PREFACE

Recently, microwave telecommunication has been developed for a wide range of applications, such as mobile phone, wireless LAN and Intelligent Transport System (ITS). Utilized microwave frequency has also increased from microwave to millimeter range in order to transmit large quantity of information with high speed. Microwave dielectric materials are continuing to play a very important role in the microwave communication systems. As a result, a large number of ceramic dielectric materials have been developed for use as dielectric resonators, capacitors, substrates and electronic packages. The key material requirements for microwave dielectric materials to be used for high frequency applications are: optimum relative permittivity, high quality factor and near-zero temperature coefficient of resonant frequency. The development of low-temperature co-fired ceramics (LTCC) has been stimulated by the benefits offered for the fabrication of miniature multilayer devices and for high level of passive integration involving the co-firing of dielectric and highly conductive metals, such as silver and copper. The present thesis entitled “SILICATE AND ALUMINATE BASED DIELECTRIC CERAMICS FOR MICROWAVE COMMUNICATION” is the outcome of a detailed investigation performed on the synthesis, characterization and microwave dielectric properties of some novel rare earth based silicates and aluminate. Accordingly the thesis is divided into seven chapters.

The first chapter gives a general introduction on low loss microwave dielectric ceramics and its importance in various areas of interest such as dielectric resonators, low temperature cofired ceramics, substrates and electronic packaging. A brief discussion about the relevance of polymer ceramic composites in microelectronic industry is also included. The major scientific and technological aspects, material requirements and applications of the dielectric ceramics are also given.
Chapter 2 presents the details of the preparation techniques adopted for the synthesis of dielectric resonators, LTCC materials and polymer ceramic composites. A brief description about the various characterization techniques used for the structural, microstructural and dielectric characterization in the purview of this thesis is also presented.

Chapter 3 gives a general idea on the importance of silicate materials for microwave applications and throws light into a new group of materials \( A{\text{RE}}_4{\text{Si}}_3{\text{O}}_{13} \) [\( A=\text{Ca, Sr and Ba; RE=\text{rare earths}} \)] belonging to the ‘apatite’ family for high frequency applications. The synthesizing conditions of these materials are optimized for the best properties. The structural investigations using XRD refinements and TEM revealed that they belong to \( P6_3/m \) space group with hexagonal symmetry. The materials exhibited a relative density less than 94\% which was improved to certain extent by the addition of a small amount of glass. The microwave dielectric properties of these materials are reported for the first time. The sintered ceramics has a relative permittivity \( (\varepsilon_r) \) less than about 20 and quality factor \( Q_u \times f \) up to 30000 GHz. The high \( \tau_f \) value \((-46 \text{ ppm/}^\circ\text{C})\) of \( \text{SrLa}_4\text{Si}_3\text{O}_{13} \) is tuned to a near zero value by the addition of suitable amount of \( \text{TiO}_2 \). The mixture rules are used to calculate the density, \( \varepsilon_r \) and \( \tau_f \) of these ceramics and the theoretical values agree well with the experimental ones.

Chapter 4 highlights the synthesis, characterization and microwave dielectric properties of two novel rare earth based silicates [\( \text{Sm}_2\text{Si}_2\text{O}_7 \) and \( \text{RE}_2\text{Ti}_2\text{SiO}_9 \) (\( \text{RE=La, Pr and Nd} \))] ceramics. The \( \text{Sm}_2\text{Si}_2\text{O}_7 \) ceramics has a tetragonal symmetry whereas \( \text{RE}_2\text{Ti}_2\text{SiO}_9 \) (\( \text{RE=La, Pr and Nd} \)) dielectric ceramics possess a monoclinic symmetry. The \( \text{Sm}_2\text{Si}_2\text{O}_7 \) ceramics sintered at 1375\(^\circ\text{C}/2\text{h}\) exhibit excellent dielectric properties: \( \varepsilon_r = 10 \) and \( \tan \delta = 0.006 \) measured at 9 GHz. The effect of various low loss glass addition on the sintering, densification and microwave dielectric properties of \( \text{Sm}_2\text{Si}_2\text{O}_7 \) is
studied. The $\text{Sm}_2\text{Si}_2\text{O}_7$ ceramics mixed with 15 wt% LBS glass lowered the sintering temperature to 975°C, whereas the 15 wt% LMZBS glass addition lowered sintering temperature of 950°C. The pure $\text{Sm}_2\text{Si}_2\text{O}_7$ ceramics and that mixed with 15 wt% LMZBS glass did not show much variation in relative permittivity with temperature. The $\text{RE}_2\text{Ti}_2\text{SiO}_9$ ($\text{RE}=$La, Pr and Nd) ceramics exhibited a relative permittivity less than 20 and relatively low $\tau_f$ value. A maximum value of $Q_u \times f$ of 33500 GHz is shown by $\text{Pr}_2\text{Ti}_2\text{SiO}_9$ ceramics. It is seen that Pr substitution for La favored the formation of solid solution in the whole range obeying Vegard’s law while Nd substitution resulted in the formation of additional phases. As the Pr content increases, an improvement in the quality factor is noted whereas the $\tau_f$ value is not much affected.

