall, Cold sintering process: a novel techniquefor low-temperature ceramic processing of ferroelectrics, J. Am. Ceram. Soc.(2016), http://dx.doi.org/10.1111/jace.14554.

[26] H. Guo, J. Guo, A. Baker, C.A. Randall, Hydrothermal-assisted cold sinteringprocess: a new guidance for low-temperature ceramic sintering, ACS Appl.Mater. Interfaces 8 (2016) 20909–20915.

[27] H. Guo, J. Guo, A. Baker, C.A. Randall, Cold sintering process for ZrO2-basedceramics: significantly enhanced densification evolution in yttria-dopedZrO2, J. Am. Ceram. Soc. (2016), http://dx.doi.org/10.1111/jace.14593.

[28] S. Funahashi, J. Guo, H. Guo, K. Wang, A.L. Baker, K. Shiratsuyu, C.A. Randall,Demonstration of the cold sintering process study for the densification andgrain growth of ZnO ceramics, J. Am. Ceram. Soc. (2016), http://dx.doi.org/10.1111/jace.14617.

[29] P.M. Ihlen, Utilisation of sillimanite minerals, their geology, and potentialoccurrences in Norway −an overview, NGU-Bull. 436 (2000) 113–128.

[30] M. Lodzinski, R. Wrzalik, M. Sitarz, Micro-Raman spectroscopy studies ofsome accessory minerals from pegmatites of the SowieMts andStrzegom-Sobótka massif, J. Mol. Struct. 744–747 (2005) 1017–1026.

[31] K. Iishi, E. Salje, C. Werneke, Phonon spectra and rigid-ion model calculationson andalusite, Phys. Chem. Miner. 4 (1979) 73–188.

[32] M.T. Sebastian, Dielectric Materials for Wireless Communication, Elsevier,Oxford, 2008.

Table and Figures

Table 1. Microwave dielectric properties of bulk Al_2SiO_5 sintered at 1525 °C, NaCl and Al_2SiO_5– NaCl composite cold sintered at 120 °C.

Material	ε_r ± 2% (Hakki)	($Q_u \times f$) ± 2% (GHz) (Cavity)	τ_f ± 2% (ppm/°C)	Density ± 1% (g/cm^3)
Al_2SiO_5 (Sintered at 1525 °C)	4.43(~16 GHz)	41,800	-17	2.00
NaCl (CSP at 120 °C)	5.22(~15.5 GHz)	12,000	-36	2.10
Al_2SiO_5–NaCl (CSP at 120 °C)	4.52 (~17 GHz)	22,350	-24	2.39

