PREFACE

Materials play a key role in technology developments and implementation. New technologies press the need for better , easily preparable , low cost materials having desired properties . Availability of materials having superior properties and knowledge of their overall behavior prompt development of better technologies or upgradation of the existing ones. Growth of devices such as LED lights, gas lighters , sensors and night vision devices, computer memories, IC chips in smart cards, supersonic missiles etc. have become possible due to availability of suitable materials. Ferroelectric ceramics such as those derived from perovskite $BaTiO_3$ are very useful materials having wide variety of applications such as PTC thermistors, piezoelectric devices , microwave phase shifters etc. These ceramics have been in demand for technological applications as well as basic research because of the fact that their properties can be suitably tailored by substitutions at Ba and Ti sites and by changing the processing variables.

During the fabrication process, the ceramic piece has to be integrated to the whole system. Therefore, in order to design the system for optimum performance, an understanding of the overall behavior would be highly useful. A ceramic system comprises small randomly oriented crystallites called grains that are joined together . The joining region, called grain boundary, has strained bonds and, therefore, its properties are different from grains due to mismatch. The overall behavior of the ceramic would have contributions from grains, grain boundaries and electrodes. In order to develop materials having desired and reproducible properties these contributions should be separated out for which technique of impedance spectroscopy has been found to be very useful. This essentially involves measurement of impedance as a function of frequency.

The impedance spectroscopic analysis is carried out by representing the ceramic system by some suitable equivalent circuit model involving lumped components such as resistor (R), capacitor (C). A parallel RC circuit has one time constant RC and can be readily used to represent one charge transfer process present in the system. A general practice is to connect more such RC circuits in series for representing a system having more charge transfer processes. Thus, a ceramic which can be treated as a grain –grain boundary –electrode system, may be represented by three parallel RC circuits connected in series. Equivalent circuits may be developed by considering all the charge transfer processes thought to be possibly present in the system. When distribution in certain properties exist, an equivalent circuit comprising a parallel RC circuit does not suffice. For analysis of these situations , use of constant phase angle elements (CPE) has been found to be very useful. However, in what way a CPE has to be connected in the model circuit is not so clear.

Therefore, in the present work simulation of the behavior of various suitable equivalent circuit models comprising CPE, resistive and capacitive components has been carried out for different values of parameters with a view to facilitate the choice of equivalent circuit models by comparing the experimental plots with the simulated ones. The work has given rise to a clear recipe for developing equivalent circuit models. An indication of presence of certain elements in a possible model to be chosen for a material is obtained by comparing the experimental plots with these simulated ones. For example, a linear portion appearing in the complex plane impedance plot may be considered as a signature of presence of series CPE in the model where as a depressed arc in the plot would indicate the presence of CPE in parallel.

In the present work, preparation and characterization of $BaTiO_3$ ceramics with substitutions of Sr, Fe and Sn has been taken up. Compositions $Ba_{1-x}Sr_xTiO_3$ (x=0.15 - 0.35), $BaFe_xTi_{1-x}O_3$ (x=0.03, 0.05, 0.10) and $BaTi_{1-x}Sn_xO_3$ (x= 0.05, 0.10, 0.15) have been prepared by solid state reaction method and characterized by X-Ray Diffraction (XRD), Scanning Electron Microscopy (SEM), Ferroelectric hysteresis (P-E) measurements, impedance spectroscopy and dielectric measurements as a function of temperature and frequency in the RF range, and dielectric measurements in the X-band of microwaves (8-12 GHz) at room temperature. These systems exhibit ferroelectric behavior which depends on values of x . The system $Ba_{1-x}Sr_xTiO_3$ has tetragonal structure and shows diffuse phase transition for all the values of x (≤ 0.35) studied. Permittivity measurements as a function of temperature and frequency have been carried out. The permittivity peak shifts towards lower temperatures as x is increased. This transition temperature is around room temperature for x=0.35. By using impedance spectroscopy , equivalent circuit models representing the data above the transition temperature are developed . These equivalent circuits would be useful for simulation purposes. Dependence of the grain and grain –boundary resistances on composition and temperature are obtained empirically. This relationship is found to be linear. The corresponding activation energies are obtained as (-1.1 x + 1.4) eV and (0.4 x + 1.0) eV respectively . Thus , as x increases, activation energy for grains decreases whereas for grain boundary, it increases. P-E hysteresis loop is observed in all the samples indicating ferroelectric behavior. Dielectric measurements at frequencies in the range 8-12 GHz are also carried out at room temperature. The value of permittivity is found to be in the range 18 - 45 and loss in the range 0.1 -0.4.

