Evaluation of hysteresis loss for rubber-ferrite composites and estimation of magnetostrictive constant ($\lambda_s$) from loop parameters

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Rubber ferrite composites (RFCs) are flexible elastomer magnets. The flexibility and mouldability of these magnets are an added attraction as far as applications are considered. They are also potential microwave absorbers. RFCs can be prepared by the incorporation of ferrite fillers in rubber matrices. The estimation of saturation magnetostrictive constant ($\lambda_s$) for RFCs poses always a problem since they are flexible and elastic in nature. So, alternate methods for the estimation of $\lambda_s$ is necessary. This paper discusses the estimation of $\lambda_s$ for ferrite fillers as well as for RFCs from hysteresis loop parameters by employing a simple relationship. The magnetostrictive constant ($\lambda_s$) for the ceramic fillers as well as the RFCs are estimated and are presented here. The results are compared.

1 Introduction

Magnetostriction is the phenomena exhibited by ferro and ferrimagnetic materials. The state of strain of the magnetic sample is dependent on the direction and the extent of magnetization. The strain caused by magnetization from the demagnetized state to the magnetic saturation is denoted by the magnetostrictive constant $\lambda$. Magnetostriction appears because a crystal is always deformed spontaneously in the direction of magnetization in each domain. As the direction of spontaneous magnetization is changed by technical magnetization, the direction of spontaneous strain of the crystal also changes, resulting in a change in the shape of the crystal as a whole. Normally, $\lambda$ is represented by the change in length to the original length and can be written as $\Delta l/l$.

This particular phenomena has important applications in making magnetostrictive transducers, which are used extensively in sonar devices. Currently, the phenomena of magnetostriction is used in analog positioning problems. They are also used in acoustic delay lines, sonar devices and oscillators.

The theory of magnetostriction is complex and models are generally related to those of magneto crystalline anisotropy since both these phenomena are due to spin-orbit coupling. The microscopic origin of magnetostriction in spinel ferrites is considered to be due to the strain potential and spin-orbit, intra spin-spin interactions, anisotropy exchange and the strain modulation of the dipolar interaction. The magnetostrictive strains are usually so small that, they can only be measured if adequately magnified by mechanical electrical or optical devices.

The precise determination of $\lambda$ or $\lambda_s$, the saturation magnetostriction constant is important. This is significant not only from the applications point of view but this also provides information as regards the nature of the domain or the domain dynamics under the application of an external magnetic field. There are several experimental methods available for the determination of $\lambda_s$. Of them, $\lambda_s$, evaluation by strain gauge methods, optical dilatometry, capacitance method and X-ray analysis need special mention. In most of the cases, these experiments are able to predict the magnitude and not the sign.

RFCs are composites containing elastomer matrices and they are elastic in nature. Under normal circumstances, they cannot be made transparent and hence optical methods cannot be employed to determine $\lambda_s$. Since they are elastic in nature, employment of strain gauge techniques for the evaluation of $\lambda_s$ also does not give accurate values. This is because, the adhesion of strain gauges to the composites, requires a small force and...
this might already cause a strain to the sample under test even before the application of a magnetic field. So, results obtained from these techniques could be erroneous, unless appropriate corrections are incorporated. This is often time-consuming and not fail-proof. This lead us to search for simpler and quicker methods to estimate $\lambda$.

2 Experimental Details

2.1 Synthesis of $\text{Ni}_{1.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ and preparation of rubber-ferrite composites

Nickel-zinc-ferrites having the general formula $\text{Ni}_{1.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ for $x$ varying from 0 to 1 in steps of 0.2, were prepared by the ceramic techniques, for the appropriate amounts of precursors necessary for the synthesis of mixed ferrites were weighed accordingly, to molecular weight considerations and pre-fired at 500 °C and then finally sintered at 1100°C for several hours. The details are cited elsewhere\(^{(2-9)}\). These powders were then characterized by using XRD and VSM. These pre-characterized powder samples were then incorporated in a natural rubber matrix for various loadings of the magnetic filler and according to a specific recipe. Ferrite fillers were loaded in phr (parts per hundred gram of rubber). RFCs containing 20, 40, 60, 80, and 120 phr were prepared by loading appropriate amount of fillers. The mixing details are given elsewhere\(^{(2-9)}\). These were then compounded into sheets of required thickness. These samples were then subjected to further studies.