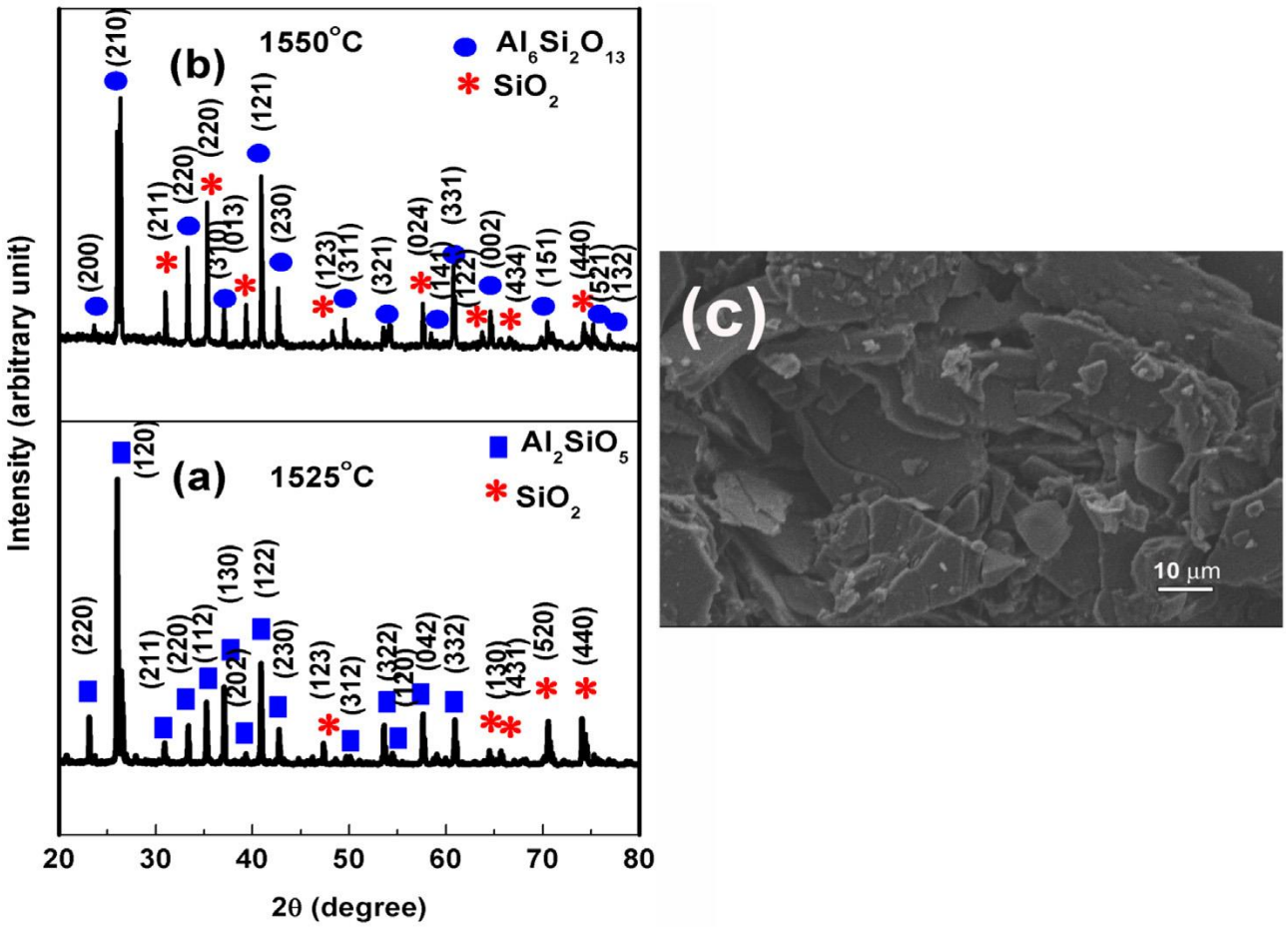

Fig. 1. (a) XRD pattern of bulk sillimanite sintered at (a) 1525°C and (b) 1550°C and (c) microstructure of fractured surface of sillimanite sintered at 1525°C using the conventional solid state ceramic method.

