Chapter 8

Dielectric permittivity and magnetic permeability studies on rubber-nickel composites in the S and X band microwave frequencies

It has been pointed out in the previous chapter that the evaluation of dielectric permittivity and magnetic permeability of composites based on rubber and nickel nanoparticles in the S (2 to 4 GHz) and X (7 to 12 GHz) band is a major step in characterizing these samples for possible microwave absorbing applications. The evaluation of these parameters and the subsequent modeling of the surface impedance equations is attempted here. The findings are presented in this chapter.

The interest in electromagnetic absorbers in the microwave region has increased in recent times due to the significant expansion in applications using electromagnetic waves in this frequency band. There are numerous electronic gadgets operating in the microwave frequencies like mobile phones, wireless local area networks, radar systems etc and the problem of electromagnetic interference has worsened significantly, demanding the use of microwave absorbers [1-3]. A good absorber has to have low or negligible reflection, sufficiently large attenuation and good heat dissipation characteristics because the absorbed energy is converted to heat within the material [4]. In stealth technology, which is another important field of application of microwave absorption, especially in aerospace applications, total absorption of electromagnetic waves without any reflection is essential since the device should go undetected by radar signals.

8.1 Microwave absorbers

The complex dielectric permittivity and magnetic permeability of a material together defines its electromagnetic wave absorbing characteristics and this emphasizes the importance of
magnetic dielectric materials. Various kinds of magnetic dielectric materials have been developed over the years to find application as microwave absorbers such as ferrites, ferrite-polymer composites and composites containing metallic magnetic particulates [5-14]. In comparison to ferrites, metallic magnetic composites have several advantages like high saturation magnetization, tunability of magnetic permeability by controlling the volume fraction of metallic particles and mechanical flexibility of the host material of the composite. One major disadvantage of metallic magnetic composite is the drop in magnetic permeability at higher applied frequencies due to skin effect and eddy current loss developed in the particulates [15]. However this drawback can be overcome by using particles of size smaller than the skin-depth of the metal with respect to the frequency of operation. The skin depth of iron in gigahertz frequencies has been estimated to be about 1 \( \mu m \) and that of nickel about 500 nm [5, 15, 16]. Iron and nickel particles below these critical size limits dispersed in suitable dielectric materials therefore are potential candidates as microwave absorbers.

The investigations on the complex dielectric and magnetic properties of composites prepared by impregnating nanometric sized nickel particles in two different elastomer matrixes namely natural rubber and neoprene rubber are described in this chapter. Natural rubber is an easily available and inexpensive material and neoprene is a synthetic rubber with several superior physical properties compared to natural rubber such as better resistance to harsh chemicals, tough flame resistance and resistance to salinity. The ease with which they can be moulded into any complex shapes and size can give both these materials an extra edge in stealth applications. The complex dielectric permittivity and magnetic permeability of these composites are evaluated in the S band (2 to 4 GHz) and X band (7 to 12 GHz) microwave frequencies using cavity perturbation technique. The details of this technique are already described in detail in chapter 2.

### 8.2 Cavity perturbation method

For a rectangular cavity, real part of the dielectric permittivity can be calculated from the relation [17, 18]

\[
\frac{\Delta f}{f_s} = 2 \frac{V_s}{V_c} (\varepsilon'_s - 1)
\]

(8.1)
Where \( \Delta f \) is the shift in resonance frequency on introduction of the sample into the cavity and \( V_s \) and \( V_c \) are the volume of the sample and cavity respectively.

