Microwave devices such as low noise oscillators, filters, patch antennas, solid state amplifiers etc. use ceramic filled Poly(tetraflouroethylene) (PTFE) substrates as the basic building block. Dielectric constant, loss tangent, dielectric thickness and temperature stability of the base substrate are of primary importance during microwave circuit design. Miniaturization of modern wireless communication systems requires novel materials with high dielectric constant and low loss tangent for the transmission of electrical signals at microwave frequencies. The circuit size can be reduced by increasing the dielectric constant of the base substrate since the wavelength of electromagnetic signal traveling through a medium is inversely proportional to square root of its dielectric constant. On the other hand, low loss tangent is essential for minimum signal attenuation together with better signal integrity to avoid cross talking. Another critical parameter is the stability of the above mentioned parameters with respect to temperature. If the dielectric constant of base substrate changes with respect to temperature, as a result, the operating frequency of the device will also vary.

Owing to its extremely low loss tangent and high service temperature, ceramic filled PTFE based laminates are the ideal choice for circuit fabrication. The relative dielectric constant ($\varepsilon_r$) of PTFE is stable over a wide range of frequencies since the molecular structure lacks dipole groups capable of resonant response. The essential features required for base substrate application are isotropic dielectric properties, good dimensional stability, bare minimum porosity, low moisture absorption, high heat dissipation etc. Even though PTFE has excellent dielectric properties, it suffers from high coefficient of thermal expansion, poor dimensional stability and low heat dissipation. Often, PTFE is filled with ceramic particulates to circumvent these deleterious properties. However, achieving uniform filler distribution through conventional polymer processing techniques is often difficult in PTFE composites due to its high melt viscosity ($10^{11}$ Pa.s.). Ordinary polymer processing techniques such as injection molding, blow molding, solvent casting etc. can not be applied in the case of PTFE. Hence, there is a perceived need to develop a novel processing
methodology to fabricate filled PTFE composite substrates with nearly isotropic dielectric properties.

First chapter of the thesis gives a detailed introduction to microwave materials, especially on planar microwave substrates. A detailed description about different classes of base materials used for microwave circuit design such as hard and soft substrates are outlined in this chapter. The advantages of soft substrates over hard microwave substrates have been discussed. Soft substrates are explained in detail with examples. In addition to microwave substrates, dielectric resonators, microwave phase shifters and microwave absorbers are also explained in this chapter. Different polarization mechanisms responsible for giving rise to dielectric properties at microwave frequency region are also explained at length. A brief discussion on the role of dielectric properties of the base material for circuit design and various packaging techniques are also given in chapter 1. Different characterization techniques and detailed specification of instruments used in the present study are described in this chapter. A detailed review of the published literature related to microwave substrates is also compiled. The basic objectives of the present work are also clearly explained in this chapter.

In the second chapter, preparation of filled PTFE composites using rutile as the particulate filler has been discussed. Structural and dielectric properties of the initial raw materials such as PTFE, rutile and microfibre glass are explained in detail. Rutile filler has been treated with silane coupling agent to preclude its moisture absorption characteristic. Three different processing methodologies were attempted to make filled PTFE composites. Sigma Mixing, Preforming followed by Sintering (SMPS process), Sigma Mixing followed by Hot pressing (SMH process) and Sigma Mixing, Extrusion, Calendering followed by Hot pressing (SMECH process) techniques were attempted to make PTFE composites. Among these, SMECH process is observed to be better suited to prepare uniformly filled planar PTFE composite substrates with bare minimum porosity. Hence, SMECH process has been used to fabricate ceramic filled PTFE substrates in the other chapters also. An optimum filler loading has been found out in PTFE matrix for reproducible microwave dielectric properties. Dielectric constant and loss tangent at microwave frequency region (X-band) of the composites were studied by waveguide cavity perturbation technique using Vector Network
Analyzer. A non-monotonic variation is observed in the dielectric constant of the composite with respect to filler loading. Dielectric constant shows better matching with that predicted using Lichtenecker model. Morphology of the composites was studied to evaluate the nature of filler distribution and also to ascertain the efficacy of SMECH process. The ultimate tensile strength of the composite has been studied as per ASTM D 638 standards and moisture absorption characteristics of samples were studied as per IPC- TM-650 2.6.2 standards. The coefficient of thermal expansion of filled PTFE composites has been studied as a function of filler loading. PTFE/rutile composite shows a dielectric constant of 10.2 with loss tangent 0.0022 at X-band region, which can be used for commercial microwave substrate applications. In order to study the effect of particle size on the microwave dielectric properties of filled PTFE composites, nano size rutile powder has also been filled in the PTFE matrix.

