This chapter provides a brief account of ceramic-polymer composites for electronic applications and various theoretical models used for predicting permittivity of the composites. The chapter presents preparation and dielectric properties of SCT filled \((n=0)\) PTFE, Polyethylene and Polystyrene. Different theoretical models were employed for effectively predicting the relative permittivity of the composites.
5.1 INTRODUCTION

Cost effective product miniaturization has been long demanding goals of electronic industries\(^1\,2\,3\,4\). Even though a good number of materials are available for microwave substrate application, the ever-increasing sophistication, packaging density and signal speed limits the use of conventional materials\(^1\,4\,5\,6\). The growth of telecommunication systems and high-speed digital devices stimulated the mankind to develop new materials, which can deliver improved performance. There are a number of ceramic materials with excellent dielectric properties and thermal properties. However, the process ability of the ceramics is poor, as they are brittle. But the excellent mechanical and low processing temperature of some of the polymers make them very good candidate for microwave substrate applications. The use of such low dielectric loss polymers is also limited by their high thermal expansion, low relative permittivity and low thermal conductivity. For an ideal substrate material it should have low dielectric loss, optimum relative permittivity, high thermal conductivity, matching coefficient of thermal expansion to that of Silicon and good mechanical properties\(^1\,3\). In a composite material, the individual properties of polymers and ceramics can be combined. Hence ceramic polymer composites can offer excellent properties for microwave substrate and electronic packaging applications. The relative permittivity of the composites can easily be tailored by the volume fraction, size and shape of low loss ceramics.

There are a number of low loss polymers that can be used for synthesizing composites for electronic applications\(^7\). The dielectric properties of the polymer used in the present investigation are given in Table 5.1. PTFE based composites have already
established as microwave substrates because of their excellent dielectric and thermal properties\textsuperscript{8,9,10,11,12}. However, composites involving other low loss, low cost polymers such as polyethylene and polystyrene are still to be explored in detail. The dielectric loss tangents of all polymers used in the present study are excellent for microwave applications.

Table 5.1 Microwave dielectric properties of the polymers used in the present investigation at 8 GHz.

<table>
<thead>
<tr>
<th>Polymer</th>
<th>Permittivity</th>
<th>Loss tangent</th>
</tr>
</thead>
<tbody>
<tr>
<td>PTFE</td>
<td>1.9</td>
<td>&lt;10\textsuperscript{-3}</td>
</tr>
<tr>
<td>Polyethylene (HDPE)</td>
<td>2.3</td>
<td>&lt;10\textsuperscript{-3}</td>
</tr>
<tr>
<td>Polystyrene</td>
<td>2.1</td>
<td>&lt;10\textsuperscript{-4}</td>
</tr>
</tbody>
</table>

Another major problem in the fabrication of a composite with desired properties is the selection of suitable filler material. Metal powders of Ag and Al are commonly used for improving the permittivity of polymer composite. However, relatively higher dielectric loss of these composites prevents their application as microwave substrates. Hence high permittivity low dielectric loss ceramic fillers are advantageous since using such fillers the permittivity can be tailored to greater extent and also the amount of filler ceramics can be reduced below 40 volume percentage for achieving $\varepsilon_r$ of 10 to 15. However, as the permittivity increases the loss tangent also increases and the number of low loss materials with $\varepsilon_r > 100$ are also limited. Some reports are available regarding BaTiO\textsubscript{3}\textsuperscript{13,14,15} SrTiO\textsubscript{3}\textsuperscript{16,17,18} and CaTiO\textsubscript{3}\textsuperscript{19} based polymer-ceramic composites. However, the higher dielectric loss of such composites precludes their immediate application. Several novel high permittivity low loss materials belonging to Sr\textsubscript{2}+\textsubscript{n}Ce\textsubscript{2}Ti\textsubscript{5+n}O\textsubscript{15+3n} (n=0 to 10) ceramics were presented in Chapter 3. These materials have permittivity in the range of 113 to 185
and low loss tangent of $< 10^{-4}$. Among these, $\text{Sr}_2\text{Ce}_2\text{Ti}_5\text{O}_{15}$ ($\varepsilon_r = 113$ and $\tan \delta = 3 \times 10^{-4}$) ceramic material was selected for preparing the composites.