The imaginary part of the dielectric permittivity is given as

\[
\left[ \frac{1}{2Q_s} - \frac{1}{2Q_c} \right] = 2 \frac{V_s}{V_c} \varepsilon'' \quad (8.2)
\]

\( Q_s \) and \( Q_c \) are the quality factors of the cavity with and without sample, given by

\[
Q_s = \frac{f_s}{\Delta f}, \quad Q_c = \frac{f_c}{\Delta f} \quad (8.3)
\]

Magnetic permeability was measured by perturbing the cavity by the samples at positions where the electric field is zero. X band measurements were possible only for even modes since this cavity was provided with a hole at the centre of the cavity. The real and imaginary parts of the complex permeability of composite samples were determined using the relations

\[
\mu'_r - 1 = \left( \frac{\lambda_g^2 + 4a^2}{8a^2} \right) \left( \frac{\Delta f}{f_s} \right) \frac{V_c}{V_s} \quad (8.4)
\]

\[
\mu''_r = \left( \frac{\lambda_g^2 + 4a^2}{16a^2} \right) \left( \frac{1}{Q_s} - \frac{1}{Q_c} \right) \frac{V_c}{V_s} \quad (8.5)
\]

\[
\lambda_g = \frac{2l}{p} \quad (8.6)
\]

where \( p \) is the number of mode in which the cavity is excited for a particular measurement.

8.3 Measurements in the S band

Measurements in the S band were carried out by a two port network analyzer (ZVB4, Rohde & Schwarz) with the help of an S band rectangular cavity having dimensions 3.4cm x 7.2cm x 30.8cm by employing cavity perturbation technique. The rectangular S-band cavity operating in TE\(_{10}\) mode was fabricated with a narrow line slot (3mm x 180mm) to insert the sample material into the cavity. Calibration was performed by the method of through-open-short-match (TOSM) before carrying out the measurements. Within the frequency range of 2 GHz to 4 GHz,
the cavity resonates at five different frequencies corresponding to the TE\textsubscript{10n} modes. The cavity was perturbed at these frequencies using rectangular composite strips. The entire set-up was calibrated using FR4 glass epoxy, a standard material with known dielectric properties in microwave band. Dielectric permittivity and dielectric loss of the composite were determined by measuring the resonant frequency ($f_r$) and the quality factor (Q) of the microwave cavity when perturbed by inserting sample in the form of strips into the cavity at a position where electric field is found to be at a maximum.

8.4 Measurements in the X band

Rectangular strips of about 5 cm in length and 2mm x 2 mm cross section were cut from the sheets of composites and the dielectric measurements in the X band microwave frequencies were done by cavity perturbation method using an Agilent 4 port network analyzer with a rectangular cavity of dimensions 1cm x 2.3cm x 15.1 cm. The cavity has a precision cut hole at the centre (along the 2.3cm side) through which the sample can be introduced and dielectric measurements were carried out at odd modes (Electric field forms an anti-node at the centre) and permeability measurements at even modes (Electric field forms a node at the centre) [19]. The equations used for determining the complex permittivity and permeability are given in the section 8.2. The complex dielectric permittivity values are determined for four different frequencies in the X-band. These four frequencies are nearly 7.224 GHz, 8.239 GHz, 9.559 GHz and 11.067 GHz corresponding to the TE\textsubscript{103} to TE\textsubscript{109} modes of the cavity used for the measurements. The magnetic permeability was measured at five frequencies which are nearly 6.8746 GHz, 7.676 GHz, 8.8621 GHz, 10.283 GHz and 11.861 GHz corresponding to the TE\textsubscript{102} to TE\textsubscript{1010} modes of the cavity.

8.5 Complex dielectric permittivity of natural-rubber-nickel composites

The variation of dielectric permittivity ($\varepsilon_r'$) of the composites for five different frequencies in the S band is depicted in figure 8.1. It was observed that the dielectric permittivity of a particular sample remains nearly a constant in the entire frequency range. Blank rubber shows a permittivity of 2.61 at a frequency of 2.304 GHz and shows a maximum of 2.67 at
dielectric permittivity between 6.03 and 6.16 in the frequency range. Within the error limits, it can be assumed that the permittivity remains unchanged within the frequency range of S band. This is consistent with a number of previous results [20,21] The increase in permittivity with the nickel content can be attributed to the presence of interfacial polarization, the formation of internal barrier layer capacitance (IBLC) or the increase in the electrical conductivity of the samples [22]. The presence of interfacial polarization can be ruled out in the GHz frequencies because it is a slower relaxation process, generally with relaxation times of the order of $10^3$ s [23]. Metallic fillers can enhance the conductivity of the samples and this can be the reason for the increase in permittivity. The increase is found to be very sharp and much more than what predicted by the mathematical models suggested by Bruggman or Baziard [20,26,27] for composites with metallic fillers. An empirical relation of the form

