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Anomalous dielectric behavior of some ferrites[a)]

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The dielectric behavior of some powdered polycrystalline samples has been studied in the frequency range of 200 Hz–100 kHz. It is shown that the dielectric behavior in these systems below the Curie temperature is not purely relaxational in its character and cannot be described by any of the models of the dielectric relaxation hitherto put forward. It is also shown that "isolation" of the particles in the powder samples plays a very important role. The origin of this abnormality is thought to be due to the mechanical resonance arising out of the magnetostrictive property of the material.

The dielectric investigation of ferrites in the low-frequency region and at high temperatures reveals a relaxation often identified as the Maxwell–Wagner (MW) polarization which was attributed to their heterogeneous structure.[1-5] The often used interpretation consists of assuming a distribution of relaxation times[6] (or a distribution of layer properties as the cause of the broadening of the relaxation process[7]). In the present communication, we put forward preliminary evidence of some abnormality in the dielectric behavior of some polycrystalline ferrites which cannot be explained by any of the relaxational models. The ferrites used in the present study were prepared by the conventional double-sintering process. The chemical analysis was done by using an atomic absorption-type Carl-Zeiss spectrometer. Mass spectrum analysis was also done to confirm the chemical composition. The x-ray diffraction pattern of the ferrite samples was obtained using Co$K\alpha$ radiation and the lattice constants were calculated in order to confirm the single-phase spinel nature. The dielectric measurements of all these ferrites in the radio frequency range (200 Hz–100 kHz) were carried out with a general radio 1615 AP capacitance test assembly and were taken in vacuum. Pellets of the ferrite powder samples were made using a hydraulic press at 10 tons/cm^2 pressure. The chemical formulas, Curie temperatures, and the other dielectric details are given in Table I.

A known amount of nickel-zinc and nickel-zinc-manganese ferrites were mixed in a molten analar grade paraffin wax with constant stirring. These were cut into pellets and were used for dielectric measurements at room temperature. The density of these wax ferrite mixtures was found to be 1.1–1.3 g/cm^3 while the density of the polycrystalline ferrite is 5.1–5.3 g/cm^3 (the density of the pellets made out of the ferrite powders was around 4.8 g/cm^3). The accuracy in the measurement of the dielectric constant is 2% and 5% in the loss. Throughout the discussion we prefer to discuss the dielectric behavior of all the samples in the form of the Cole–Cole (CC) plots as shown in all four figures. The peculiarity of the dielectric behavior of all these ferrite samples in their ferromagnetic phase is that the spectral shape cannot be explained by any of the spectral shape functions[8,9] suggested so far to describe the dipolar relaxation process in other materials, thus indicating that the behavior is not "pure" relaxational in its character. A few examples of this behavior can be very clearly seen in the case of Ni-Zn 1 [Fig. 1(a)] and Ni-Zn-Mn 1 [Fig. 2(b)]. In the case of Ni-Zn 1 and Ni-Zn-Mn 1 the curves corresponding to the dielectric loss versus frequency in a double logarithmic plot shows slopes of the curves at the exceeding ± 1 intermediate frequencies indicating an unmistakable sign of a resonance process[6] (not shown in the figures). This behavior can be seen as a "bulge" at the intermediate frequencies in the CC plots. [see Figs. 1(a), 2(a), and 2(b)]. From the discussion of Jonscher (Ref. 8), the "bulge" at intermediate frequencies in the above CC plots is confirmed as not due to the low-frequency dispersion (LFD) behavior associated with a narrow barrier region in series with a less dispersive bulk material). The above abnormality increases with temperature [Fig. 2(a)] and decreases with the dilution of the Ni content (in the present compositional range) and completely disappears in

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TABLE I. The chemical formulas, Curie temperatures, and the dielectric details of the samples used in the study.

