Elastic behaviour of Sn doped Ni-Zn ferrites

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Abstract- The polycrystalline Sn added Ni-Zn ferrites were prepared using the conventional sintering method. The monophasic nature of the samples was characterized using X-rays. The elastic behavior has been measured in the temperature range 80-410ºC using the composite oscillator method. The values of Young's and shear modulus are corrected to theoretical density using the method of Mackenzie method. The variation of Young's and rigidity modulus with temperature follows Watchman's equation and a linear equation respectively.

Index Terms- ferrites, XRD, composite oscillator method, elastic properties

I. Introduction

Ni-Zn ferrites are well known materials used for applications in the megahertz frequency range [1, 2]. Efforts are being made to develop low power loss Ni-Zn ferrites operating at higher frequencies in conformity with the demands arising from miniaturization of electronic devices. The performance ferrites are improved through better mechanical properties. In order to increase usage of Ni-Zn ferrites at high frequency, the properties may be optimized by the addition of Sn.

An extensive study of the composition, magnetic field and temperature dependence of the elastic behaviour of Cobalt- Zinc [1], Nickel-Zinc[2] and Magnesium-Manganese [3] ferrites has yielded very interesting results. In continuation, it was thought desirable to undertake a study of the dependence of the elastic properties of the Sn doped NiZn ferrites on composition and temperature. The obtained results of such a study are presented in this communication.

II. EXPERIMENTAL

Sn doped Ni-Zn ferrites having the composition $Ni_{0.4}Zn_{0.6}Sn_xFe_{2-x}O_4$ (x = 0.02, 0.04, 0.06, 0.08 and 0.1) were prepared by double sintering method, starting with highly pure (98.5%) chemicals. The powder were mixed in a ball mill for 6h and then pressed into toroidal and disks forms at 190 Mpa pressure. The pressed samples were calcinated at 720ºC/2h. The final sintering was carried out at 930^o C/4h followed by slow cooling in the furnace with about 10ºC/min.

III.RESULTS AND DISCUSSIONS

The sintered samples were characterized using X-ray diffraction and it was found that the samples were monophasic. The bulk density of the samples was determined from volume weight measurements in air at room temperature. The values of densities were found to increase with an addition of Sn to Ni-Zn ferrites. With the help of X-ray and bulk densities of the samples, the percentage of porosity was estimated and is found to be in between 10-20%. Using the XRD data, lattice constant (a) of all the samples under investigation was estimated and it is found that the lattice constant varies linearly with an increase of Sn content in Ni-Zn ferrites (Fig.1).

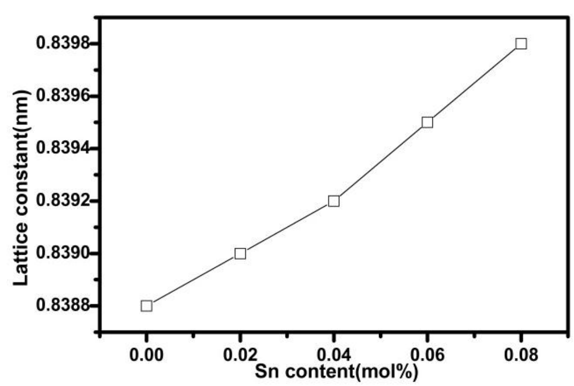

Fig.1. Lattice constant versus Sn content

The elastic behaviour of Sn doped NiZn ferrites have been measured using a piezoelectric composite oscillator method [4]. The appropriate quartz crystal transducer was cemented to the specimen with phenyl salicylate and two parts of sodium silcofluoride mixed with one part of barium sulphate in one drop of water glass for room and high temperature measurements, respectively. For the Young's modulus (E) measurements, the piezoelectric crystals were cut to oscillate at a resonance frequency of 120. 455 ± 0.002 kHz in longitudinal mode. Shear modulus (G) was measured on the same specimen sections, using shear crystals oscillating at approximately 165.215 ± 0.006 kHz. The Young's and shear moduli were then determined in the usual way using the expressions:

Table 1: Room temperature elastic moduli data.

