STUDIES ON THE DIELECTRIC CONSTANT, AC ELECTRICAL CONDUCTIVITY AND AC ELECTRICAL MODULUS OF V_2O_5 ADDITION LITHIUM-ZINC FERRITES

SM. RUBAYATUL ISLAM, M. RASEL SHIKDER AND M. SAMIR ULLAH*

Department of Physics, Bangladesh University of Engineering and Technology, Dhaka-1000

*Corresponding author e-mail: samirullah@phy.buet.ac.bd

Received on 09.01.2023, Revised received on 05.04.2023, Accepted for publication on 07.04.2023

DOI: https://doi.org/10.3329/bjphy.v30i1.68379

ABSTRACT

This study investigates the electrical transport features of V_2O_5 added Li-Zn ferrites. The ferrites have the compositions $Li_{0.25}Zn_{0.50}Fe_{2.25}O_4 + xV_2O_5$ (where $x=0.4,\ 0.8$ and 1.2 wt %) and are prepared by the solid-state reaction technique. Frequency-dependent dielectric constant and ac conductivity of the samples have been investigated at room temperature. An enhancement of dielectric constant and ac conductivity is observed with V_2O_5 addition and displayed a decreasing trend for higher V_2O_5 addition. The Nyquist impedance plots of the samples display a single semicircular arc, indicating that the conduction mechanism of Li-Zn ferrites occurs through the grain property.

Keywords: V_2O_5 addition, Dielectric constant, Nyquist impedance plots and Electric modulus

1. INTRODUCTION

The spinel-type of Li-Zn ferrites is a magnetic material that has been used in microwave devices in electronics industry for high frequency applications due to their substantial magnetic induction, high mechanical strength and low dielectric loss [1-4]. There are various sintering aids such as V_2O_5 , Bi_2O_3 , MnO_2 , MoO_3 and Nb_2O_5 have been used to enhance the grain formation and compaction of the samples that can tailor the electromagnetic properties of the materials [5-10]. However, spinel ferrites require high sintering temperatures to achieve densification, which can lead to the abnormal grain growth and deterioration of their electromagnetic properties [11]. V_2O_5 has been found to play a critical role as a sintering aid in improving mass transport mechanisms and densification of the samples [12-13]. However, there is insufficient data on the electrical transport features of V_2O_5 -added Li-Zn ferrites. Therefore, this study aims to investigate the dielectric constant, ac electrical conductivity, ac electrical modulus, and impedance spectroscopy of Lithium-Zinc ferrites with the addition of V_2O_5 .

2. MATERIALS AND METHOD

Li₂CO₃, ZnO, and Fe₂O₃ of high purity powders were used as raw materials to prepare the desired samples. These powders were weighed with appropriate ratio and mixed thoroughly by milling in a mortar-pestle for 5 h. Then the mixed powder was pre-heated at 750 °C for 3 h. Calcined powders were then combined with varying proportions of V₂O₅ (0.4, 0.8 and 1.2 wt%). Using a hydraulic press the resulting mixture was pressed into disc-shaped samples, which were sintered in air at 1050 °C for 3 h. An impedance analyzer (WAYNE KERR 6500B) was used to investigate the dielectric constant, ac conductivity, and impedance spectroscopy of the disc-shaped sample as a function of

frequency. The surface morphology of the samples has been inspected with the field emission scanning electron microscopy (FESEM: JSM-7600F, JEOL).

3. RESULTS AND DISCUSSION

3.1 Dielectric constant and tangent loss factor

The frequency dependent dielectric constant (ε') at ambient temperature is presented in **Fig. 1**. It demonstrates that ε' decreases as the frequency increases, which follows Maxwell-Wagner polarization [14].

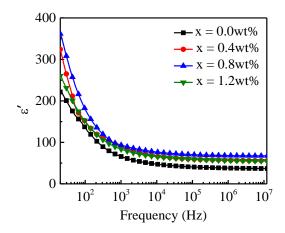


Fig.1 Real part of permittivity ε' against the applied frequency

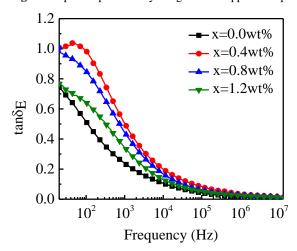


Fig. 2 Loss tangent for different V₂O₅ content against frequency

It is commonly acknowledged that the polarization of a specimen is influenced by various factors including the density of sintering, microscopic structure, and pore space of the ferrites [15]. **Figure 2**

shows the variation of the dielectric loss factor or tangent loss factor $(tan\delta_E)$ with applied frequency. The condition for highest loss factor $(tan\delta_E)$ is $\omega\tau=1$, where $\omega=2\pi f_o$, τ is the time takes for a system to return to its equilibrium state after being perturbed, called relaxation time, and f_o corresponds to a specific frequency where resonance take place [16]. Hence, $tan\delta_E$ step up to a peak value as the hopping frequency of the electron is close to the employed frequency. At higher frequencies, inclusion of V_2O_5 to Li-Zn ferrites results in lower dielectric losses, which could make them suitable for use in high-frequency devices.

