4.1 Introduction:

The present chapter deals with the characterization of the yCMPBT composites, where the yCMPBT refers to y[CFMO-PBT]. Initially ac conductivity of composites is discussed in the light of presence of interfacial polarization. Further the dc resistivity is also measured at room temperature for yCMPBT composites. The discussion is followed by the presentation of results on dielectric properties of the composites. Here variation of $\varepsilon_r$, $\varepsilon'$ that is real part of dielectric constant, $\varepsilon''$ that is imaginary part of dielectric constant and loss tangent tan$\delta$ are presented and analyzed for their variation with temperature and frequency. In the present case dielectric constant refers to relative permittivity of the material. Importantly the magnetoelectric effect and magnetodielectric effect in these systems are elaborated in detail in section-4.4 and section-4.5 respectively. The theoretical background of the parameters being studied is already discussed in Chapter-I and Chapter-II, therefore whenever required the reference is made to these chapters.

4.2: Electrical Conductivity:

4.2.1: AC Conductivity:

The polar materials like ferrite and ferroelectric systems are known to possess electrical conduction due to small polarons. It could be interesting to investigate these features for ME composites also. For conduction due to small polarons the conductivity is observed to obey the following relation [1-3].

$$ (\sigma_{ac}-\sigma_{dc}) = \omega^2 \Gamma^2 / (1+\omega^2 \Gamma^2) $$

Here $\omega$ is angular frequency and $\Gamma$ is staying time of the small polarons. It is observed that the $\Gamma$ is of the order of $10^{-10}$ seconds for these compositions and therefore $\omega^2 \Gamma^2$ is $\ll 1$. Within this approximation the equation 4.1 could be written as.

$$ \log(\sigma_{ac}-\sigma_{dc}) = \Gamma^2 \log\omega^2 $$
Figures 4.1a and 4.1b show variation of ac conductivity as a function of logω² for yCMPBT composites sintered at 1150°C and 1200°C respectively. From equation 4.1 above it is expected that for conduction to be due to small polarons, the log(σ_{ac}−σ_{dc}) v/s logω² should be linear with slope equal to Γ². From figures 4.1a and 4.1b it is seen that for frequencies above 10kHz, the behavior of (σ_{ac}−σ_{dc}) is linear.

Thus it could be concluded that dominant mode of conduction in these systems is small polarons [1-5]. It is known that the interfacial polarization is an effect that occurs only at lower frequencies and the behavior of log(σ_{ac}−σ_{dc}) is linear, uniform for frequencies greater than 10kHz. Therefore it is expected that log(σ_{ac}−σ_{dc}) v/s logω² may deviate from linearity for frequencies below 10kHz.

Here figure 4.1a shows the log-log behavior of yCMPBT composites sintered at 1150°C, while figure 4.1b shows the log-log behavior of yCMPBT composites sintered at 1200°C. The following observations are noteworthy:

1. For frequencies above 10kHz the variation is linear and slope of the lines is almost independent of y.
2. The conductivity is minimum for y=0.5. Initially the σ decreases with y up to y=0.5 and then increases for y=0.6.
3. The deviation is observed from the linear behavior for frequencies less than 10kHz. A deviation becomes predominant as y increases from 0.2 to 0.6. It is known that the ME composites posses a contribution due to interfacial polarization [6-8]. The interfacial polarization may cause a change in slope of the log(σ_{ac}−σ_{dc}) behaviour. As expected, the change in slope is maximum for y=0.5 and 0.6.

Thus it could be concluded that the conduction in the individual phases is due to small polarons [9-12] and presence of interfacial polarization reduces the effective staying time of the polarons, especially at low frequencies.
Figure 4.1a: Variation of ac conductivity as a function of $\log \omega^2$ for yCMPBT with $T_s=1150^\circ C$

Figure 4.1b: Variation of ac conductivity as a function of $\log \omega^2$ for yCMPBT with $T_s=1200^\circ C$
4.2.2 DC resistivity:

Figure-4.2 shows variation of dc resistivity as a function of $y$ for $y$CMPBT composites sintered at 1150°C and 1200°C. Figure clearly shows the decreasing trend of $\rho$ with increase in $y$. In general, the $\rho$ of the material increases slightly for $T_s$ changing from 1150°C to 1200°C. This feature may appear because of the grain growth and formation of voids as the $T_s$ is increased [6]. Similar observations are observed in the present case. The table-4.1 shows the variation of $\rho$ with $y$ for $y$CMPBT composites of for varying $T_s$. From the table it is observed that $\rho$ decreases up to $y=0.5$ and then shows slight increase for $y=0.6$.