$$\varepsilon_r' = \frac{\varepsilon_r''}{(1 - y)^8}$$

(8.7)

is used to fit the experimentally observed values and is in close agreement with the model. Here, $\varepsilon_r'$, $\varepsilon_r''$, and $y$ are the effective permittivity of the composite, permittivity of the polymer host material and the volume ratio of the metallic filler respectively. The variations of permittivity according to various models are shown in figure 8.2. The reason for such a sharp increase in.

![Figure 8.1](image.png)

**Figure 8.1:** Variation of dielectric permittivity of natural rubber based composites with frequency in the S band
permittivity is not known. It is possible that more than one factor is at work which contribute to the increase in the permittivity like enhancement of electrical conductivity or internal barrier layer capacitance.

![Figure 8.2: Variation of dielectric permittivity with volume ratio of nickel in the S band compared with different models (natural rubber based composites)](image_url)

The dielectric loss of the material in the S band is shown in figure 8.3. The loss remains nearly a constant for the entire range of 2 GHz to 4 GHz. The loss in the case of blank rubber lies in the range of 0.012 and 0.014. A flat curve indicates the absence of any relaxation process in this frequency regime. The electronic polarization of the molecules can only be the relevant polarization mechanism in these frequencies [23]. Polarization of the molecules is in synchronization with the applied signals since these signals have much lower period compared to the relaxation time of electronic polarization. But when the content of nickel in the composites increases, there is a steady increase in the dielectric loss too. This may be due to the increase in the reflectance of material with higher nickel content. Composite with 100 phr nickel content has maximum dielectric loss which is almost 9 times the loss of the blank rubber. It is evident that the dielectric loss can be enhanced by adding nickel particles in natural rubber and this opens up scope for developing materials for absorption of electromagnetic waves in microwave frequencies.
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Figure 8.3: Dielectric loss with frequency in the S band (natural rubber based composites)

8.6 Complex dielectric permittivity of neoprene-rubber-nickel composites

It was observed from the rf dielectric studies that neoprene rubber possesses a higher dielectric permittivity compared to natural rubber [chapter 7]. This enhancement in $\varepsilon'$ was observed in S band microwave frequency also. Figure 8.4 depicts the variation of $\varepsilon'$ with frequency and as observed in the case of natural rubber, there is a steady increase in $\varepsilon'$ with

Figure 8.4 Variation of dielectric permittivity of neoprene-rubber based composites with frequency in the S band
increase in content of nickel in the composites. The increase in $\varepsilon'$ follows the empirical relation suggested by Baziard as shown in figure 8.5.

![Figure 8.5: Variation of dielectric permittivity with volume ratio of nickel in the S band compared with Baziard's model (neoprene-rubber based composites)](image)

The behaviour of dielectric loss in neoprene based composites is different from that of natural rubber based composites. Dielectric loss of all the composite samples increases steadily with frequency and shows saturation towards the higher edge of the s band. Further, the dielectric loss in neoprene rubber based composites is much higher than that in natural rubber based composites. This high loss in neoprene rubber matrix can be observed in all frequency ranges from rf to X band. The variation of dielectric loss is depicted in figure 8.6.
8.7 Magnetic permeability of the composites