The bulk density (ρ_B) and porosity (P%) of the investigated specimen are charted in **Table 1.** The data revealed that ρ_B increases with raising the value of V_2O_5 concentration up to 0.8 wt%, meanwhile the porosity decreases. This can be explained by the fact that V_2O_5 acts as a sintering aid, which improves the densification of the samples. However, P% increased at x=1.2 wt%, which might be associated with the subsequent decrease in sample density. The inclusion of V_2O_5 (0.4, 0.8, and 1.2 wt%) in Li-Zn ferrites results in a higher dielectric constant compared to pure Li-Zn ferrites. This is due to the enhanced polarization resulting from the increased densification of V_2O_5 included Li-Zn ferrites, particularly at lower frequency regions. Furthermore, frequency derivative of dielectric constant of V_2O_5 included Li-Zn ferrites decreases slowly at higher frequencies, indicating that they exhibit higher frequency stability compared to pure Li-Zn ferrites.

3.2 Electrical conductivity

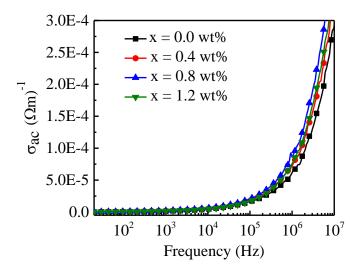


Fig.3 Electrical conductivity (σ_{ac}) against frequency

The change in frequency dependent ac electrical conductivity (σ_{ac}) at room temperature is plotted in **Fig. 3.** The conductivity of ferrites is given by, $\sigma_{ferrite} = \sigma_{dc} + \sigma_{ac}(\omega)$, where σ_{dc} represents the frequency independent term, called the dc conductivity and σ_{ac} denotes ac conductivity, a function of frequency [17]. Electron hopping occurs at the octahedral B-site and the concentration of Fe³⁺ ions is the primary factor accountable for the movement of electrons within localized sites.

Composition	$\rho_B (g/cm^3)$	P%	D (µm)
$Li_{0.25}Zn_{0.50}Fe_{2.25}O_4$	4.30	15	5.3
$Li_{0.25}Zn_{0.50}Fe_{2.25}O_4 + 0.4 \text{ wt}\%V_2O_5$	4.37	13	5.9
$Li_{0.25}Zn_{0.50}Fe_{2.25}O_4 + 0.8 \text{ wt } \%V_2O_5$	4.51	10	7.7
$Li_{0.25}Zn_{0.50}Fe_{2.25}O_4 + 1.2 \text{ wt } \%V_2O_5$	4.42	12	6.6

Table 1: Bulk density (ρ_B) , porosity (P%) and average grain size(D) of the compositions

The conductivity of the material remains relatively constant at lower frequencies due to the high resistivity of the grain boundaries, which are more operative comparative to the exceedingly conductive grains. As a result, the frequency at which electrons jump between Fe^{2+} and Fe^{3+} ions is low and insignificant in the lower frequency range. However, the conductivity increases rapidly at higher frequencies due to the activity of the conductive grains. This is because the highly conductive grains are more active than the poorly conductive grain boundaries. Therefore, the improvement in conductivity is attributed to an increase in electron hopping between Fe^{2+} and Fe^{3+} ions. The rising of ac electrical conductivity of V_2O_5 (0.4, 0.8 and 1.2 wt %) included Li-Zn ferrites is observed which could be attributed to the compaction and larger grain size (**Table 1**).

3.3 Electric modulus

Variation of the real part of electric modulus (M') as a function of frequency is depicted in Fig.4.