In chapter 3, ceramics having higher dielectric constant than rutile are filled in the PTFE matrix with an objective to improve the effective dielectric properties of the composite systems. The particulate fillers such as SrTiO$_3$ and CaTiO$_3$ were prepared through solid state ceramic preparation route. The phase formation, morphology and particle size of the fillers were studied using powder X-ray diffraction, Laser Raman, SEM and particle size analyses. PTFE/SrTiO$_3$ and PTFE/CaTiO$_3$ composites have been prepared through SMECH process. Optimum filler loading in both composites were found out. Microwave dielectric, mechanical and thermal properties of filled composites have been evaluated with respect to filler loading. The dielectric constant of both PTFE/SrTiO$_3$ and PTFE/CaTiO$_3$ composites show matching with values predicted using Lichtenecker model. Dielectric constant of 13.1 and 11.9 were obtained for SrTiO$_3$ and CaTiO$_3$ filled PTFE composites at optimum filler loading of 63 and 61 wt% respectively. The composites show comparatively higher loss tangent than rutile filled PTFE composites. The moisture absorption characteristics of these composites are also on the higher side.

Chapter 4 explains the preparation of high dielectric and low loss ceramic fillers and filled PTFE composites made out them. Sr$_2$Ti$_9$O$_{20}$ and Ca$_2$Ti$_9$O$_{20}$ fillers were prepared through conventional solid state ceramic route. Phase formation was studied using powder X-ray diffraction and Laser Raman analyses. The microstructure of the sintered ceramic pellets
was studied using SEM techniques. The dielectric properties of sintered \( \text{Sr}_2\text{Ti}_9\text{O}_{20} \) and \( \text{Ca}_2\text{Ti}_9\text{O}_{20} \) ceramics were evaluated at microwave frequency region using Hakki and Coleman post resonator technique. X-ray diffraction studies show that \( \text{Sr}_2\text{Ti}_9\text{O}_{20} \) is multiphase in nature consisting of rutile and \( \text{SrTiO}_3 \) where as \( \text{Ca}_2\text{Ti}_9\text{O}_{20} \) appeared to be a mixture of rutile and \( \text{CaTiO}_3 \). PTFE/\( \text{Sr}_2\text{Ti}_9\text{O}_{20} \) and PTFE/\( \text{Ca}_2\text{Ti}_9\text{O}_{20} \) composites were prepared through SMECH process. Microwave dielectric properties, moisture absorption characteristics, tensile properties and linear coefficient of thermal expansion were studied as a function of filler loading. At optimum filler loading, PTFE/\( \text{Sr}_2\text{Ti}_9\text{O}_{20} \) and PTFE/\( \text{Ca}_2\text{Ti}_9\text{O}_{20} \) composites show a dielectric constant of 14.4 and 12.9 respectively with loss tangent less than 0.0041. These composites have better mechanical strength together with low moisture absorption content. The CTE of the composites reduces to \(<20\ \text{ppm/}^\circ\text{C} \) at optimum filler loading which is comparable to that of copper conducting layer. In order to study the effect of multiphase nature of the filler in the dielectric properties of the composites, ceramics were calcined at three different temperatures and filled in the PTFE matrix through SMECH process. It is observed that, no difference in the effective dielectric constant of the composite is observed with respect to calcination temperature. SEM studies reveal that the fillers are uniformly distributed in the PTFE matrix without much porosity.

Fifth chapter discusses the preparation of high dielectric and temperature stable PTFE/ceramic composite substrates for outdoor wireless applications. Two approaches were attempted to obtain temperature stable substrates. In the first approach, fillers having different temperature coefficient of dielectric constant have been incorporated in the PTFE matrix very close to the optimum loading condition through SMECH process. Fillers having relatively high negative temperature coefficient of dielectric constant \( (\tau_{\varepsilon r}=-300\ \text{ppm/}^\circ\text{C}) \) result in composites with low positive \( \tau_{\varepsilon r} \). For near zero temperature coefficient of dielectric constant \( (\tau_{\varepsilon r}=+30\ \text{ppm/}^\circ\text{C}) \) fillers, the composite exhibited relatively high positive temperature coefficient of dielectric constant. It is observed that by judiciously selecting the filler with suitable \( \tau_{\varepsilon r} \) and its loading fraction, temperature stable substrates can be realized in PTFE composite systems. In the second approach discussed in chapter 5, a secondary polymer is added to PTFE/ceramic composite system. Thermoplastic poly ether ether ketone
(PEEK) having nearly same processing temperature was added to rutile loaded PTFE composites. At 8 wt% PEEK addition, the $\tau_{\varepsilon}$ of -40ppm/°C was achieved.

A general conclusion derived from the results obtained in chapter 2, 3, 4 and 5 are compiled and the scope for future work is discussed in chapter 6.

**Patent filed**

1. High permittivity low loss temperature stable flexible microwave laminates
   Indian Patent 2008 (submitted)

**Research papers published**


3. Preparation and characterization of high permittivity and low loss PTFE/CaTiO$_3$ microwave laminates. Polymer Composites 2008 (available online)


Papers presented at conferences

1. Temperature Stable Microwave Substrate Based on PEEK/TiO₂ Composite System MMA- 2006, Finland

2. Effect of Particle Size on the Microwave Dielectric Properties of Silica Filled PTFE Substrates, MMA-2006, Finland

3. Fabrication of SrTiO₃ filled PTFE Composites for Microwave Substrate Applications ICMARS 2006 Jodhpur, India

4. PTFE/Ceramic Flexible Laminates for Microwave Substrates Applications, APSYM 2008, CUSAT, Kochi, India