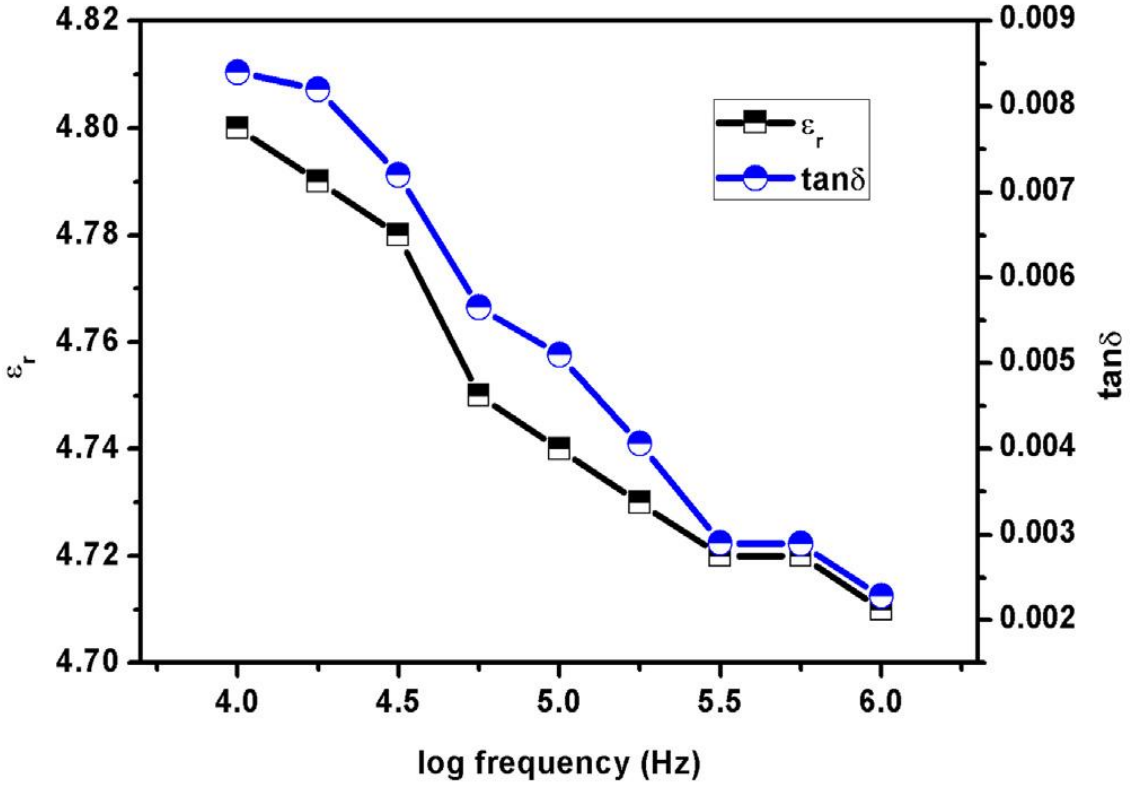

Fig. 2. Variation of dielectric properties with radio frequency of bulk Al_2SiO_5 sintered using conventional ceramic route at 1525°C.

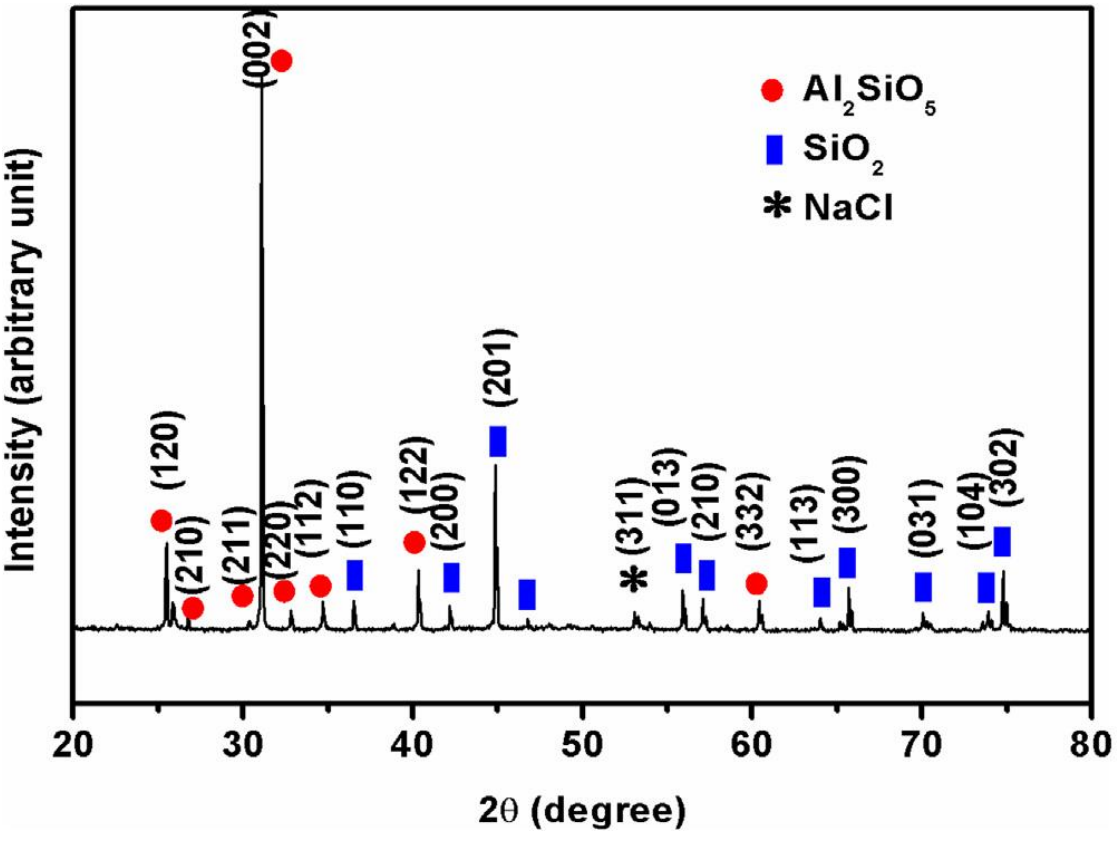

Fig. 3. XRD pattern of Al_2SiO_5– NaCl composite cold sintered at 120°C.

