Complex perovskite oxides with high dielectric constants are widely used in technological applications such as capacitors, resonators and filters. High dielectric constants allow smaller capacitive components, thus offering the opportunity to decrease the size of the electronic devices\textsuperscript{1}. The $\varepsilon'$ (dielectric constant) value of these oxides exhibits only small variation with change in temperature and frequency, which is desirable for technological applications. The high value permittivity dispersion observed in these oxides may be explained in terms of dipole relaxation related to point defects and the low frequency dispersion effect is influenced by marked electrical conductivity\textsuperscript{2,4}.

Various relaxation processes seem to coexist in real perovskite crystals or ceramics, which contain number of different energy barriers due to point defects appearing during the technological process. Therefore, the departure of the response from the ideal Debye model in solid-state samples, resulting from the interaction between dipoles, cannot be disregarded\textsuperscript{5}. The situation in solid solutions or compounds is complex, leading to ambiguity of analyses based on particular models with formulae having many parameters\textsuperscript{6}.

Niobium based perovskite oxides attract attention as compounds, since they show a combination of promising electrical and mechanical properties for use in the electronic industries\textsuperscript{7-10}. Recently, we have investigated the dielectric behaviour of niobium based lead free ceramic system, barium-aluminium-niobate $\text{BaAl}_{1/2}\text{Nb}_{1/2}\text{O}_3$\textsuperscript{11}. An analysis of the real and imaginary parts of the dielectric permittivity with frequency has been performed, assuming a distribution of relaxation times as confirmed by Cole-Cole plots as well as the scaling behaviour of the imaginary part of impedance spectra. The high value permittivity dispersion has been performed, assuming a distribution of relaxation times as confirmed by Cole-Cole plots as well as the scaling behaviour of the dielectric loss spectra. This scaling behaviour has suggested that the distribution of relaxation times is temperature independent\textsuperscript{11}. In this paper we investigate the electric relaxation properties of the strontium-magnesium-niobate, $\text{SrMg}_{1/3}\text{Nb}_{2/3}\text{O}_3$ (SMN) ceramic in a temperature range from room temperature to 613 K by means of dielectric spectroscopy. The electric modulus formalism is applied to analyse the dielectric data. It is demonstrated that this approach is a very effective and simple to characterize relaxation properties of the ceramics.

It is to be mentioned here that the use of the electric modulus formalism gives insight into the bulk response and is effective in the case of conducting samples. Hence, the local behaviour of the defects can be separated from electrode effects\textsuperscript{5}. Such an approach allows us to overcome the difficulties caused by high electrical conductivity, which can mask the dielectric relaxation processes.
Experimental Procedure

In analogy to our previous work\textsuperscript{11}, the ceramic method was employed for the synthesis of SMN. The raw chemicals used in this study were reagent grade SrCO\textsubscript{3}, MgO and Nb\textsubscript{2}O\textsubscript{5}. The mixed raw chemicals in stoichiometric ratio were calcined in a Pt crucible at 1300°C in air for 9 h and brought to room temperature under controlled cooling. The calcined sample was pelletized into a disk using polyvinyl alcohol as binder. Finally, the disks were sintered at 1350°C for 4 h and cooled down to room temperature by adjusting the cooling rate.

The X-ray powder diffraction pattern of the sample was taken at room temperature using a Philips PW1877 automatic X-ray powder diffractometer. To measure the electrical properties, gold electrodes were formed on both surfaces of sintered disks. The frequency dependence of the dielectric constant and loss tangent (\(\tan \delta\)) was obtained using an LCR meter (Hioki) in the temperature range from room temperature to 613 K and in the frequency range of 50 Hz-1 MHz. All the dielectric data were collected while heating at a rate of 0.5°C min\textsuperscript{-1}. These results were found to be reproducible.

Results and Discussion

Figure 1 shows the X-ray diffraction pattern of the sample taken at room temperature. All the reflection peaks of the X-ray profile were indexed and lattice parameters were determined using a least-squares method with the help of a standard computer programme (POWD). Good agreement between the observed and calculated interplaner spacing (\(d\)-values) suggests that the compound is having monoclinic structure at room temperature with \(\beta=93.3^\circ\) (\(a=3.9256\) Å, \(b=6.8557\) Å and \(c=4.7437\) Å). X-ray diffraction confirms that the specimen is single phase.