5.2 THEORETICAL MODELING

The dielectric properties of disordered heterogeneous materials have received considerable fundamental attention since nearly the time of Maxwell and these investigations continue to be centrally important for developing a deeper understanding leading to a wide variety of innovative technological applications in many branches of engineering\textsuperscript{20}. Although, experiments and simulations are designed to probe the dielectric behavior of composite materials, the prediction of dielectric properties of these media remains an outstanding problem in materials physics. Formulas for the permittivity of heterostructures were originally discussed by Maxwell and Faraday in the middle of the nineteenth century as part of their comprehensive studies of electricity and magnetism. Later on Clausious (1897), Maxwell- Garnet\textsuperscript{21} (1904) discovered new relations for isotropic dielectric mixtures. In the early 20\textsuperscript{th} century Lichteneker\textsuperscript{22} derived a famous logarithmic relation for dielectric composites. Logarithmic relation has been widely used to fit permittivity of various composites. Effective medium approximation was first introduced by Bruggemen\textsuperscript{23} in 1935. A twist to the story developed in the year of 1957 when percolation theory was introduced by Broadbent and Hammersley\textsuperscript{24}. The availability of high performance computers made the problem much more interesting and several other models were developed.
The precise prediction of permittivity of a composite material is very important for the design of packaging materials and substrates. Several quantitative rules and simulation techniques have been proposed for the prediction of permittivity. However, very little experimental evidence was provided to support the rule of mixtures. In the present study the following equations were used to calculate the effective permittivity of the various composites.

\[ \ln \varepsilon_{\text{eff}} = f \ln \varepsilon_i + (1-f) \ln \varepsilon_m \quad \text{Lichteneker Eqn. (5.1)} \]

\[ \frac{\varepsilon_{\text{eff}} - \varepsilon_m}{\varepsilon_{\text{eff}} + 2\varepsilon_m} = f \frac{\varepsilon_i - \varepsilon_m}{\varepsilon_i + 2\varepsilon_m} \quad \text{Maxwell Garnet Eqn. (5.2)} \]

\[ \varepsilon_{\text{eff}} = \frac{\varepsilon_m (1-f) + \varepsilon_f \left[ \frac{3\varepsilon_m}{\varepsilon_i + 2\varepsilon_m} + \frac{3f(\varepsilon_i - \varepsilon_m)}{(\varepsilon_i + 2\varepsilon_m)} \right]}{(1-f) + f \left[ \frac{3\varepsilon_m}{\varepsilon_i + 2\varepsilon_m} + \frac{3f(\varepsilon_i - \varepsilon_m)}{(\varepsilon_i + 2\varepsilon_m)} \right]} \quad \text{Smith Eqn. (5.3)} \]

\[ \varepsilon_{\text{eff}} = \varepsilon_m \left[ 1 + \frac{f(\varepsilon_i - \varepsilon_m)}{\varepsilon_m + n(1-f)(\varepsilon_i - \varepsilon_m)} \right] \quad \text{EMT Model (5.4)} \]
where $\varepsilon_{\text{eff}}$, $\varepsilon_i$, and $\varepsilon_m$ are the permittivity of the composites, filler, matrix respectively, $f$ is the volume fraction of the ceramic and $n$ is the fitting parameter. Among these equations, Lichteneker equation is the most commonly used relation for predicting the permittivity of the composites. Smith and Jayasundere\textsuperscript{26} derived an equation for predicting permittivity of 0-3 composites assuming spherical fillers. In this case a bi-phase system consisting of dielectric spheres with permittivity $\varepsilon_i$ is dispersed in continuous medium of permittivity $\varepsilon_m$.