Table-4.1 variation of $\rho$ with $y$ for the $y$CMPBT composites

<table>
<thead>
<tr>
<th>$y$</th>
<th>Log $\rho$dc (Ω·m) ($T_s = 1150^\circ$C)</th>
<th>Log $\rho$dc (Ω·m) ($T_s = 1225^\circ$C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2</td>
<td>8.5</td>
<td>8.69</td>
</tr>
<tr>
<td>0.3</td>
<td>7.93</td>
<td>8.02</td>
</tr>
<tr>
<td>0.4</td>
<td>5.79</td>
<td>5.85</td>
</tr>
<tr>
<td>0.5</td>
<td>4.67</td>
<td>4.74</td>
</tr>
<tr>
<td>0.6</td>
<td>5.79</td>
<td>5.55</td>
</tr>
</tbody>
</table>
Figure 4.2: Variation of $\log_{10}\rho_{dc}$ as a function of $y$ for MPBT composites.
4.3 Dielectric properties of CMPBT composites:

4.3.1 Introduction:
The dielectric constant in case of yCMPBT composites is expected to have two contributions, one due to parent PBT ferroelectric and other due to the interfacial polarization occurring at the interfaces between ferrite (CFMO) and ferroelectric (PBT) phases. The interfacial polarization occurs because of the difference in the resistivity and dielectric constant of these two distinct phases. As y increases contribution due to ferroelectric phase may decrease, while the contribution due to interfacial polarization may increase as y(1-y). The y(1-y) type of variation becomes maximum for y=0.5. This is a typical behavior of binary composites.

From the discussion above it could be seen that at low frequencies the dominant mode of polarization is the interfacial polarization [13, 14].

4.3.2 Variation of dielectric constant (ε) as a function of composition:

Figure-4.3 shows variation of dielectric constant (ε) represented as relative permittivity, as a function of temperature at 1kHz for yCMPBT composites for y= 0.2 to 0.6 sintered at 1150°C. For the purpose of reference the variation of ε for y=0 is also shown in figure-4.3. From the figure it is observed that all the compositions exhibit a diffused phase transition (DPT) in the vicinity of 190°C, consistent with the earlier reports [15-17].

Similar observations are obtained for yCMPBT composites sintered at 1200°C as shown in figure-4.4. From figures-4.3 and 4.4 it is observed that ε initially increases with y for y=0.2 to 0.5 and then suddenly decreases for y=0.6.
Figure 4.3: Variation of dielectric constant with temperature for varying $y$ for CMPBT composites sintered at 1150$^\circ$C

Figure 4.4: Variation of dielectric constant with temperature for varying $y$ for CMPBT composites sintered at 1200$^\circ$C
4.3.3 Variation of dielectric constant as a function of temperature and frequency:

The DPT may occur because of the presence of two separate phases or could be due to a relaxor type behavior. This feature could be understood clearly from the variation of $\varepsilon$ with frequency. Here figures-4.5 to 4.7 and figures-4.8 to 4.10 show variation of $\varepsilon$ as a function of temperature and frequency for yCMPBT composites sintered at 1150°C and 1200°C respectively. It is observed that $\varepsilon$ possess large dispersion at low frequencies as predicted using the Maxwell-Wagner model for the presence of interfacial polarization. It is also observed that $\varepsilon$ decreases with increasing frequency. Further it is observed that the $T_c$ is independent of frequency and no relaxor type behavior is present in the yCMPBT composites.

Tables-4.2 and 4.3 give variation of dielectric constant ($\varepsilon$) and electrical quality factor $Q$ with $y$ for yCMPBT composites at $f=1kHz$ and $T_s$ equal to 1150°C and 1200°C respectively. The tables also show values of $\varepsilon$ and $Q$ at $T=200°C$, where the $\varepsilon$ becomes maximum. From table it is observed that $\varepsilon$ shows an increasing trend up to $y=0.5$. The magnitude of $\varepsilon$ and $Q$ are interdependent for a given value of $y$. 
Figure 4.5: Variation of dielectric constant $\varepsilon$ as a function of temperature for varying frequency for 0.3CMPBT sintered at 1150$^0$C.