Sample No.	Sample	Chemical formulas	Curie temperature[a] T_c (°C)[a]	Experimental values		
				ϵ_∞ [b]	$\epsilon_0 - \epsilon_\infty$ [b]	ϵ''_{max}
1	Ni-Zn 1	$Ni_{0.6}Zn_{0.33}Fe_2O_{4\pm}$	~425	1.6	23.9	16.5
2	Ni-Zn 2	$Ni_{0.27}Zn_{0.8}Fe_2O_{4\pm}$	~70	4.5	16.3	4.6
3	Ni-Zn 3	$Ni_{0.13}Zn_{0.87}Fe_2O_{4\pm}$	~0	7.5	24.5	6.3
4	Ni-Zn-Mn 1	$Ni_{0.17}Zn_{0.44}Mn_{0.29}Fe_2O_{4\pm}$	> 100	1.65	23.1	14.0
5	Ni-Zn-Mn 2	$Ni_{0.27}Zn_{0.44}Mn_{0.29}Fe_2O_{4\pm}$	> 100		···	0.207
6	Ba	$BaFe_{12}O_{19}$	~450	3.0	42.0	22.5
7	Co-Zn	$Co_{0.79}Zn_{0.21}Fe_2O_4$	> 700	5.5	23.0	9.4
8	Wax + Ni-Zn 1	···		6.1	5.9	1.75
9	Wax + Ni-Zn 2	···	···	4.9	4.25	0.70
10	Wax + Ni-Zn 3	···	···	5.225	3.625	0.875
11	Wax + Ni-Zn-Mn 1	···	···	5.2	6.6	1.55
12	Wax + Ni-Zn-Mn 2	···	···	4.5	3.3	0.3

[a] Taken from Ref. 14.
[b] Extrapolated values.

Ni-Zn 3 [Fig. 1(b)], which happens to be in a paramagnetic state. In order to show that the abnormality is not due to an artifact of the experiment, we have studied the primary relaxational behavior of one polyurethane sample. As can be seen in Fig. 3, there is no "abnormality"[8] and the experimental points can be fitted to the popular Hevriliak–Negami equation.[9]

Since the response to a sinusoidal electromagnetic field is under consideration, it is logical to enquire about both the electrical and the magnetic processes and the interaction between the two. Regarding the magnetic processes in the present experimental range of frequencies, neither dimensional resonance nor skin effect play a part here.[10] From the studies of earlier workers,[6,11] for the concentrations of Ni and Zn in the Ni-Zn samples used in the present study, the saturation magnetization and the magnetic permeability decrease from Ni-Zn 1 to Ni-Zn 3, which may play a part in the gradual absence of the abnomality. The previous studies[11] on Ni-Zn 1 indicated a rise of saturation magnetization with temperature for the temperature range given in Fig. 2(a), which perhaps might have influenced the rise of abnormality with increase in temperature. It can also be noted that there are reports[12–15] of magnetic relaxations in Ni-Zn ferrites in the present experimental range of frequencies and temperature.

To get some further insight into the picture, we studied the dielectric behavior of the ferrite dispersed in a medium of wax. As expected, addition of wax has decreased the dielectric strength ($\epsilon_0 - \epsilon_\infty$) (Table I) as in the case of any nonferromagnetic material, but what is more interesting is the change in the shape of the CC plots with a "near" absence of the "abnormality" (Fig. 4). Hence, we are tempted to attribute the abnormal behavior in the pure ferrites to the mechanical resonance of the system resulting from the magne-

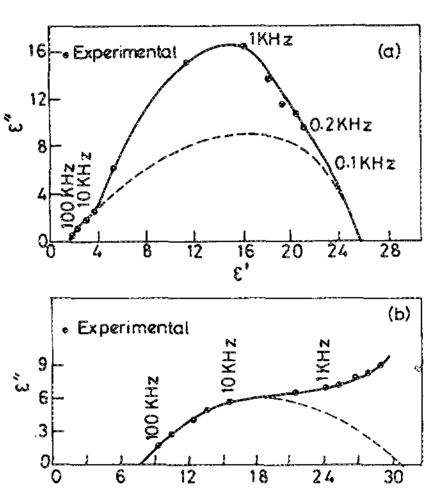

FIG. 1. (a) The complex plane representation of the dielectric behavior of Ni-Zn 1 at 50 °C. (b) The complex plane representation of the dielectric behavior of Ni-Zn 3 at 50 °C.