Effective medium analysis assumes that each constituent is surrounded by the same effective medium. It assumes that the local electric and magnetic fields are the same in the volume occupied by each component in the composite. The analysis is done in the approximation of non-interacting inclusions (each inclusion is subject to the same mean-field, unperturbed by the presence of other inclusions). The accuracy and range of validity of EMT is not easy to establish. In order to predict the permittivity of the composites precisely, Rao et al.\textsuperscript{34} developed the Effective Medium Theory (EMT) model. In the EMT model, composites can be treated as an effective medium whose permittivity is obtained by averaging over the permittivity of the constituents. The schematic representation of the microstructure of polymer ceramic composites is shown in Fig. 5.1 (a). In order to account for the major features of a composite microstructure, a random unit cell is defined as core of $\varepsilon_i$ surrounded by a shell of host matrix $\varepsilon_m$ as shown in Fig. 5.1 (b) [where $\varepsilon_i$, $\varepsilon_m$ and $\varepsilon_{\text{eff}}$ are the permittivity of ceramics, matrix and composites respectively]. The basic concept of effective medium is that when a random unit cell is embedded in an effective medium it cannot be detected in the electromagnetic experiment\textsuperscript{34}. Otherwise, the random unit cell can be replaced by a material characterized by $\varepsilon_{\text{eff}}$. The EMT model leads to the equation.
(5.4) to predict the effective permittivity of the ceramic-polymer composites which also includes a fitting parameter $n$.

![Fig. 5.1 Schematic diagram of (a) microstructure of composite materials and (b) Random unit cell embedded in the effective medium.](image)

5.3 EXPERIMENTAL PROCEDURE

$\text{Sr}_2\text{Ce}_2\text{Ti}_5\text{O}_{15}$ ceramics were prepared by the solid-state ceramic route. $\text{Sr}_2\text{Ce}_2\text{Ti}_5\text{O}_{15}/\text{PTFE}$ (Hindustan Fluorocarbons, Hyderabad, India) composites were prepared by powder processing technology. A detailed description of the experimental procedure is described in chapter 2, section 2.3.1. Polystyrene and polyethylene based composites were prepared by sigma blending technique or melt mixing technique as described in chapter 2, section 2.3.2. The composites were characterized by X-ray diffraction technique using CuKα radiation (Philips X-Ray Diffractometer). The surface morphology of the composites was studied by scanning electron microscope (JEOL-JSM 5600 LV, Tokyo, Japan). The low frequency dielectric properties were measured by LCR meter (Hioki 3532-50). Electroded samples having diameter of about 14 mm and
thickness 1 mm were used for this measurement. HP 8510 C Vector Network Analyzer was used to characterize the composite materials at microwave frequencies. For this purpose rectangular samples of dimensions (40x3x2 mm³) were made and the dielectric properties were measured using cavity perturbation technique. The micromechanical properties of polystyrene/SCT composites were measured using micro hardness tester (Clemex 4, Germany). Both the surfaces of the samples were polished to have optically flat surface for indentation. The specimen was subjected to a load of 50 g and dwell time of 10 s. For pure ceramic sample the load was increased to 500 g. A total of 5 readings were taken to get average hardness.

5.4 PTFE-Sr₂Ce₂Ti₅O₁₅ COMPOSITES

The PTFE-SCT composites were prepared by powder processing technology. The measured of density by Archimedes method shows a decrease in relative density from 85 to 75 % with the increase in filler content. Hence samples with higher filler content show poor densification. This can be due to the increased void formation with filler content. Fig 5.2 gives the XRD pattern of PTFE and its Sr₂Ce₂Ti₅O₁₅ filled composites. The pattern of PTFE [Fig. 5.2 (a)] shows a strong crystalline peak superimposed over an amorphous halo as reported³⁵. Fig. 5.2 (b) and (c) show the XRD patterns of PTFE-Sr₂Ce₂Ti₅O₁₅ composites with ceramic volume percentage 30 and 60 respectively. The XRD peaks corresponding to pseudo-cubic Sr₂Ce₂Ti₅O₁₅ were indexed. Fig. 5.3 shows the SEM pictures of pure SCT ceramic (used in the present investigation) and PTFE-Sr₂Ce₂Ti₅O₁₅
composites with different volume fractions. Irregularly shaped particles with average size less than 10 µm is visible from figure 5.3 (a). It can be seen that Sr₂Ce₂Ti₅O₁₅ particles are well dispersed in the PTFE matrix (see Fig. 5.3 (b)), however at higher mixing ratios agglomeration of ceramic particles were observed [See Fig. 5.3 (c)]. Hence for higher mixing ratios the connectivity among the ceramic particles increases which in turn improves the properties.