Figure 4.6: Variation of dielectric constant $\varepsilon$ as a function of temperature for varying frequency for 0.5CMPBT sintered at 1150$^0$C.
Figure 4.7: Variation of dielectric constant $\varepsilon$ as a function of temperature for varying frequency for 0.6CMPBT sintered at $1150^\circ$C.

Figure 4.8: Variation of dielectric constant $\varepsilon$ as a function of temperature for varying frequency for 0.3CMPBT sintered at $1200^\circ$C.
Figure 4.9: Variation of dielectric constant $\varepsilon$ as a function of temperature for varying frequency for 0.4CMPBT sintered at 1200°C.

Figure 4.10: Variation of dielectric constant $\varepsilon$ as a function of temperature for varying frequency for 0.5CMPBT sintered at 1200°C.
Table -4.2: Variation of dielectric constant $\varepsilon$ and $Q$ at 1 kHz for the composites sintered at 1150$^\circ$C.

<table>
<thead>
<tr>
<th>$y$</th>
<th>$\varepsilon$ at room temperature</th>
<th>$Q$ at room temperature</th>
<th>$\varepsilon_{\text{max}}$</th>
<th>$Q$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2</td>
<td>50.4</td>
<td>11.6</td>
<td>227</td>
<td>1.89</td>
</tr>
<tr>
<td>0.3</td>
<td>78.3</td>
<td>5.97</td>
<td>930</td>
<td>0.3</td>
</tr>
<tr>
<td>0.4</td>
<td>77.3</td>
<td>3.37</td>
<td>1119</td>
<td>0.39</td>
</tr>
<tr>
<td>0.5</td>
<td>100.3</td>
<td>5.48</td>
<td>1140</td>
<td>0.03</td>
</tr>
<tr>
<td>0.6</td>
<td>18.5</td>
<td>15.3</td>
<td>73</td>
<td>0.16</td>
</tr>
</tbody>
</table>

Table – 4.3: Variation of dielectric constant $\varepsilon$ and $Q$ at 1 kHz for the composites sintered at 1200$^\circ$C.

<table>
<thead>
<tr>
<th>$y$</th>
<th>$\varepsilon$ at room temperature</th>
<th>$Q$ at room temperature</th>
<th>$\varepsilon_{\text{max}}$</th>
<th>$Q$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2</td>
<td>229</td>
<td>9.68</td>
<td>1307</td>
<td>2.17</td>
</tr>
<tr>
<td>0.3</td>
<td>198</td>
<td>3.15</td>
<td>2341</td>
<td>1.13</td>
</tr>
<tr>
<td>0.4</td>
<td>264</td>
<td>2.67</td>
<td>3271</td>
<td>0.56</td>
</tr>
<tr>
<td>0.5</td>
<td>411</td>
<td>1.01</td>
<td>6886</td>
<td>0.21</td>
</tr>
<tr>
<td>0.6</td>
<td>35</td>
<td>4.13</td>
<td>224</td>
<td>0.02</td>
</tr>
</tbody>
</table>
4.3.4 Variation of loss tangent $\tan\delta$ as a function of temperature and frequency:

Figure-4.11 shows variation of loss tangent $\tan\delta$ as a function of temperature for various frequencies between 1kHz to 1MHz for 0.4CMPBT composite with $T_s=1150^\circ$C. From the figure it is observed that value of $\tan\delta$ decreases with increasing frequency. Similar results are observed for 0.4CMPBT composite sintered at 1200$^\circ$C as shown in figure-4.12. The behavior is as expected from the basic theory of dielectric materials [section-2.7]. At low frequencies $\tan\delta$ v/s temperature behavior shows large dispersion, which is a characteristic of presence of the interfacial polarization [6-8].