2.2 Structural characterization

Structural parameters were determined and the monophasic nature of the fillers was ensured by employing X-ray powder diffractometer (XRD). The diffractogram of these powder samples were recorded on a Philips (PW 1130) X-ray diffractometer using CuK$_\alpha$ radiation, $\lambda=1.5418\text{Å}$.

2.3 Magnetic measurements

Room temperature magnetic measurements for the ceramic filler and the RFCs were carried out by employing a vibrating sample magnetometer model 4500 from EG & G PARC. Parameters such as, saturation magnetisation ($\sigma_s$), remanent magnetisation ($\sigma_r$) and magnetic field strength (coercivity) $H_c$ were obtained from these magnetic measurements.

3 Estimation of $\lambda$ from Magnetic Measurements

The saturation magnetostriction of a polycrystalline specimen depends on the properties of its individual crystals and the way in which they are arranged. The energy loss is attributed to the creation and annihilation of the domain wall surface during its displacement. This means that, the value of saturation magnetostriction depends on the energy loss. In polycrystalline materials, the grains are oriented randomly. The saturation magnetostriction obtained here is an average over the orientations.

It is known from first principles that, simple relationships exist between $\lambda$ and the total energy loss ($W_t$) when considering the hysteresis loop of a ferro or ferrimagnetic material. Normally, $W_t$ is related to $M_t$ and $H_c$ and they are, in turn, determined by loop constants. Here, the shape of the hysteresis loop play a very important role and so, do the ratio $M_t/M_c$. In literature\(^{(21)}\), three types of materials are mentioned and they are characterized by loop constants. The equations connecting the hysteresis loss with $M_t$ and $H_c$ are different for different materials and they are the following. For single domain fine particles:

$$W_t = 1.98 \cdot M_t \cdot H_c$$ \quad (1)

For fine particles with multi domain structure:

$$W_t = \frac{3.21 M_t H_c}{1.3}$$ \quad (2)

For a cubic magnetic material:

$$W_t = \frac{4.3 \times M_t \times H_c}{1.2}$$ \quad (3)

It is imperative that, the exact nature of the loop and the loop constants of the system under investigation is to be determined. For this, the observed $W_t$ from hysteresis loop parameters (VSM measurements) was fitted for various combinations of $M_t$ and $H_c$ [Eqs (1) to (3)] and it was found that, the calculated $W_t$ and the observed $W_t$ obey the equation, $W_t = \frac{4.3 \times M_t \times H_c}{1.2}$. This fact was ascertained by calculating $W_t$ and comparing it with the observed $W_t$, obtained from VSM measurements. These calculations were extended for the $\text{Ni}_{1.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ ceramic series as well as for the ferrite incorporated composites. Figs 1 and 2 show...
the nature of $W_6$ with $x$ for these samples. It is to be noted that, the $W_6$ calculated by using Eq. (3) is in good agreement with the observed $W_6$ from VSM. So, it is right to assume that, the present mixed ferrite system and the RFCs under investigation also came under the third category as described in literature\(^\text{21}\) and they are governed by Eq. (3).

However, this relationship requires a knowledge of $\sigma_0$. One of the two extreme assumptions are possible like, stress is uniform throughout but strain varies from grain-to-grain or, strain is uniform and stress varies. It has been reported in the literature that, $\sigma_0$ is universal in nature, especially, for polycrystals and it is of the order of $10^4$ N/m\(^2\) (Ref. 22). The origin of $\sigma_0$ is assumed to be a constant which is not clear and dealt very scarcely\(^\text{22}\). This value of $\sigma_0$ was employed for the estimation of $\lambda$. Relationships connecting the saturation magnetostriction constant $\lambda$, and the energy loss $W_6$ is also possible. In such a case, it is to be assumed that all process of magnetisation reversal take place exclusively through the displacement of 180° walls. $\lambda$ is related with $W_6$ by the equation:

$$W_6 = 4.3 \pi \lambda \sigma_0$$

(4)

Here, hysteresis loss is expressed in terms of magnetostrictive anisotropy because: irreversible process of magnetisation is very much affected by the fluctuations of internal stress, rather than the magnitude of crystal anisotropy. From Eq. (4), $\lambda$ can be deduced as follows:

$$\lambda = \frac{W_6}{4.3 \pi x \sigma_0}$$

(5)

where, $\lambda$ is the magnetostrictive constant $W_6$ is the hysteresis loss factor and $\sigma_0$ is the internal stress.