Fig. 5.2 XRD pattern of (a) PTFE (b) PTFE + 30 volume percentage and (c) 60 volume percentage of SCT composites.
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Fig. 5.3 SEM micrographs of (a) Sr$_2$Ce$_2$Ti$_5$O$_{15}$ powders and PTFE-Sr$_2$Ce$_2$Ti$_5$O$_{15}$ composites containing (b) 10, (c) 30 and (d) 60 volume percentage of the ceramics.

The surface energy of PTFE is relatively low and so it is difficult for PTFE to have good adhesion with ceramics. In such cases the use of coupling agents will improve filler matrix interfacial strength. Fig. 5.4 shows the fractograph of 10 volume percentage PTFE-Sr$_2$Ce$_2$Ti$_5$O$_{15}$ composites. The presence of PTFE fibrils in the fractured surface is an indication of better adhesion between ceramics and PTFE$^{36}$.

Figs. 5.5 (a) and (b) show the variation of permittivity and dielectric loss at 1 MHz and 8 GHz respectively. The permittivity and dielectric loss of the composites increase with the increase in Sr$_2$Ce$_2$Ti$_5$O$_{15}$ content. The increase in permittivity is expected as the ceramic have high $\varepsilon_r$ compared to that of PTFE matrix. Also at higher mixing ratios the
connectivity (see Fig. 5.3 c) among the ceramic particles increases which in turn increases the permittivity. The connectivity of the individual components has maximum importance as it controls the electric flux pattern as well as mechanical and thermal properties. The permittivity and loss tangent of the composites are affected by a number of factors such as porosity, moisture content and interface between the two components in the composite. The dielectric loss of the PTFE-Sr$_2$Ce$_2$Ti$_5$O$_{15}$ composites increases with increase in ceramic content. As the volume fraction of ceramic increases, the interface between PTFE and Sr$_2$Ce$_2$Ti$_5$O$_{15}$ increases. Hence the increase in dielectric loss with mixing ratio is attributed to the increased interfacial polarization at higher volume fractions of the PTFE-Sr$_2$Ce$_2$Ti$_5$O$_{15}$ composites.

Fig. 5.4 SEM fractograph of PTFE-Sr$_2$Ce$_2$Ti$_5$O$_{15}$ composites containing 10 volume percentage of ceramics.
Fig. 5.5 Variation in permittivity and loss tangent of PTFE-Sr$_2$Ce$_2$Ti$_5$O$_{15}$ composites at (a) 1 MHz and (b) 8 GHz.

Fig. 5.6 (a) and (b) depict the comparison of effective permittivity of the composites at 1 MHz and 8 GHz with the values predicted by equations described in the section 5.2. Lichtenecker’s and Jayasundere’s mixing rules fit well with the experimental
data at lower mixing ratios (up to 0.3\textit{vf}). At higher mixing ratios the predicted $\varepsilon_r$ values are much higher than the experimental. Maxwell-Garnet relations predict lower $\varepsilon_r$ compared to the experimental data. Hence Lichtenecker's, Jayasundere's and Maxwell-Garnet relations cannot be used to predict the $\varepsilon_r$ of PTFE-Sr$_2$Ce$_2$Ti$_5$O$_{15}$ composites. The permittivity of the composites not only depends on the individual dielectric properties of the components but also on the shape and size of the ceramic particle, porosity, interface between components and on the homogeneity of distribution. It will be difficult to include all these parameters in calculations. In EMT model, $n$ is the correction factor to compensate for the shape of the fillers used in polymer ceramic composites. The value of $n$ is determined empirically which is found to be 0.165 for the PTFE-Sr$_2$Ce$_2$Ti$_5$O$_{15}$ composites. A small value of $n$ shows that the filler particles have nearly spherical shape\textsuperscript{38}. At 1 MHz, measured and predicted $\varepsilon_r$ are nearly equal up to 30 volume percentage of Sr$_2$Ce$_2$Ti$_5$O$_{15}$ ceramics. However, as volume fraction of the ceramics increases beyond 0.3\textit{vf} a slight deviation from the predicted value of $\varepsilon_r$ is observed in PTFE-Sr$_2$Ce$_2$Ti$_5$O$_{15}$ composites. At 8 GHz the measured $\varepsilon_r$ is in good agreement with the EMT model. Hence it can be concluded that EMT model can be used to predict the effective permittivity of the PTFE-Sr$_2$Ce$_2$Ti$_5$O$_{15}$ composites. Even though relative permittivity of the PTFE is improved, the dielectric loss tangent increased considerably. The increase in loss tangent can be attributed to the increased porosity of the composite. Hence the higher dielectric loss of the composites precludes their immediate practical applications.
Fig. 5.6 Comparison of experimental and theoretical permittivity of PTFE-Sr$_2$Ce$_2$Ti$_5$O$_{15}$ composites at (a) 1 MHz (b) 8 GHz.