For the remaining compositions [y=0.2, 0.3, 0.5 and 0.6], the variation of loss tangent $\tan\delta$ v/s temperature with varying frequencies is also studied and similar results are observed as discussed above.
Figure 4.11: Variation of tanδ as a function of temperature for varying frequencies for 0.4CMPBT composite sintered at 1150°C

Figure 4.12: Variation of tanδ as a function of temperature for varying frequencies for 0.4CMPBT composite sintered at 1200°C
4.3.5: Variation of dielectric constant and tanδ as a function of frequency:

To understand the effect of interfacial polarization, the real part $\varepsilon'$ and imaginary part $\varepsilon''$ of the complex permittivity is also determined. The variation of $\varepsilon'$ as a function of frequency is shown in figures-4.13 and 4.14 for the $y$CMPBT composites sintered at 1150°C and 1200°C respectively, while variation of $\varepsilon''$ as a function of frequency for the same compositions is shown in figures-4.15 and 4.16. From figures-4.13 and 4.14 it is observed that $\varepsilon'$ rapidly decreases with frequency for lower values of frequency and remains constant for higher values of frequencies. From figure-4.15 and figure-4.17 it is seen that $\varepsilon''$ also show similar variations with frequency. The high value of dielectric constant at lower frequency is explained on the basis of space charge polarization due to inhomogeneous dielectric structure and resistivity of the samples [18].

For the composite systems it is known that the dielectric constant posses two contributions one due to parent ferroelectric phase that is PBT and other due to interfacial polarization between PBT and CFMO particles. The interfacial polarization occurs because of the difference in the resistivity and dielectric constant of PBT and CFMO phases [19]. Further the interfacial polarization could be understood in terms of the Maxwell-Wagner model and Koops phenomenological theory [15, 20-21]. To understand the observed behavior of $\varepsilon$ in perspective of these models, the variation of loss tangent tanδ as a function of frequency is determined in the frequency range 500Hz to 1MHz.

The variation of tanδ as a function of logf for varying $y$ and $T_s$ for $y$CMPBT composites is as shown in figures-4.17 and 4.18. The variation of tanδ with logf is also consistent with above mentioned theories. From these figures it is observed that for $y=0.2$ and $y=0.6$, tanδ passes through a resonance peak. The resonance occurs at the frequency where the time required for the charge to transfer across the interface
matches with reciprocal of applied frequency. Similar observations are reported for the other titanate systems [22].

From the above discussion it could be said that the dielectric properties of $y$CMPBT composites posses a large contribution of interfacial polarization for $y=0.5$. 
Figure 4.13: Variation of $\varepsilon'$ v/s log$\omega$ for yCMPBT composites, Ts=1150$^0$C

Figure 4.14: Variation of $\varepsilon'$ v/s log$\omega$ for yCMPBT composites, Ts=1200$^0$C
Figure 4.15: Variation of $\varepsilon''$ v/s $\log \omega$ for yCMPBT composites, $Ts=1150^0\text{C}$

Figure 4.16: Variation of $\varepsilon''$ v/s $\log \omega$ for yCMPBT composites, $Ts=1200^0\text{C}$
Figure 4.17: Variation of $\tan\delta$ as a function of $\log f$ of yCMPBT for $f=1\text{kHz}$ and $T_s=1150^\circ \text{C}$

Figure 4.18: Variation of $\tan\delta$ as a function of $\log f$ of yCMPBT for $f=1\text{kHz}$ and $T_s=1200^\circ \text{C}$
4.4: Magnetoelectric properties of CMPBT composites:

4.4.1 Introduction:

The dynamic ME coefficients $\alpha$ and $\beta$ of the composites $y$CMPBT are measured at 850Hz using custom designed measurement unit.

Linear and quadratic magnetoelectric coefficients $\alpha$ and $\beta$ are determined using the following relation,

$$\alpha = \frac{(dv / dh)*1}{t}$$

and

$$\beta = \frac{(dv / dH)*1}{2h_o}$$

Where:

$v$ : The rms value of voltage developed across the sample in response to an ac magnetic field ($h$).

$t$ : Thickness of the sample.

$h_o$ : rms value of applied magnetic field.

$H$ : Applied dc magnetic field.

The details of measurement techniques are already discussed in chapter-II.

4.4.2: Linear ME coefficient $\alpha$ as a function of composition at different sintering temperature:

Figure-4.19 shows variation of linear magnetoelectric coefficient ($\alpha$) as a function of $y$ for $y$CMPBT composites sintered at 1150°C and 1200°C. The table-4.4 shows the variation of $\alpha$ with $y$. The $\alpha$ is observed to be maximum for $y$=0.5. This feature is attributed to $y(1-y)$ type proportionality of $\alpha$. The high value of $\alpha=23$mV/Oe/cm for 0.5CMPBT at Ts= 1200°C is most interesting and useful feature of the present observations. From the table-4.4 it is also seen that $\alpha$ increases for increasing sintering temperature Ts, which could be attributed to the increased grain size and resulting
improved magnetomechanical coupling [19]. This feature is similar to observations on nickel ferrite and PZT systems [23]