The frequency (angular) dependence of the real (\(\varepsilon'\)) and imaginary (\(\varepsilon''\)) parts of dielectric constant of SMN as a function of temperature is plotted in Figs 2a and 2b respectively. At the temperature 473 K, dielectric constant gradually decreases with increasing frequency with a value of 174 at 50 Hz. With increasing temperature, it increases apparently, which becomes even more significant at low frequency (below 150 Hz). The observed increase in dispersion with increased temperature suggests that ac conductivity in SMN is a bulk effect. At elevated temperatures (>553 K), the dielectric constant at low frequency is rather high (>600), and it falls against frequency at first and then becomes more or less stabilized down to above 150 Hz (Fig. 2a). The low frequency dispersion in Fig. 2a indicates the absence of any inhomogeneties arising owing to Maxwell-Wagner polarization.

Fig. 1—XRD pattern of \(\text{SrMg}_{1/3}\text{Nb}_{2/3}\text{O}_3\) at room temperature

![Fig. 1—XRD pattern of \(\text{SrMg}_{1/3}\text{Nb}_{2/3}\text{O}_3\) at room temperature](image)

Fig. 2—Frequency dependence of (a) \(\varepsilon'\) and (b) \(\varepsilon''\) of \(\text{SrMg}_{1/3}\text{Nb}_{2/3}\text{O}_3\) at various temperatures

![Fig. 2—Frequency dependence of (a) \(\varepsilon'\) and (b) \(\varepsilon''\) of \(\text{SrMg}_{1/3}\text{Nb}_{2/3}\text{O}_3\) at various temperatures](image)
Figure 2b plots the frequency (angular) dependence of imaginary part of dielectric constant ($\varepsilon''$) at various temperatures. Similar to the dependence of dielectric constant on temperature, the dielectric loss increases with increasing temperature. This indicates the thermally activated nature of the dielectric relaxation of the system.

For the quantitative analysis of dielectric spectra the Cole-Cole equation has been used:

$$\varepsilon^* = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + (i\omega\tau)^{1-\alpha}} \quad \ldots \quad (1)$$

Here $\varepsilon_\infty$ is the high frequency limit of the permittivity, $\varepsilon_s - \varepsilon_\infty$ is dielectric strength, $\omega (= 2\pi\nu)$ is the angular frequency, $\tau$ is the mean relaxation time and $\alpha$ represents the distribution of relaxation time. Figure 3 depicts a Cole-Cole representative plot for $T = 523$ K. It is evident from the plot that the relaxation process differs from the purely Debye process (for which $\alpha = 0$). The parameter $\alpha$ as determined from the angle subtended by the radius of Cole-Cole circle with the $\varepsilon'$ axis passing through the origin of the $\varepsilon''$-axis is found to be 0.16. The Cole-Cole plot explains the polydispersive nature of dielectric relaxation of SMN.

If one assumes that all dielectric loss in the temperature range studied is due to conductivity, the appropriate formula is

$$\sigma' = \omega\varepsilon_0\varepsilon'' \quad \ldots \quad (2)$$

Here $\sigma'$ is the real part of the conductivity. The frequency spectrum of the conductivity for SMN is shown in Fig. 4 at different measuring temperatures. The conductivity shows a dispersion which shifts to higher frequency side with the increase of temperature. It is seen from Fig. 4 that $\sigma'$ decreases with decreasing frequency and becomes independent of frequency after a certain value. Extrapolation of this part towards lower frequency will give $\sigma_{dc}$ thus obtained follows Arrhenius law given by

$$\sigma_{dc} = \sigma_0 \exp \left[ -\frac{E_\sigma}{k_B T} \right] \quad \ldots \quad (3)$$

with activation energy $E_\sigma = 1.8$ eV as shown in Fig. 5.