5.5 POLYSTYRENE, POLYETHYLENE-Sr$_2$Ce$_2$Ti$_5$O$_{15}$ COMPOSITES

One of the major disadvantages of PTFE based composites prepared by powder processing route described in the earlier section is the higher dielectric loss caused by poor
densification of the composites. This can be improved by changing the preparation technique. Another method used for making polymer-ceramic composite is the melt mixing or sigma blending technique. However, in order to fabricate PTFE based composite high temperature (>250 °C) is required, also PTFE has very high viscosity compared to other thermoplastic polymers like polyethylene or polystyrene. Both polyethylene and polystyrene showed excellent dielectric properties. Hence for the sigma blending purpose polystyrene and polyethylene were selected. Polystyrene is a low dielectric loss polymer having low processing temperature. Some reports\textsuperscript{39,40,41} are available in the literature investigating the dielectric and thermal properties of polystyrene-AlN composites. However, the low relative permittivity of the AlN ceramics prevents better tailoring of the 
\textit{\varepsilon}_r. Khastigir \textit{et al.}\textsuperscript{41} reported the dielectric properties TiO\textsubscript{2}-polystyrene composites. But the dielectric properties of these composites were not investigated at microwave frequencies. Polyethylene is also a well known polymeric insulating material. It has got high dielectric strength, low dielectric loss and good mechanical properties. Even though there are a number of reports\textsuperscript{42,43} showing the dielectric properties of metal filled polyethylene, properties of the low loss ceramic filled polyethylene is less investigated. Hence properties of Sr\textsubscript{2}Ce\textsubscript{2}Ti\textsubscript{5}O\textsubscript{15} ceramic loaded polyethylene and polystyrene were presented. These composites were prepared by melt mixing method as described in the chapter 2.

5.5.1 Results and Discussion

Fig. 5.7 (a) and (b) shows the variation in experimental and theoretical densities of polystyrene-SCT and polyethylene-SCT composites respectively with respect to the filler volume. The matrix and the filler have densities of 1.038 g/cm\textsuperscript{3} (polystyrene) 0.95 g/cm\textsuperscript{3}
(polyethylene) and 5.15 g/cm³ (SCT). It is found that as the filler loading increases the experimental density follows the theoretical one. However, the difference between experimental and theoretical densities increases marginally with respect to ceramic content. This can be due the increased porosity with filler content. This variation is more prominent in PE-SCT composites. The percentage densification of PS-SCT composites varied from 97 to 98.5 while that of PE-SCT composites varied from 93 to 97 percentages.

Fig. 5.7 Comparison of experimental and theoretical densities of (a) PS-SCT (b) PE-SCT composites.
Fig. 5.8 (a) shows the DSC thermo gram of pure polystyrene and 40 volume percentage PS-SCT composite. It can be noted that PS exhibits endothermic peak approximately at 100 °C corresponding to the glass transition temperature.

![Diagram](a)

![Diagram](b)

Fig. 5.8 Heating and cooling DSC curves of (a) pure and 40 volume percentage ceramic loaded PS-SCT composites (b) 40 volume percentage SCT loaded polyethylene.
A very broad peak is also observed for 40 volume percentage PS-SCT composites at about 100 °C confirming no significant change in glass transition temperature with filler loading. The DSC thermogram of 40 volume percentage ceramic loaded HDPE is shown in Fig. 5. 8 (b). In the PE-SCT composite there has been no change in melting point of polyethylene is observed with filler content.