4.4.3: Quadratic Magnetoelastic coefficient $\beta$ as a function of composition at different sintering temperature:

Figure-4.20 shows variation of quadratic magnetoelectric coefficient ($\beta$) as a function of $y$ for yCMPBT composites sintered at 1150°C and 1200°C. The table-4.4 also shows the variation of $\beta$ with $y$. As required for the ME composites the magnitude of $\beta$ is fairly small and the $\beta$ decreases with increasing sintering temperature. This too is also a device related property of ME composites. To determine the magnitude of $\beta$ the ME output ($v$) is measured as a function of applied dc magnetic field $H$ between 0 to 4.5 kOe. It is observed that as $y$ increases the $v$ increases with $H$. This is expected from the proportionality of ME output $v$ with $\lambda$. The $\lambda$ is known to follow the variation of $M_s$ with $H$. As $\lambda$ increases with $H$ the ME output should also increase with $H$ as observed in present case. From table-4.4 it is seen that similar to the variation of $\alpha$, $\beta$ also becomes maximum for $y = 0.5$. Here $\alpha$ and $\beta$ both are expected to follow the relation $y(1-y)$ and the present observations of $\beta$ confirm this prediction. The $\alpha$ and $\beta$ both pass through a broad maxima at $y=0.5$, as expected from the above relation.

4.4.4: ME coefficient ($\alpha$) as a function of frequency and composition:

Figures-4.21 and 4.22 show the variation of magnetoelectric coefficient $\alpha$ as a function of log$f$ for yCMPBT composites sintered at 1150°C and 1200°C respectively. From the basic theory of ME effect $\alpha$ is proportional to $(\lambda*k_m*d)/\epsilon$, where $\lambda$ is coefficient of magnetostriction, $k_m$ is magnetomechanical coupling coefficient, $d$ is piezoelectric constant and $\epsilon$ is dielectric constant. This means that $\alpha$ is inversely proportional to $\epsilon$. As the $\epsilon$ is observed to decrease with frequency (figures-4.7and 4.8), the $\alpha$ is expected to increase with increasing frequency and similar are the present observations.
Figure 4.19: Variation of linear ME coefficient $\alpha$ as a function of $y$ at different sintering temperature for $y$CMPBT composites.

Figure 4.20: Variation of linear ME coefficient $\beta$ as a function of $y$ at different sintering temperature for $y$CMPBT composites.
Figure 4.21: Variation of ME coefficient $\alpha$ with logf for varying yCMPBT sintered at $1150^0$C.

Figure 4.22: Variation of ME coefficient $\alpha$ with logf for varying yCMPBT for $T_s=1200^0$C.
Table 4.4: Variation of magnetoelastic coefficients $\alpha$ and $\beta$ with $y$ for the composites sintered at 1150°C and 1200°C

<table>
<thead>
<tr>
<th>$y$</th>
<th>$\alpha$(1150°C) mv/Oe/cm</th>
<th>$\alpha$(1200°C) mv/Oe/cm</th>
<th>$\beta$(1150°C)$10^{-4}$ mv/Oe²/cm</th>
<th>$\beta$(1200°C)$10^{-4}$ mv/Oe²/cm</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2</td>
<td>10.61</td>
<td>13.36</td>
<td>16.5</td>
<td>7.38</td>
</tr>
<tr>
<td>0.3</td>
<td>11.75</td>
<td>16</td>
<td>9.3</td>
<td>8.11</td>
</tr>
<tr>
<td>0.4</td>
<td>16.4</td>
<td>12.88</td>
<td>7.85</td>
<td>12.6</td>
</tr>
<tr>
<td>0.5</td>
<td>20.5</td>
<td>23</td>
<td>41.3</td>
<td>13.3</td>
</tr>
<tr>
<td>0.6</td>
<td>9</td>
<td>8.5</td>
<td>42.8</td>
<td>4.4</td>
</tr>
</tbody>
</table>
4.5: Ferroelectric and Ferromagnetic properties of yCMPBT composites:

Here figure 4.22a shows variation of P-E hysteresis for yCMPBT (y=0.3, 0.4 and 0.5) composites sintered at 1200°C. Further table-4.4a shows variation of $P_{\text{max}}$ and $P_{t}$ for the compositions specified above. The hysteresis loops of all the compositions corresponding to the loops for polycrystalline ferroelectric compositions. Further from table-4.4a it could be seen that for yCMPBT composites the $P_{\text{max}}$ decreases as y increases. The $P_{\text{max}}$ increases with decreasing y could be correlated with the content of PBT phase, it could be seen that the content of PBT phase increases with decrease in y.