4 Results and Discussion

For the purpose of comparison and checking the validity of this simple relationship, data available from literature on similar systems, namely, Ni\(_{1-x}\)Zn\(_{x}\)Fe\(_2\)O\(_4\) were made use of $\lambda$, was calculated for Ni\(_{1-x}\)Zn\(_{x}\)Fe\(_2\)O\(_4\) with various $x$ for two series of samples fired at different temperatures and they are shown in Fig. 3. This was done by employing the reported values of $M$, and $H$, and substituting these values in Eq. (5). The value of $\lambda$, for nickel-zinc-ferrite series was experimentally found out by Smit & Wijn\(^\text{22}\) and compared with the calculated values. Table 1 shows a gist of the $\lambda$, values reported by various scientists for various ferrites. $\lambda$, reported for Ni\(_{1-x}\)Zn\(_{x}\)Fe\(_2\)O\(_4\) by using experimental techniques was also plotted for
different $x$ and is shown in Fig. 3. These were also compared with the calculated $\lambda$, (using the reported $M$, and $H$, values of Ni$_{1-x}$Zn$_x$Fe$_2$O$_4$ ferrite samples by employing Eq. (5). An increase in $\lambda$ was observed with increase in zinc concentration. Etienne du Tremolet de Lacheisserie has found out the magnetostriction coefficients for Ni$_{1-x}$Zn$_x$Fe$_2$O$_4$ at 77 K and it is found to be lying between $105 \times 10^{-6}$ and $63 \times 10^{-6}$ (Ref. 24). He also analyzed the $x$ dependence of $\lambda$ for Ni$_{1-x}$Zn$_x$Fe$_2$O$_4$. The $\lambda$ for NiFe$_2$O$_4$ at room temperature was first reported by Smit & Wijn, and it is found to be $-26 \times 10^{-6}$ (cf Table 1). But, Murthy & Rao$^{25}$ report that, for NiFe$_2$O$_4$, the value of $\lambda$ is $-24 \times 10^{-6}$. These discrepancies could be because of the influence on the preparative conditions on the value of $\lambda$. That is, as sintering temperature increases, the absolute value of $\lambda$ decreases. This decrease was correlated with the appearance of Fe$^{3+}$ in the nickel ferrite. Since Fe$_3$O$_4$ exhibit a positive $\lambda$ value, hence, any appearance of Fe$^{3+}$ in the nickel-ferrite markedly reduces the absolute value of its $\lambda$, coefficient$^{24}$.

This estimation only provides the magnitude of the magnetostriction and not the sign. This is one of the drawbacks of this method of estimation. However, a survey of the literature reveals that, except magnetite most of the ferrites exhibit a negative magnetostrictive constant. So, intuitively one can indirectly gain an idea about the sign of $\lambda$. Moreover, a knowledge of anisotropy constants can also give indications about the sign. So, going by the vast experimental data available on similar systems sign can be attributed to the $\lambda$.

Table 1 — Linear magnetostriction coefficient for some mixed ferrites (reported by Smit & Wijn$^{23}$)

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>$\lambda \times 10^6$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni$<em>{0.1}$Zn$</em>{0.04}$Fe$_2$O$_4$</td>
<td>-5</td>
</tr>
<tr>
<td>Ni$<em>{0.5}$Zn$</em>{0.5}$Fe$_2$O$_4$</td>
<td>-11</td>
</tr>
<tr>
<td>Ni$<em>{0.6}$Zn$</em>{0.4}$Fe$_2$O$_4$</td>
<td>-16</td>
</tr>
<tr>
<td>Ni$<em>{0.1}$Zn$</em>{0.5}$Fe$_2$O$_4$</td>
<td>-21</td>
</tr>
<tr>
<td>NiFe$_2$O$_4$</td>
<td>-25</td>
</tr>
<tr>
<td>Fe$_3$O$_4$</td>
<td>40</td>
</tr>
<tr>
<td>CoFe$_2$O$_4$</td>
<td>-110</td>
</tr>
<tr>
<td>MnFe$_2$O$_4$</td>
<td>-5</td>
</tr>
<tr>
<td>MgFe$_2$O$_4$</td>
<td>-6</td>
</tr>
<tr>
<td>Li$_{0.5}$Fe$_2$O$_4$</td>
<td>-8</td>
</tr>
</tbody>
</table>