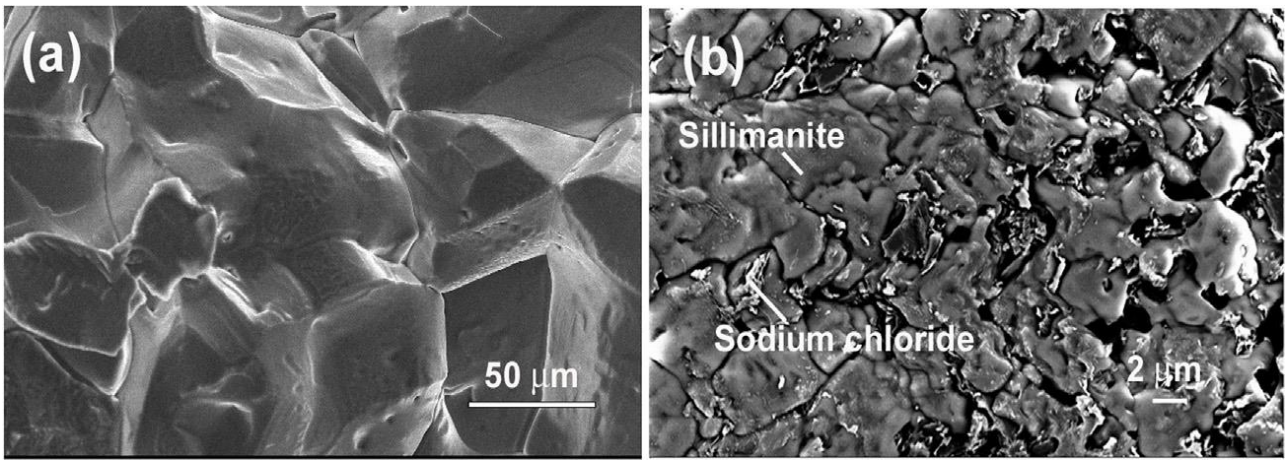

Fig. 4. (a) microstructure of the fractured surface and (b) backscattered image of Al_2SiO_5-NaCl cold sintered at 120°C.