Chapter 5 discusses in detail the synthesis and characterization of various polymer ceramic composite using $\text{Sm}_2\text{Si}_2\text{O}_7$ ceramics as filler for electronic packaging applications. The polymers used in the investigation are PTFE, Polyethylene and Polystyrene. The dielectric, thermal and mechanical properties of these polymer-ceramic composites has been investigated. The effects of coupling agent and filler particle size on the above properties of PTFE/$\text{Sm}_2\text{Si}_2\text{O}_7$ composites are also studied. For a filler loading of 0.5 $\nu_f$, PTFE composite has $\varepsilon_r = 3.82$ and tan $\delta = 0.0136$ (at 9 GHz), $k_c = 1.76$ W/m°C, $\alpha_c = 36$ ppm/°C, Vickers’ microhardness of 13 kgf/mm²; PE composite has $\varepsilon_r = 5.28$ and tan $\delta = 0.0091$ (at 9 GHz), $k_c = 2.97$ W/m°C, $\alpha_c = 60$ ppm/°C, Vickers’ microhardness of 17 kgf/mm² and PS composite has $\varepsilon_r = 4.60$ and tan $\delta = 0.0110$ (at 9 GHz), $k_c = 0.29$ W/m°C, $\alpha_c = 36$ ppm/°C, Vickers’ microhardness of 56 kgf/mm². Several theoretical model approaches have been employed to predict the relative permittivity, thermal conductivity and coefficient of linear expansion of the composite systems and the results were compared with that of
experimental data. All theoretical predictions were found to be valid for low filler contents.

Chapter 6 outlines the applicability of $0.83 \text{ ZnAl}_2\text{O}_4\cdot0.17 \text{ TiO}_2$ (ZAT) dielectric ceramic based glass and polymer composites for LTCC substrate and electronic packaging applications respectively. The ZAT dielectric ceramic possess excellent thermal and microwave dielectric properties but with a high sintering temperature of $1450^\circ C$. The first section of the Chapter 6 discusses the efforts taken to reduce the sintering temperature by glass addition for use as LTCC substrate material. Among the various glasses added, BBSZ is found to lower the sintering temperature without much affecting the microwave dielectric properties. The XRD and SEM of BBSZ glass added ZAT composites suggested the existence of no additional secondary phases. The addition of 10 wt% BBSZ glass reduced the sintering temperature to $950^\circ C$ with reasonably good microwave dielectric properties. The composites also possessed high chemical compatibility with silver. The results bring out the possibility of using ZAT/10 wt% BBSZ composites for LTCC substrate applications. The second section of this chapter compares the physical, dielectric and thermal properties of ZAT loaded PTFE and PE composites. The PE/ZAT composites possessed a high relative density when compared with the PTFE/ZAT composites. For lower filler content the fillers are uniformly dispersed in the matrix and as the filler content increased the agglomeration also increased and resulted in porosity. The dielectric properties of both the composites showed that ZAT loaded with PE composites exhibited good dielectric properties and also a very low water absorption value of less than 0.1%. The results show that ZAT filler loaded composites have better properties than that loaded with $\text{Sm}_2\text{Si}_2\text{O}_7$ filler and can be used for electronic packaging applications.

The seventh chapter gives the conclusion of the thesis and scope for future work.