The magnetic permeability of both natural rubber and neoprene rubber based composites were measured at two frequencies in the S band. Magnetic permeability decreases with frequency. The studies reveal that the variations in magnetic permeability behaviour of both types of composites are identical as depicted in figure 8.7 and 8.8. The origin of magnetic permeability is from the embedded ferromagnetic nickel nanoparticles in the matrix and the elastomer matrix does not offer any contribution towards the magnetic properties. It was observed that the natural rubber based composites exhibit higher magnetic permeability and higher elastic modulus of natural rubber over neoprene rubber could be the reason for this. Magnetic loss also shows the same kind of change in both classes of composites samples as shown in figures 9 and 10. Due to instrument limitations the real and imaginary parts of the magnetic permeability are plotted for only two frequencies in the S band. The variation of the real part is as expected and it shows a steady decrease and this is attributed to the presence of skin effect and eddy current loss in nickel particles. Even nanometer sized particles can have these effects in a reduced scale, if not as prominent as in the case of bulk material. But it is difficult to derive any conclusion about the true nature of magnetic loss from the observed data.
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**Figure 8.7:** Variation of magnetic permeability in the S band (natural rubber based composites)

**Figure 8.8:** Variation of magnetic permeability with frequency in the S band (neoprene-rubber based composites)
8.8 Complex dielectric permittivity in X band

The variation of the real part of the relative dielectric permittivity ($\varepsilon'_r$) of natural-rubber-nickel composites is depicted in figure 8.11 and that of neoprene-rubber-nickel composites in figure 8.12. It can be observed that, in both types of composites, the permittivity of a particular composite sample remains constant for the entire frequency range of 7 – 12 GHz. Frequency dependant dispersion of permittivity is completely absent in the X band frequency. The polarization mechanism operating in the gigahertz frequency is purely electronic or orientational
with relaxation times smaller than the time period of the applied signals. Interfacial polarization, which is the basic reason for the dispersion at radio frequency (rf) regime, has no role to play in the microwave frequencies as it does not produce dispersion in $\varepsilon'_r$ because of its much smaller relaxation time. But $\varepsilon'_r$ was found to increase with the increase of mass ratio of filler in the composite as evident from figure 8.11 and figure 8.12. This phenomenon of increase in $\varepsilon'_r$ with filler concentration can be attributed to the enhancement of electrical conductivity of the composites due to the incorporation of metal particles as filler.

It can be observed from figure 8.11 and figure 8.12 that the dielectric permittivity of neoprene rubber composites is much higher than that of natural rubber composites for a given filler concentration. Neoprene rubber is made of polar molecules and the polar nature of the molecules is retained even after the cross-linking of the molecular chains. Polar molecules of neoprene can undergo orientational polarization and can give rise to enhanced polarization compared to the molecules of natural rubber and this explains its higher dielectric permittivity.

Figure 8.11: Variation of dielectric permittivity of natural rubber based composites with frequency in the X band
Figure 8.12: Variation of dielectric permittivity of neoprene-rubber based composites with frequency in the X band

Figure 8.13 and figure 8.14 depict the variation in dielectric loss ($\varepsilon_r''$) with frequency of natural-rubber-nickel and neoprene-rubber-nickel composites respectively.

In neoprene-rubber-nickel composites, as frequency increases $\varepsilon_r''$ registers a steady increase and this phenomenon is observed in all the five samples. This can be attributed to interfacial polarization and the resulting energy loss due to relaxation polarization. Even though the absence of interfacial polarization was confirmed from the absence of frequency dispersion in $\varepsilon_r'$, small amount of charge accumulation may be present around metal particles even at GHz frequencies. The electric conductance loss may also be contributing at higher frequencies and this could be the reason for enhanced loss for 100 phr sample towards the higher edge of the frequency band [24]. Also there is a steady increase in $\varepsilon_r''$ with the increase in the concentration of nickel in the composites suggesting the presence of electrical conductance loss.
Natural-rubber-nickel composites show a similar kind of variation in dielectric loss but at 8.25 GHz, there appears a relaxation peak. Orientational polarization of the polymer chains of natural rubber may have a relaxation times close to this frequency and causing the relaxation peak. But the dielectric loss increases towards the higher frequency side and increases sharply at higher loading fractions. Conductance loss due to the presence of metallic inclusions is attributed to this enhancement in dielectric loss.
8.9 Complex magnetic permeability in the X band