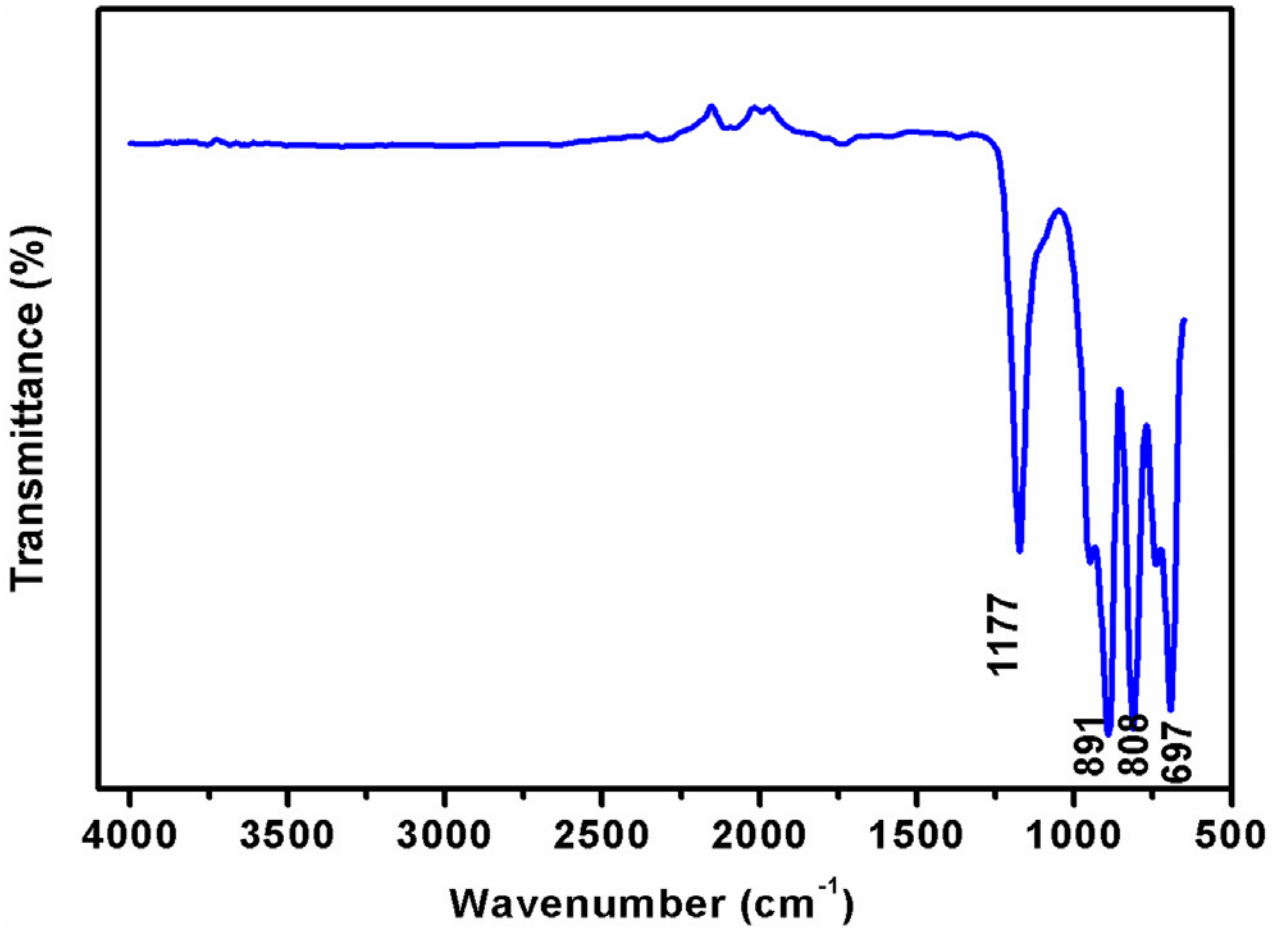

Fig. 5. Room temperature FT-IR spectrum of Al_2SiO_5– NaCl composite cold sintered at 120°C.

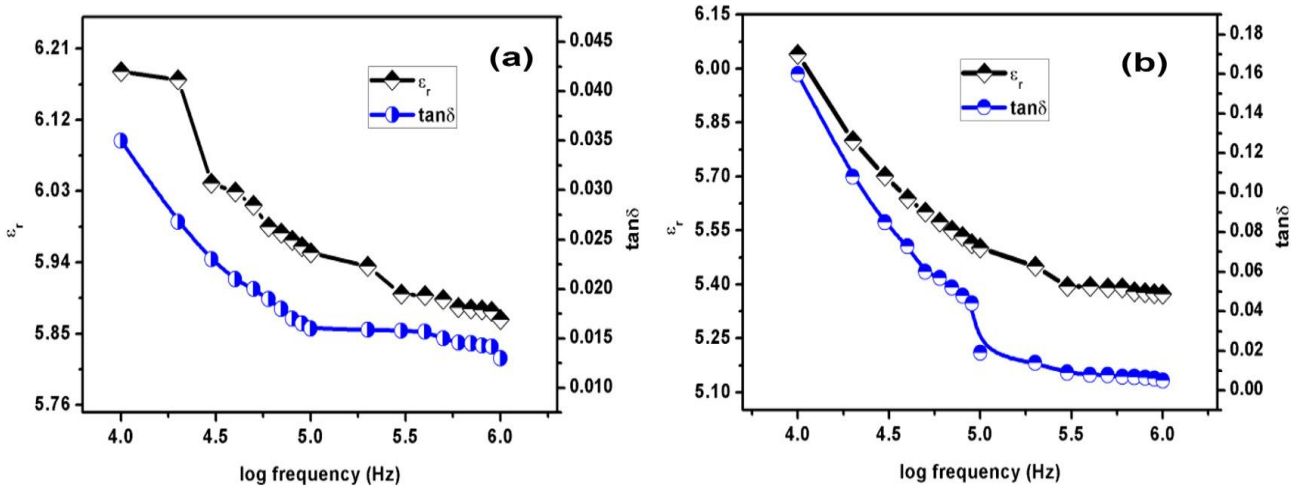

Fig. 6. Variation of dielectric properties with radio frequency of (a) pure NaCl (b) Al_2SiO_5– NaCl composite cold sintered at 120°C.