Compositions with x =0.03 , 0.05 and 0.10 in the system $BaFe_xTi_{1-x}O_3$ have been prepared and characterized . It was found that these samples contain coexisting tetragonal and hexagonal phases, the latter increasing with the doping level. Dielectric measurements have been carried out as a function of frequency and temperature. Composition with x = 0.03 exhibits a relaxor behavior whereas sample with x =0.05 shows diffuse phase transition. Contributions of tetragonal and hexagonal phases , grain boundaries and electrodes to the overall dielectric behavior have been separated out using impedance spectroscopic analysis and equivalent circuit models representing the data obtained. Dielectric measurements at frequencies in the range 8-12 GHz are also carried out at room temperature.

Compositions with x= 0.05, 0.10 and 0.15 in the system $BaTi_{1-x}Sn_xO_3$ have been prepared and characterized . Compositions with x = 0.05 and 0.10 are found to be in tetragonal phase while x=0.15 is in cubic phase. The samples with x = 0.05 and 0.10 show diffuse phase transition. The permittivity peak shifts towards lower temperatures as x is increased Equivalent circuit models representing the data above the transition temperature are developed using impedance spectroscopy. P-E hysteresis loop is observed in the samples with x = 0.05 and x=0.10. Dielectric measurements at frequencies in the range 8-12 GHz are also carried out at room temperature.

As a possible application of the materials studied in the present work, effort was made to develop a design of rectangular dielectric resonator antenna (RDRA) operating in X- band of microwaves (8-12 GHz) based on dielectric constant values found for these materials. The X band is used for radar, satellite communication and terrestrial broadband. Recent developments of communication technologies aimed at increasing data rate by orders of magnitude have pressed the need for antennas having large band width so as to accommodate large number of channels, high gain, and small size. In general, higher frequency bands offer larger bandwidth capabilities in communication. Dielectric resonator antennas (DRA) have attracted much attentions recently due to their low cost, absence of losses at high frequencies that arise because of presence of metallic parts in usual antennas and flexibility in design . A DRA consists of a small puck of a dielectric material of certain shape and size. The size varies as $(\varepsilon)^{-1/2}$ so that desired miniaturization is possible by using suitable high permittivity dielectrics.

The present study indicates that some systems, such as $Ba_{1-x}Sr_xTiO_3$ (x = 0.35), and $BaTi_{1-x}Sn_xO_3$ (x= 0.15) have permittivity and dielectric loss (tan δ) around 40 and 0.1 respectively. It has been reported by Zivkovic (2012) enhancement in bandwidth of wire antenna by loading it with a material having dielectric loss around 0.05. Ceramic systems studied in the present work have losses in the range 0.1 to 0.4. In order to explore the possibility of using these in development of antenna , design and simulation of rectangular dielectric resonator antenna (RDRA) was pursued with a view to propose a suitable design where these materials may be utilized. Design of an aperture coupled RDRA array comprising three elements of permittivity equal to 40 and resonance frequency of 10 GHz is proposed and the effect of varying the value of loss on its performance is observed. High Frequency Structure Simulator (HFSS) software was used. The results of our preliminary studies indicate that when the outer elements are aperture coupled and the middle one is parasitic the band width slightly increases by 3% (from 1.32 GHz to 1.36 GHz) and gain decreases only by 2.7 % (from 5.97 dB to 5.81 dB) as the loss in middle element is increased from 0 to 0.1 keeping the outer elements loss less. The far field pattern remains almost same as loss is increased. It indicates that low cost, easily preparable but slightly lossy (tan δ ~0.1) materials , such as $Ba_{1-x}Sr_xTiO3$ (x = 0.35) ceramics having permittivity and loss around 40 and 0.1 respectively studied in the present work may be used in conjunction with low loss dielectrics in development of antennas by applying the present RDRA design with some trade off with gain .