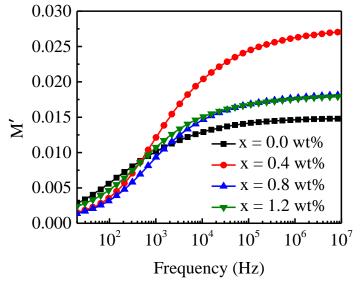


Fig. 4 Variation of M' with the applied frequency

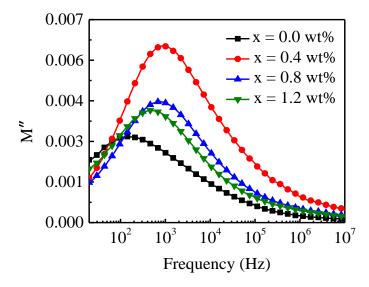


Fig. 5 Imaginary part of electric modulus (M'') against frequency

M' is very much low at lower frequency region, indicates that the electrode and/or ionic polarization is present in all samples. Dispersion is observed in the high frequency range due to the mobility of the charge carriers in limited distance. This is possibly caused by the deficiency in restoring force that can control the mobility of the charge carriers when an electric field is applied [18]. Frequency dependence of the imaginary part of electric modulus (M'') of studied samples are illustrated in **Fig.** 5. M'' rises up to a relaxation peak value at low frequency region as frequency increases. Further increase in frequency lower the value of M'' and approaches to a constant value due to the potential wells of the carriers at high frequency. The irregularity in the peaks suggests the occurrence of distinct relaxation times, indicating a non-Debye type of relaxation.

3.4 Nyquist impedance plots

To explore the impact of grain and grain boundary on the electrical characteristics of V_2O_5 added Li-Zn ferrites, Nyquist impedance plots/Cole-Cole plots ($Z''vs\ Z'$) have been scrutinized for studied samples. A Cole-Cole plot can be used to determine the individual contributions of the grain and grain boundary resistances to the overall resistance of a specimen [19-21].

Figure 6 shows the Cole-Cole plot for investigated samples. It can be seen that the semicircle areas are smaller for V_2O_5 -incorporated Li-Zn ferrites than that of Li-Zn ferrites, indicating a decrease in resistance and an increase in conductivity. This suggests that the semicircular arc corresponds to transportation caused by grain in the superior-frequency region, which is likely due to the larger grain sizes in V_2O_5 -enhanced Li-Zn ferrites [22]. Therefore, the conduction mechanism of V_2O_5 added Li-Zn ferrites takes place through the grain property.

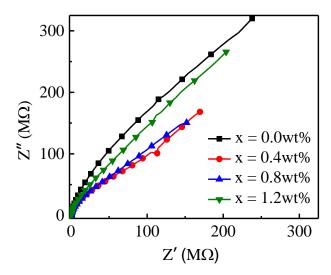


Fig. 6 Nyquist impedance plots/Cole-Cole plot

4. CONCLUSIONS

The inclusion of V_2O_5 in Li-Zn ferrites resulted in an increase in their dielectric constants as compared to the pure system. This can be attributed to the effect of interfacial polarization and densification of the samples. The lower values of dielectric losses found at higher frequencies might have possible technical application in high frequency devices. The Cole-Cole plots have revealed that the semicircle areas of V_2O_5 added Li-Zn ferrites are smaller than the pure Li-Zn ferrites. This may be due to the lowering in resistance, which leads to greater conductivity of V_2O_5 added Li-Zn ferrites.

ACKNOWLEDGEMENTS

The authors are thankful to the Materials Science Division, Atomic Energy Center Dhaka-1000, and Department of Nanomaterials and Ceramic Engineering (NCE), Bangladesh University of Engineering and Technology, Dhaka-1000 for their technical support to carry out this research work.

References

- Y. Gao, Z. Wang, R. Shi, J. Pei, H. Zhang, X. Zhou, "Electromagnetic and microwave absorption properties of Ti doped Li–Zn ferrites" J. Alloys Compd. 805, 934-941(2019)
- [2] Y. Gao, Z. Wang, J. Pei, H. Zhang, "Structural, elastic, thermal and soft magnetic properties of Ni-Zn-Li ferrites" J. Alloys Compd. **774**, 1233-1242 (2019)
- [3] A.V. Anupama, V. Rathod, V. M. Jali, B. Sahoo, "Composition dependent elastic and thermal properties of Li-Zn ferrites" J. Alloys Compd. 728, 1091-1100 (2017)
- [4] V. Rathod, A. V. Anupama, V. M. Jali, V. A. Hiremath, B. Sahoo, "Combustion synthesis, structure and magnetic properties of Li-Zn ferrite ceramic powders" Ceram. Int. 43, 14431-14440 (2017)
- [5] M. Samir Ullah, M. Firoz Uddin, A. A. Momin, M. A. Hakim, "Effect of V₂O₅ addition on the structural and magnetic properties of Ni–Co–Zn ferrites" Mater. Res. Express, 8 (1), 016102 (2021)