It was observed that the real part of complex permeability ($\mu'_r$) decreases with increase in frequency in both classes of composites. It is well-known that the permeability of ferromagnetic nickel drops sharply at gigahertz frequencies and becomes close to unity at 10 GHz [20]. The influence of skin effect and eddy current loss are present in nanometer sized particles also, in a reduced magnitude. However the decrease in $\mu'_r$ is not as sharp as the reported behavior of bulk nickel and this can be attributed to the reduced size of nickel particle. The variation of $\mu'_r$ with frequency is shown in figure 8.15 (natural-rubber-nickel composites) and in figure 8.16 (neoprene-rubber-nickel composites).

![Figure 8.15: Variation of magnetic permeability with frequency of the natural rubber based composites in the X band](image)

The magnetic loss ($\mu''_r$) increases initially and both composites show a peak at about 8 GHz and then drops slightly (figures 8.17 and 8.18). This is due to the resonant vibration of the nickel particles in the oscillatory magnetic field [24]. Further, the loss is more pronounced in neoprene based composites, even though the pattern of variation remains the same, as evident from the graphs. It appears that the neoprene matrix offers less resistance to resonant vibrations of nickel particles on application of an oscillatory electromagnetic field.
Figure 8.16: Variation of magnetic permeability with frequency of neoprene-rubber based composites in the X band

Figure 8.17: Variation of magnetic loss with frequency of the natural rubber based composites in the X band
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**Figure 8.18:** Variation of magnetic loss with frequency of the natural rubber based composites in the X band

### 8.10 Input impedance and reflection loss

The microwave absorption characteristics of a material are analyzed on the basis of its complex permittivity and permeability. Good absorbers should have low reflection coefficient and good absorption coefficient. Electromagnetic waves entering a material are absorbed effectively if there is a good impedance matching between the material and the free space and the condition for which can be written down as follows [24]

$$\frac{\mu'}{\varepsilon'} = 1$$  \hspace{1cm} (8.9)

The neoprene-nickel composites in these studies show that $\varepsilon'_r$ varies between 2.5 and 5 with respect to different loadings and $\varepsilon'_r$ varies between 1.2 and 2.2 and it is difficult to meet the condition given by equation (6) for any particular sample or in any given frequency. The propagation constant $\gamma$ of electromagnetic waves in a material is given by the relation

$$\gamma = \alpha + i\beta$$  \hspace{1cm} (8.10)

where $\alpha$ is the attenuation constant and $\beta$ is the phase constant. In terms of the complex permittivity and permeability the propagation constant is expressed by the relation...
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\[ \gamma = j \frac{2\pi f}{c} \sqrt{\mu_r \varepsilon_r} \quad (8.11) \]

Separating the real and imaginary parts of equation (11) gives the attenuation constant \( \alpha \) and phase constant \( \beta \). [25] as

\[ \alpha = \frac{\sqrt{2}}{c} \pi f \left( \sqrt{\left(\mu''_r \varepsilon''_r - \mu'_r \varepsilon'_r\right) + \sqrt{\left(\mu''_r \varepsilon''_r - \mu'_r \varepsilon'_r\right)^2 + \left(\varepsilon'_r \mu''_r - \varepsilon''_r \mu'_r\right)}} \right) \quad (8.12) \]

and

\[ \beta = \frac{\sqrt{2}}{c} \pi f \left( \sqrt{\left(\mu'_r \varepsilon'_r - \mu''_r \varepsilon''_r\right) + \sqrt{\left(\mu'_r \varepsilon'_r - \mu''_r \varepsilon''_r\right)^2 + \left(\varepsilon'_r \mu''_r + \varepsilon''_r \mu'_r\right)}} \right) \quad (8.13) \]

The input impedance of a wave absorber \((Z_{in})\) with a single layer, backed by a metallic reflector is given by the relation

\[ Z_{in} = Z_0 \sqrt{\frac{\mu_r}{\varepsilon_r}} \tanh(\gamma t) \quad (8.14) \]