It is interesting to note that the present composites also exhibits magneto hysteresis loops. The table 4.4b shows the variation of saturation magnetization ($M_s$), remnant magnetization ($M_r$) and coercive field ($H_c$) as a function of y for yCMPBT composites sintered at 1150°C. It is observed the $M_s$, $M_r$ and $H_c$ increases with increase in y. The present observations could be correlated with increasing content of CMFO (ferrite phase) in the composites as y increases.

![Figure 4.22a: Shows variation of P-E hysteresis for yCMPBT](image)

Figure 4.22a: Shows variation of P-E hysteresis for yCMPBT
Table 4.4a: Variation of maximum polarization ($P_{\text{max}}$) and remnant polarization ($P_r$) for $y$CMPBT composites.

<table>
<thead>
<tr>
<th>$y$</th>
<th>$P_{\text{max}}$ ($\mu$C/cm$^2$)</th>
<th>$P_r$ ($\mu$C/cm$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>2.1</td>
<td>1.71</td>
</tr>
<tr>
<td>0.3</td>
<td>0.34</td>
<td>0.19</td>
</tr>
<tr>
<td>0.4</td>
<td>0.29</td>
<td>0.14</td>
</tr>
<tr>
<td>0.5</td>
<td>0.15</td>
<td>0.06</td>
</tr>
</tbody>
</table>

Table 4.4b: Shows the variation of saturation magnetization ($M_s$), remnant magnetization ($M_r$) and coercive field ($H_c$) for $y$ CMPBT composites sintered at 1150°C.

<table>
<thead>
<tr>
<th>$y$</th>
<th>$M_s$</th>
<th>$M_r$</th>
<th>$H_c$ (Oe)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2</td>
<td>28.85</td>
<td>7.3</td>
<td>120</td>
</tr>
<tr>
<td>0.3</td>
<td>50.1</td>
<td>35.6</td>
<td>180</td>
</tr>
<tr>
<td>0.4</td>
<td>83.9</td>
<td>65.6</td>
<td>187.5</td>
</tr>
<tr>
<td>0.5</td>
<td>114</td>
<td>98.8</td>
<td>210</td>
</tr>
<tr>
<td>0.6</td>
<td>130</td>
<td>113</td>
<td>217.5</td>
</tr>
</tbody>
</table>
4.6: Magnetodielectric properties of CMPBT composites:

4.6.1 Introduction:

In case of PZT-MZF system it is argued that the stress induced on the piezoelectric system causes an increase in polarization and decrease in the dielectric constant $\varepsilon$. The stress induced by the magnetic field would be positive or negative depending on positive or negative value of $\lambda$. Thus $\varepsilon$ may decrease with applied magnetic field $H$ for positive value of $\lambda$, while $\varepsilon$ may increase with $H$ for $\lambda$ being negative. In the present case the CFMO possess a negative value of $\lambda$, therefore it is expected that $\varepsilon$ increases with increasing $H$. The effect discussed above is categorized as Gridnev’s effect [24]. Therefore the present yCMPBT composites are expected to show a positive $\text{Mc}$ due to Gridnev’s contribution, while the negative $\text{Mc}$ if the Catalan type contribution dominates [25-27]. As reported by Catalan the magnetocapacitance ($\text{Mc}$) could be positive or negative dependant on the nature of the composites [28-29]. If the composite appears as a multilayer laminate or like superlattice, the magnetocapacitance is expected to be negative, while for the 3-0 type connectivity of the composite, the magnetocapacitance is positive [28-30].

In the present case we are using 0.5CMPBT for investigation of magnetocapacitance owing to its high value of $\beta$ (table-4.4). In case of 0.5CMPBT the ferrite phase as well as ferroelectric phase exists in equal weight proportions. Therefore the Catalan type contribution is expected to be equivalent to the supperlattice type connectivity and the $\text{Mc}$ could be negative if the Catalan type contribution dominates [28-29].