- [6] H. Bahiraei, C. K. Ong, "Influence of MoO₃ additive on grain growth and magnetic properties of Mg_{0.3}Cu_{0.2}Zn_{0.5}Fe₂O₄ ceramics sintered at low temperature J. Mater. Sci.: Mater. Electron. 32(22), 26967-26974 (2021)
- [7] Y. Yang, J. Li, H. Zhang, G. Wang, G. Gan, Y. Rao, "Effects of Bi₂O₃-MnO₂ additives on tunable microstructure and magnetic properties of low temperature co-fired NiCuZn ferrite ceramics" J. Mater. Sci.: Mater. Electron. **31**(**15**), 12325-12332 (2020)
- [8] F. Xie, L. Jia, F. Xu, G. Gan, J. Li, Y. Li, Y. Li, H. Zhang, "Low-temperature sintering Li-Zn-Ti-Mn ferrite ceramics: synthesis, microstructure, and enhanced ferromagnetic properties with CuO-V₂O₅ additive" J. Mater. Sci.: Mater. Electron. 29(15), 13337-13344 (2018)
- Y. Jin, H. Zhu, Y. Xu, H. Zhu, H. Zhou, Y. Jin, "Effects of Nb₂O₅–WO₃ additive on microstructure and magnetic properties of low temperature-fired NiCuZn ferrites. J. Mater. Sci.: Mater. Electron. 26, 2397– 2402 (2015)
- [10] B. P. Rao, O. Caltun, I. Dumitru, L. Spinu, "Complex permeability spectra of Ni-Zn ferrites doped with V₂O₅/Nb₂O₅" J. Magn. Magn. Mater. 304, 749–751 (2006)
- [11] K. Raju, G. Venkataiah, D. H. Yoon, "Effect of Zn substitution on the structural and magnetic properties of Ni-Co ferrites" Ceram. Int. **40(7)**, 9337-9344 (2014)
- [12] J. Du, G. Yao, Y. Liu, J. Mia, G. Zu, "Influence of V₂O₅ as an effective dopant on the sintering behavior and magnetic properties of NiFe₂O₄ ferrite ceramics" Ceram. Int. 38, 1707-1711 (2012)
- [13] R. Lebourgeois, S. Duguey, J. P. Ganne, J. M. Heintz, "Influence of V₂O₅ on the magnetic properties of nickel-zinc-copper" J. Magn. Magn. Mater. 312, 328–330 (2007)
- [14] J.C. Maxwell, "A Treatise on Electricity and Magnetics" Vol. 1, 374-387, London (1873)
- [15] J. S. Ghodake, R. C. Kambale, S. V. Salvi, S. R. Sawant, S. S. Suryavanshi, "Electric propeties of Co substituted Ni-Zn ferrites" J. Alloys Compd. 486, 830–834 (2009)
- [16] Navneet Sing, Ashish Agarwal, Sujata Sanghi, "Dielectric relaxation, conductivity behavior and magnetic properties of Mg substituted Zn-Li ferrites", Curr. Appl. Phys. 11, 783-789 (2011)
- [17] Alina Manzoor, Muhammad Azhar Khan, Muhammad Yaqoob Khan, Majid Niaz Akhtar, Altaf Hussain, "Tuning magnetic and high frequency dielectric behavior in Li-Zn ferrites by Ho doping", Ceram. Int. **44(6)**, 6321-6329 (2018)
- [18] A. A. Momin, R. Parvin, M. F. Islam, A. K. M. Akther Hossain, "Structural, magnetic and electrical properties of Multiferroics xLi_{0.1}Ni_{0.2}Mn_{0.6}Fe_{2.1}O₄–(1-x)Bi_{0.8}Y_{0.2}FeO₃ composites" J. Magn. Magn. Mater. 526, 167708 (2021)
- [19] M. Firoz Uddin, M. Samir Ullah, S. Manjura Hoque, F. A. Khan, A. A. Momin, Sm. Rubayatul Islam, Faizus Salehin and M. A. Hakim, "Electrical transport properties of V₂O₅ added Ni-Co-Zn ferrites" J. Adv. Dielect. 11, 2150025 (2021)
- [20] M. Belal Hossen, and A.K.M Akther Hossain, "Complex impedance and electric modulus studies of magnetic ceramic Ni_{0.27}Cu_{0.10}Zn_{0.63}Fe₂O₄." J. Adv. Ceram, 4(3), 217-225 (2015)
- [21] S. Nasrin, M. Sharmin, M. A. Matin, A. K. M. Akther Hossain, M. D. Rahman, "Study the impact of sintering temperature on electromagnetic properties of (1-y)[Ba_{0.9}Ca_{0.1}Zr_{0.1}Ti_{0.9}O₃] + y[Ni_{0.25}Cu_{0.13}Zn_{0.62}Fe₂O₄] composites", Appl. Phys. A, 127 59 (2021)
- [22] M. Kaiser, "Influence of V₂O₅ ion addition on the conductivity and grain growth of Ni-Zn-Cu ferrites" Curr. Appl. Phys., 10, 975-984 (2010)