Where \( Z_0 \) is the impedance of free space, and the reflection coefficient is given by the relation

\[ \Gamma = \frac{Z_{in} - Z_0}{Z_{in} + Z_0} \quad (8.15) \]

and reflection loss expressed in decibel (R) is given as

\[ R = 20 \log_{10} |\Gamma| \text{ dB} \quad (8.16) \]

The electrical wavelength \( (\lambda) \) in the material can be calculated from the phase constant \( \beta \) for any given frequency as

\[ \lambda = \frac{2\pi}{\beta} \quad (8.17) \]

At the air material interface total cancellation of reflected wave can occur by interference between the incident and the reflected waves. The condition for this can be deduced by assuming that the dielectric loss in the medium is small compared with the dielectric permittivity [25]. The equation can be expressed as

\[ t_0 = \frac{c}{4.\pi f} \frac{1}{\sqrt{\mu'_r \varepsilon'_r}} \left( 1 + \frac{1}{8} \tan^2 \delta_\mu \right)^{-1} \quad (8.18) \]

where \( \delta_\mu \) is the magnetic loss tangent.
Based on the equations given above, the propagation constant of the composites was calculated and from which the reflection loss at various frequencies were estimated. In this study the permeability and permittivity were not measured for the same frequencies, since the measurements were carried out at even and odd resonating modes of the cavity. The real part of the permittivity of a particular sample nearly remains a constant in the entire frequency range of X band and the average value of this permittivity was assumed to be valid for all frequencies and used in the calculations. The values of $\varepsilon'$, $\mu'$ and $\mu''$ were estimated for the entire range of X band in steps of 0.1 GHz by interpolating the plots with the help of a commercial computer software for extrapolation and interpolation of graphs.

![Reflection loss plot](image)

*Figure 8.19: Reflection loss with frequency in natural-rubber-nickel composites*
**Figure 8.20:** Variation in thickness with frequency corresponding to complete cancellation of waves by interference in natural-rubber-nickel composites

**Figure 8.21:** Reflection loss with frequency in neoprene-rubber-nickel composites
Figure 8.22: Variation in thickness with frequency corresponding to complete cancellation of waves by interference in neoprene-rubber-nickel composites

For natural rubber based composites the reflection loss with frequency does not show any resonance and shows a steady decrease with increase of frequency (figure 8.19). A maximum of -10.5 dB loss is registered for a frequency of 12 GHz. The thickness corresponding to the cancellation of reflected waves also decrease steadily with frequency as depicted in figure 8.20.

For neoprene-rubber based composites the variation of reflection loss with frequency is shown in figure 8.21. It can be observed from the graphs that the frequency corresponding to maximum reflection loss is shifted towards the lower frequency side as the concentration of nickel particles increases in the composites. This result is consistent with previous reports.

The thickness for cancellation of reflected wave is calculated and the results are shown in figure 8.22. There is a steady decrease in thickness corresponding to total annihilation of waves by interference.

8.11 Conclusion

The complex dielectric permittivity and magnetic permeability of natural-rubber-nickel and neoprene-rubber-nickel nanocomposites were evaluated in the X-band of microwave
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frequencies from 7 GHz to 12 GHz. The real part of dielectric permittivity does not change with frequency but registers a steady increase with increase in the concentration of loading of nickel nanoparticles. The dielectric loss shows an increase with frequency in general and for natural rubber based composites there appears a shallow relaxation peak around 8.25 GHz. In both these of composites, dielectric loss increases with increase in the concentration of filler which could be the result of conductance loss. Real part of permeability shows a steady decrease with increase of frequency and the behaviour is identical in both types of composites. The magnetic loss shows a small relaxation at 8 GHz and then decreases. Magnetic resonance of ferromagnetic nickel particles is attributed to the observed peak in magnetic loss The input impedance and reflection loss were calculated and for neoprene rubber, resonance is observed and found to shift towards higher frequency side with the increase in concentration of nickel nanoparticles.
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