The very basic fact about ac conductivity in disordered solids is that $\sigma'$ is an increasing function of frequency (any hopping model has this feature). In a hopping model it is possible to distinguish different characteristic regions of frequency. At low frequencies where the conductivity is constant, the transport takes place on infinite paths. For a region of frequencies where the conductivity increases strongly with frequency, the transport is dominated by contributions from hopping infinite clusters.
The dielectric properties of materials can be expressed in various ways, using different representations. Although these alternative representations are equally valid\textsuperscript{14}, they may often provide new insight into the dielectric and electrical properties of materials. For example, a comparison of the complex dielectric permittivity \( \varepsilon^* \) and electric modulus \( M^* \) representation allows us to distinguish local dielectric relaxation (e.g. dipole reorientation) from long-range electrical conductivity. Based on values of the activation energy and the characteristic relaxation time, it is possible to ascribe a given process to a specific mechanism. To achieve this purpose, we applied different representations to analyse the same data.

We have adopted the modulus formalism to study the relaxation mechanism in SMN. From the physical point of view, the electrical modulus corresponds to the relaxation of the electric field in the materials when the electric displacement remains constant. The usefulness of the modulus representation in the analysis of the relaxation properties has been demonstrated for polycrystalline ceramic\textsuperscript{15}. In the modulus formalism, an electric modulus \( M^*(\omega) \) is defined in terms of the complex dielectric permittivity \( \varepsilon^*(\omega) \) by

\[
M^*(\omega) = \frac{\varepsilon^*(\omega)}{\varepsilon'(\omega)^2 + \varepsilon''(\omega)^2} \quad \text{… (5)}
\]

Based on Eq. (5) we have changed the form of presentation of the dielectric data from \( \varepsilon'(\omega) \) and \( \varepsilon''(\omega) \) to \( M'(\omega) \) and \( M''(\omega) \). The obtained modulus spectra \( M'(\omega) \) and \( M''(\omega) \) are depicted in Figs 6a and 6b. Data presented in this way exhibit a pronounced relaxation peak for \( M''(\omega) \) that moves towards lower frequencies during cooling of the sample. Consequently, it means that the relaxation rate for this process decreases with decreasing temperature. As a convenient measure of the characteristic relaxation time one can choose the inverse of frequency of the maximum peak position, i.e., \( \tau_m = \omega_m^{-1} \). Thus, we can determine the temperature dependence of the characteristic relaxation time as shown in Fig. 7, which satisfies Arrhenius law. From the numerical fitting analysis, we found the value of the activation energy = 1.76 eV. We have scaled each \( M'' \) by \( M''_m \) (\( M''_m \) is the peak value of imaginary part of electric modulus) and each frequency by \( \omega_m \) where \( \omega_m \) corresponds to the frequency of the peak position of
$M''$ in the $M''$ versus log $\omega$ plots in Fig. 8. The overlap of the curves for all the temperatures indicates that the dynamical processes are nearly temperature independent.

The frequency dependence of real ($Z'$) and imaginary part ($Z''$) parts of impedance of SMN at various temperatures is shown in Figs 9a and 9b respectively. In the accessible frequency range, in Fig. 9b the spectrum at each temperature exhibits one relaxation peak whose peak frequency, $\omega_m$, increases with increasing temperature. The shape of the $Z''$ spectra is independent of temperature, as can be seen from Fig. 10, where both the axes are normalized.
Figure 12 shows an Arrhenius plot of $Z''$ and the corresponding activation energy of 2.04 eV for relaxation is found to be close to the activation energy for $M''$.

**Conclusions**

The frequency-dependent dielectric dispersion of strontium-magnesium-niobate, SrMg$_{1/3}$Nb$_{2/3}$O$_3$ ceramic synthesized by the solid state reaction technique is investigated in the temperature range from room temperature to 613 K. The X-ray diffraction of the sample at room temperature shows monoclinic phase. An analysis of the real and imaginary parts of impedance is performed, assuming a distribution of relaxation times as confirmed by Cole-Cole plot as well as the scaling behaviour of the imaginary part of impedance spectra. The scaling behaviour suggests that the distribution of relaxation times is temperature independent. The frequency dependent electrical data were analyzed in the framework of the conductivity and modulus formalisms. Both these formalisms provided for qualitative similarities in relaxation times.

**References**