Sample No.	Sintering Temp /Time (^{o}C)/ (hr)	Lattice constant a (nm)	Bulk density	X-ray density	E (10^{11} dy/cm^2)	G (10^{11} dy/cm^2)	κ (10^{11} dy/cm^2)	σ
0.00	930/4	0.8388	4.35	5.48	18.02	6.43	30.41	0.4
0.02	930/4	0.8390	4.64	5.57	19.47	7.01	33.04	0.4
0.04	930/4	0.8392	4.97	5.66	19.69	7.04	27.05	0.38
0.06	930/4	0.8395	5.1	5.75	20.79	7.67	23.77	0.35
0.08	930/4	0.8398	5.21	5.84	22.5	8.33	25.75	0.35

$E = (\lambda_l f_1)^2 \rho$, $G = (\lambda_S fs)^2 \rho$, where ρ density, λ the wavelength, f the resonant frequency and subscripts l and s refer to longitudinal and shear waves, respectively. The fundamental resonant frequency of every specimen was adjusted within 0.2% of that of the composite-bar resonator by careful polishing of the end faces of the specimen. The composite resonator was mounted in a metal chamber evacuated at 10^{-4} torr. The accuracy of measurement of E and G are 0.1%.

The elastic constants such as Young's (E) and shear modulus (G) were measured at room temperature for all samples and obtained results presented in the Table 1. The other constants, such as bulk modulus (k) and Poisson's ratio (σ) were computed using the general relationships: $K = E/ 3(3G – E)$, $\sigma = E/ 2G – 1$ and results are given in the table. It can be seen from the table that the values of Young's and shear modulus are found to increase with an increase in an addition of Sn to NiZn ferrites. This shows that the atomic binding strength has been decreased with an increase of addition of Sn to NiZn ferrites. The value of Poisson's ratio is found to be same for all the samples under investigation. The average value of Poisson's ratio is 0.31.

The elastic behaviour of the samples has been determined over the temperature range of 300 – 450°C. Fractional variation in frequency was measured and related to fractional modulus variations through the expansion coefficient by

$$E_T \text{ or } G_T / E_{RT} \text{ or } G_{RT} = (1 + \alpha \Delta T)^{-1} \quad (f_{ST}/ f_S)^2 \qquad \ldots\ldots \ (1)$$

Where E_{RT}, G_{RT} and f_S are Young's, shear modulus and resonant of the specimen at the reference temperature (room temperature or 300K), E_T or G_T and f_{ST} are the same quantities at any other temperature, T is the difference between the measured and reference temperature, and α is the coefficient of linear expansion over the measured temperature range. The temperature of the sample was measured with the help of Cr-Al thermocouple with an accuracy of ± 0.1 K.

The values of the coefficient of thermal expansion (α) for the present specimens are measured using the dial gauge method. It is found that the variation of coefficient of thermal expansion (α) with temperature is found to be same for all samples showing a peak at the Curie temperature of the sample. The value of Curie temperature of the present samples is 380ºC. The following equation is formed using the least square method and the same was used in the Eq. 1.

$$\alpha = 6.412 \times 10^{-6} + 0.64 \times 10^{-8} (T) + 0.382 \times 10^{-11} (T^2) \qquad (2)$$

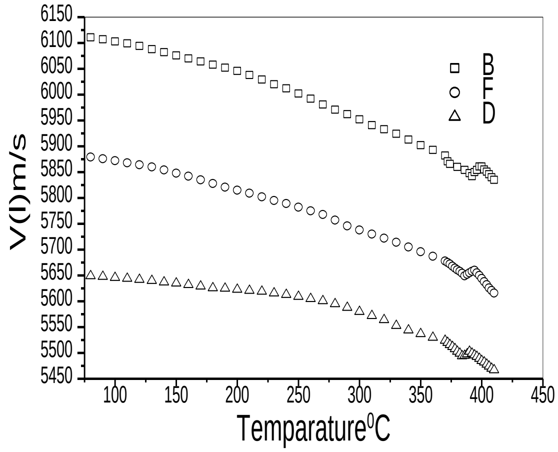

Fig 2: Temperature variation of Vl for Sn doped NiZn ferrites

Figs. 2 and 3 show the temperature dependence of longitudinal velocity (V_l) and shear velocity (Vs) for three ferrites under investigation. The values of V_l and Vs are obtained with the help of elastic constants data. It can be seen from the figure that the values of V_l and Vs decreases continuously with an increase of temperature (T) and reach a minimum temperature, called T_1. Then the value of V_l and Vs increases with temperature and attaining a maximum value at temperature T_2. Thereafter V_l and Vs decreases with further increase of temperature up to 430ºC. No hysteresis has been observed in these samples during cooling. More or less similar behaviour has been observed for all other samples. Murthy et.al [5, 6] observed a similar anomalous behavior in elastic properties of polycrystalline Ni-Zn and Co-Zn ferrites in the vicinity of the Curie temperature. A similar variation of Young's modulus with temperature also has been observed by Noikov et.al [7] in the case of NiCaMn ferrites. Kawai et.al [8] have made an extensive study of the temperature variation of Young's modulus in the case of single crystal $Mn_{0.53}Zn_{0.35}Fe_2O_4$ and observed an anomalous behavior near the Curie temperature.