Fig. 5.9 (a) and (b) shows surface morphology of 40 volume percentage SCT loaded polystyrene and polyethylene respectively. Although the ceramic particles are dispersed in the polymer matrix occasional agglomeration can be observed. It can be confirmed from the figure that the polystyrene covers the ceramic better than polyethylene. Fig. 5.9 (c) and (d) shows the fracto-graphs of 40 volume percentage of SCT loaded PS and PE respectively. The fractograph clearly shows ceramic particles are loosely bound to the polyethylene matrix compared to polystyrene. This can be due to the poor adhesion between the ceramic and polyethylene. The observed percentage densification of the PS-SCT composites (97 to 98.5 %) can be well correlated with its microstructures. However, in the case of PE-SCT composite some porosity is visible in the fractograph which corresponds to its low densification compared to PS-SCT composites.

Micro hardness of the composites gives additional information on micromechanical properties with respect to filler loading. Micro indentation technique has been increasingly used in characterizing homopolymers and polymer composites. The variation in micro hardness with filler loading for PS-SCT and PE-SCT composites is shown in Fig. 5.10. The hot pressed and well-polished polystyrene has average Vickers’s hardness of 21.5 Kg/mm² compared to 8 Kg/mm² of pure HDPE. It can be noted that as the ceramic content
increases the hardness also increases. More than 50% increase in hardness is observed for 50 volume percentage PS-SCT and PE-SCT composites. However, it is immaterial to compare with the micro hardness of 865 obtained for the sintered and dense SCT for a load of 500g. Mechanical properties of the PS-SCT composites are better compared to that of PE/SCT composites.

Table 5.2 shows the dielectric properties of PS-SCT and PE-SCT composites measured at 1 MHz. The relative permittivity and loss tangent of the PS-SCT composites increase with filler loading. It is well known that both permittivity and loss tangent depend
on electronic, ionic, orientation and space charge polarizations. The space charge polarization arises because of heterogeneity in the sample and it decreases with frequency. SCT is polycrystalline while PS and PE is amorphous and non polar. The increase in \( \varepsilon_r \) and dielectric loss of PS-SCT and PE-SCT composites with filler content can be attributed to the increased interfacial polarization.

A maximum \( \varepsilon_r \) of 14 and loss tangent of 0.00696 is observed for 50 volume percentage loading in PS-SCT composite. In the case of PE-SCT composite for 50 volume percentage loading the permittivity and loss tangent are 13, 0.0081 respectively at 1 MHz.
Table 5.2 Dielectric properties of PS-SCT and PE-SCT composites at 1 MHz.

<table>
<thead>
<tr>
<th>Volume percentage of SCT</th>
<th>(\varepsilon_r) PS-SCT</th>
<th>(\tan\delta) PS-SCT</th>
<th>(\varepsilon_r) PE-SCT</th>
<th>(\tan\delta) PE-SCT</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>3.3</td>
<td>0.00177</td>
<td>2.6</td>
<td>0.00163</td>
</tr>
<tr>
<td>10</td>
<td>3.9</td>
<td>0.00268</td>
<td>3.6</td>
<td>0.00252</td>
</tr>
<tr>
<td>20</td>
<td>5.4</td>
<td>0.00379</td>
<td>5.</td>
<td>0.00323</td>
</tr>
<tr>
<td>30</td>
<td>8.2</td>
<td>0.00449</td>
<td>7.3</td>
<td>0.00468</td>
</tr>
<tr>
<td>40</td>
<td>9.6</td>
<td>0.0053</td>
<td>9.5</td>
<td>0.00680</td>
</tr>
<tr>
<td>50</td>
<td>14</td>
<td>0.00696</td>
<td>13</td>
<td>0.00810</td>
</tr>
</tbody>
</table>