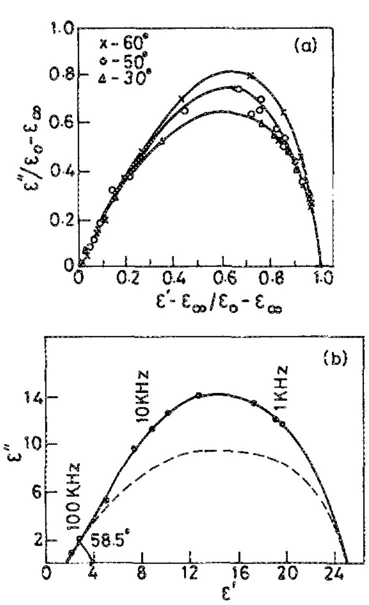

FIG. 2. (a) The complex plane representation of the dielectric behavior of Ni-Zn-Mn 1 at 50 °C. (b) Temperature variation of the dielectric behavior of Ni-Zn 1 shown as a normalized complex plane plot.

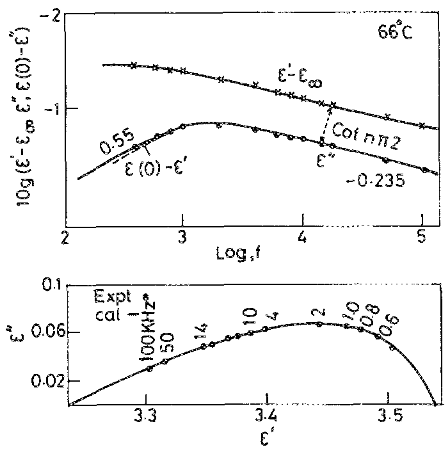

FIG. 3. Dielectric behavior of one polyether-urethane sample. (a) Double logarithmic representation of the real and imaginary parts of $\epsilon^* - \epsilon_\infty$ with frequency. (b) Complex plane representation for the same.

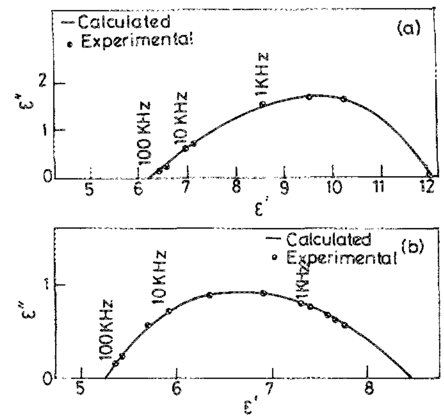

FIG. 4. (a) Complex plane representation of the dielectric behavior of wax-Ni-Zn 1 at room temperature. (b) Complex plane representation of the dielectric behavior of wax-Ni-Zn 3 at room temperature.

toelastic coupling (magnetostriction) in the material,[16] with wax offering a high damping to this process in the wax-ferrite composite resulting in a near-relaxational behavior. The anomalous behavior which is under discussion can also be found in the cases of Fe_3O_4 at 111 K and $Mn_{0.6}Fe_{2.4}O_4$ at 88 K studied by the earlier workers,[2,4] which seemed to have escaped their attention.

As it is very well known that the dielectric magnetic and mechanical relaxations are very much coupled to each other in every material as discussed by Hill and Jonscher elsewhere,[17,18] the peculiarity one must face in the case of ferrites is that the putative magnetic loss and the dielectric loss appear to have closely allied relaxation times and it would be difficult to consider that one of these is due to an internal mechanism and the other to an external MW process because all the fundamental properties like magnetostriction, anisotropy, saturation magnetization, domains, grains, and Curie point and conductivity, etc., are all coupled to each other.[11] Hence, there is no reason to believe that the dielectric behavior in the present ferrites corresponds to two independent processes. The scene is more complicated with the possibility that there may exist a distribution of resonance frequencies[11] associated with the distribution in the grain sizes.

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