The anomalous behaviour of elastic modulus with temperature observed in the vicinity of Curie temperature (T_C) for polycrystalline ferrites qualitatively can be explained by considering the temperature variation of magnetic crystalline anisotropy energy. The temperature variation of the magneto-crystalline anisotropy (k_1) for Sn doped NiZn ferrites, has been measured by Vasudev [9] and concluded that k_1 becomes zero at 370ºC, i.e., at a few degrees below the Curie temperature (380ºC). It can be seen from Fig.2 that the V_l for the ferrites attains minimum and maximum values at 370ºC (T_1) and 380ºC (T_2), respectively. Thus, the temperatures T_1 and T_2 coincide with the temperatures at which the magneto-crystalline anisotropy constant becomes zero and at the Curie temperature of the presently investigated ferrites. The magneto crystalline anisotropy (k_1) can be considered as a measure of the magnetic energy barrier to the movement of domains in magnetic materials. The domains will be free to move at a temperature where $k_1 = 0$, the substance undergoes a minimum strain for a given stress and Young's modulus/or shear modulus approaches a minimum value due to the modulus effect as a result of domain wall motion. As the temperature of the sample is increased beyond this temperature, for a given stress, the strain increases upto Curie point. In other words, the ultrasonic velocity increases and attains a maximum at Curie temperature. Beyond this temperature the value of V_l and Vs decreases with an increase of T, since at Curie temperature the ferrite loses its spontaneous magnetization and becomes paramagnetic.

In order to get the complete information about anomalous behavior, a study of the variation of V_l and Vs with temperature in a saturation magnetic field (380 mT) has been carried out by us [10]. It was found that the anomalous behavior observed in the vicinity of Curie temperature disappears and the values of V_l and Vs decreases continuously with an increase of temperature. This is because the domain wall movement is 'frozen' completely by the application of the saturation magnetic field.

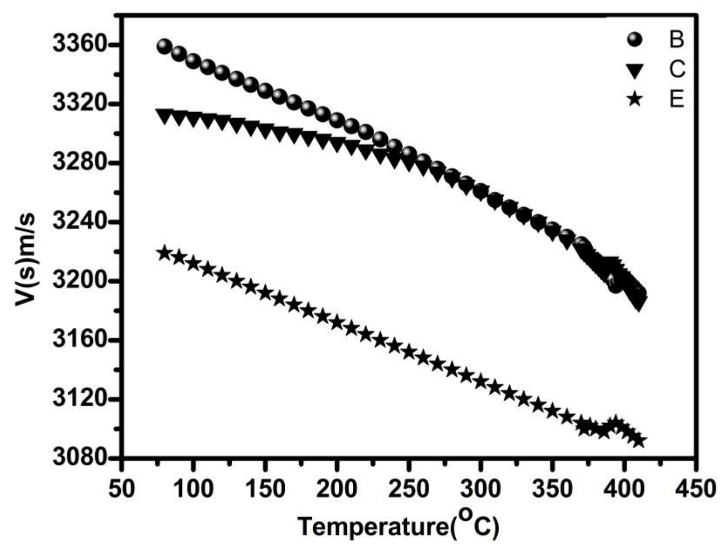

Fig.3. Temperature variation of Vs

IV.CONCLUSIONS

The Young's and shear modulus were increasing with an increase of Sn addition to Ni-Zn ferrites. The longitudinal (V_l) and shear velocity (V_s) were found to be decreasing continuously with an increase of temperature (T) for the present samples. The longitudinal waves for the Sn added Ni-Zn ferrites attains maximum and minimum values around 370°C for T_1 and 380°C for T_2. The magneto crystalline anisotropy(k_1) also becomes zero at 370°C for present ferrites.

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