Fig. 3 — $\lambda$ versus composition for Ni$_{1-x}$Zn$_x$Fe$_2$O$_4$
The graph depicting the variation of $\lambda_s$ with $x$ for samples fired at higher temperature indicates that $\lambda_s$ is higher for particles with lower $H_c$. The variation pattern of $\lambda_s$ with $x$ is the same for $\text{Ni}_1\text{Zn}_x\text{Fe}_2\text{O}_4$ for both calculated and measured values from literature. However, barring compositions at $x = 0$ and $x = 1$ (cf. Fig. 3), the variation pattern for $\lambda_s$ versus $x$ is almost the same. This could be because no $\lambda_s$ values were reported earlier for $\text{ZnFe}_2\text{O}_4$ and it was assumed to be an anti-ferromagnetic material. In the present study, we observed a net magnetization of 3.118 emu/gm for fine particle zinc-ferrite and this behaviour has been discussed. It is also possible that, at $x = 0$, the $\text{Ni}^{2+}$ will be occupying the tetrahedral sites instead of octahedral sites. This observation is supported by complimentary evidences and is reported by Chinnasamy et al. The estimated $\lambda_s$ for the samples under investigation is less than that of the reported.

The same principle was applied to estimate $\lambda_s$ for RFCs. The $\lambda_s$ values of rubber-ferrite composites are larger, compared to that of its ceramic component. The compositional dependence of $\lambda_s$ for RFCs shows same pattern as that observed for ceramic fillers. Fig. 4 shows the variation of magnetostriction constant with zinc concentration for RFCs. It is also to be noted that, the saturation magnetostriction decreases with increase of weight fraction of the magnetic filler. The details are shown in Fig. 5. The $\lambda_s$ of RFCs exhibit a cumulative property of the matrix and the filler. The $\lambda_s$ of RFCs is characteristic of a dilute system. The values range from $6.98 \times 10^{-5}$ to $0.1356 \times 10^{-5}$. This is because, the matrix is non-magnetic and are assumed that there is no filler-matrix interaction. Moreover, magnetization measurements carried out on RFCs show that, the $H_c$ of the composites and $H_c$ of the filler, are the same. So, naturally, the $\lambda_s$ is determined by the amount of the filler or rather the $M_s$ of the filler.

It is to be noted that, the employment of Eq. (5) provides only approximate values for $\lambda_s$.
ceramic as well as the composites can be determined experimentally, a comparative study can be carried out. This, together with some modelling and simulation, can throw light in the origin of $\sigma_0$ and thus, could even tell us, whether there is a large change in $\sigma_0$ or this could be assumed as a constant. This estimation does not provide the sign for the estimated $\lambda_0$. This is one of the drawbacks of this method of estimation of $\lambda_0$.

5 Conclusion

Estimation of magnetostriction constant $\lambda_0$ was carried out by utilizing the hysteresis loop parameters and employing a simple relationship. The magnetic parameters obtained from the vibrating sample magnetometer is used to estimate the magnetostriction constant ($\lambda_0$). One of the main drawbacks of this estimation is that, the sign of $\lambda_0$ cannot be predicted directly. From the literature, it can be seen that, except magnetite most of the ferrites exhibit a negative magnetostrictive constant. Results on the estimation of $\lambda_0$ for the ceramic nickel-zinc-ferrite lies in the range $13.44 \times 10^{-5}$ to a lower value of $0.51 \times 10^{-5}$. The estimated $\lambda_0$ values of RFCs are found to be less, compared with that of ceramic ferrite samples.

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References