4.6.2: Variation of Dielectric constant as a function of frequency at different applied Magnetic field:

Figures-4.23 and 4.24 show variation of dielectric constant $\varepsilon$ as a function of frequency for various applied magnetic field $H$ for 0.5CMPBT composites sintered at 1150$^\circ$C and 1200$^\circ$C respectively. It is observed that the magnetocapacitance ($\text{Mc}$) is positive throughout for the composite sintered at 1150$^\circ$C, while it is initially negative and then becomes positive for the composite sintered at 1200$^\circ$C. At higher sintering temperature it is excepted that electromechanical coupling could be more and both the Catalan type and Gridnev type contribution could be large. Thus the negative value of
Mc for the applied field up to 2kOe for 0.5CMPBT composite with T_s=1200\(^{0}\)C, could be due to Catalan type contribution, while the positive value of Mc for H>2kOe could be because of the Gridnev type contribution.

The table-4.5 shows variation of Mc with frequency for 0.5CMPBT composite for T_s=1150\(^{0}\)C and 1200\(^{0}\)C. From the table-4.5 it is seen that for T_s=1150\(^{0}\)C overall magnitude of Mc is large as compared to the magnitude of Mc for T_s=1200\(^{0}\)C. Primarily this feature could be attributed to higher value of quadratic magnetoelectric coefficient \(\beta\) for 0.5CMPBT sintered at 1150\(^{0}\)C as compared to the 1200\(^{0}\)C (table -4.4). Further it is seen that the Mc for T_s=1200\(^{0}\)C shows both negative as well as positive values. For the applied magnetic field H < 2kOe, the dielectric constant decreases with H, and Mc is negative. For further increase in H the \(\varepsilon\) increases with H and shows a positive value of Mc.

4.6.3: Variation of tan\(\delta\) as a function of frequency at different applied Magnetic field:

The behavior of tan\(\delta\) for these composites is as shown in figures-4.25 and 4.26. Here the figure-4.25 shows variation of loss tangent tan\(\delta\) as a function of frequency and applied magnetic field H for 0.5CMPBT composite with T_s=1150\(^{0}\)C, while figure 4.26 shows the same behavior for 0.5CMPBT composite sintered at 1200\(^{0}\)C. The variation in tan\(\delta\) is complimentary to the variation of \(\varepsilon\) and theoretical models as proposed by Gridnev and Catalan [24, 28]. Here tan\(\delta\) passes through a peak at electromechanical resonance frequency corresponding to the radial mode of oscillations. This behavior is also as expected from the basic theory of piezoelectric systems.
Figure 4.23: Variation of \( \varepsilon \) as a function of \( \log f \) for varying \( H \) for \( y \text{CMPBT} \) for \( T_s=1150^0\text{C} \)

Figure 4.24: Variation of \( \varepsilon \) as a function of \( \log f \) for varying \( H \) for \( y \text{CMPBT} \) composites with \( T_s=1200^0\text{C} \)
Figure 4.25: Variation of $\tan\delta$ as a function of $\log f$ for varying $H$ for yCMPBT composites with $Ts=1150^0\text{C}$

Figure 4.26: Variation of $\tan\delta$ as a function of $\log f$ for varying $H$ for yCMPBT composites with $Ts=1200^0\text{C}$
Table 4.5: Variation of Magnetodielectric capacitance (Mc) with frequency for CMPBT composites sintered at 1150°C and 1200°C.

<table>
<thead>
<tr>
<th>Frequency Hz</th>
<th>Mc (^{(1150°C)})</th>
<th>Mc(_1) (^{(1200°C)})</th>
<th>Mc(_2) (^{(1200°C)})</th>
</tr>
</thead>
<tbody>
<tr>
<td>500</td>
<td>11.8</td>
<td>-5.54</td>
<td>6.03</td>
</tr>
<tr>
<td>1000</td>
<td>6.23</td>
<td>-4.35</td>
<td>4.8</td>
</tr>
<tr>
<td>10000</td>
<td>12.73</td>
<td>-1.96</td>
<td>5.9</td>
</tr>
<tr>
<td>100000</td>
<td>4.97</td>
<td>-1.3</td>
<td>2.26</td>
</tr>
<tr>
<td>500000</td>
<td>4.22</td>
<td>-1.1</td>
<td>2.02</td>
</tr>
<tr>
<td>1000000</td>
<td>5.33</td>
<td>-0.95</td>
<td>1.57</td>
</tr>
</tbody>
</table>
References:

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