Fig. 5.11 shows the variation in microwave dielectric properties of PS-SCT and PE-SCT composites with filler volume at 8 GHz. The dielectric properties at microwave frequencies also follow the path of that obtained at radio frequencies. As the volume of percentage of the ceramic increases, the loss tangent as well as \(\varepsilon_r\) increases. For most of the polymers the dielectric loss at microwave frequencies consists of background loss or high and low frequency tails outside this region\(^7\). Hence the increase in dielectric loss and permittivity is due to the properties of the SCT filler. It is worth to note that the order of loss tangent is nearly the same for all volume fractions of the PS-SCT composites. However, the dielectric loss of the PE-SCT composite is relatively higher. For practical applications it is always better to consider composites with ceramic loading less than 40 volume percentage so that flexibility of the polymer is partially retained. For 40 volume percentage loading, relative permittivity \(\sim 10\) with a dielectric loss tangent of \(3 \times 10^{-4}\) and 0.005 is observed for of PS-SCT and PE-SCT composites respectively.

The comparison of experimental and theoretical permittivity of the PS-SCT and PE-SCT composite is shown in Fig. 5.12. The most commonly used relation is Lichteneker's logarithmic law of mixing. However, this relation overestimated the
permittivity values of PS-SCT composites at higher volume fractions. In the case of PE-
SCT composite a slight variation is also observed at higher volume percentage. Smith and
Jayasundere\textsuperscript{26} derived an equation for predicting permittivity of 0-3 composites assuming
spherical fillers. But Smith’s equation cannot predict the permittivity values of PS-SCT
composites precisely.

![Graph](image)

**Fig. 5.11** Variation of permittivity and dielectric loss for (a) PS-SCT and (b) PE-SCT
composites at 8 GHz.
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This can be due to the fact that the used fillers are not spherical. Maxwell Garnet equation also gives considerably lower estimate for the permittivity in both PS-SCT and PE-SCT composites.

The measured permittivity shows deviation from the predicted values for almost all theoretical models especially in PS-SCT composites. This can be due to the various factors, which affect the permittivity of the composites other than filler volume and $\varepsilon_i$. The permittivity of composites also depends on the homogeneity of distribution of the filler, shape and size of fillers and inter-face between ceramics and polymers. It will be a difficult task to include all the above parameters in to single equation. However, Rao et al.\textsuperscript{34} introduced effective medium theory (EMT model) to calculate the permittivity of the composites. The EMT model uses a ceramic morphology fitting factor $n$, which is determined by curve fitting. The fitting factor $n$ is subjected to the ceramic material only. In the PS-SCT and PE-SCT composites the permittivity predicted by EMT model matches with experimental values except for 50 volume percentage of the ceramic. Among the several models tried, EMT model gives a reasonably good fit for the permittivity PS-SCT and PE-SCT composites. The obtained values of $n$ are 0.17 and 0.15 for PS-SCT and PE-SCT composite respectively. The values of $n$ are in good agreement with that of PTFE-SCT composites ($n= 0.165$). When the ceramic loading increases (>0.4 vt) almost all model loses their validity. This can be due to the imperfect dispersing of the ceramic particles at higher filler content. Among the four models tried in general, EMT model gives a better fitting for the relative permittivity of the SCT based composites.
5.6 CONCLUSIONS

PTFE-Sr$_2$Ce$_2$Ti$_5$O$_{15}$ composites are prepared by powder processing method. The dielectric properties of the composites were studied at 1 MHz and 8 GHz. As the volume
fraction of $\text{Sr}_2\text{Ce}_2\text{Ti}_5\text{O}_{15}$ increases the permittivity and dielectric loss increases. The higher
dielectric loss of the PTFE composite at higher ceramic loading is due to poor
densification of the composite. Polystyrene-SCT and polyethylene-SCT composites were
prepared by melt mixing technique. The DSC of the composites shows no significant
change in the glass transition temperature with filler volume. Micro hardness test shows
improvement in mechanical properties with filler loading. The permittivity and the loss
tangent of the PS-SCT and PE-SCT composites increase with filler content. Both PS-SCT
and PE-SCT composites shows excellent dielectric properties at microwave frequency. It is
found that EMT model is effective in predicting permittivity of PTFE-SCT, PE-SCT, PS-
SCT composites. PS-SCT and PE-SCT composites can be made with permittivity ranging
from 3 to 14 having stable dielectric loss less than $10^{-3}$. Hence these composites can be
considered for microwave substrates applications.
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