Impedance Spectroscopy of Nanocrystalline MgFe$_2$O$_4$ and MnFe$_2$O$_4$ Ferrite Ceramics: Effect of Grain Boundaries on the Electrical Properties

Dalibor L. Sekulić$^{1*}$, Zorica Z. Lazarević$^2$, Čedomir D. Jovalekić$^3$, Aleksandra N. Milutinović$^2$, Nebojša Z. Romčević$^2$

$^1$Faculty of Technical Sciences, University of Novi Sad, Trg Dositeja Obradovića 6, Novi Sad, Serbia

$^2$Institute of Physics, University of Belgrade, P.O. Box 68, Pregrevica 118, Zemun, Belgrade, Serbia

$^3$The Institute for Multidisciplinary Research, University of Belgrade, Kneza Višeslava 1, Belgrade, Serbia

Abstract:
Two ferrite ceramic materials, MgFe$_2$O$_4$ and MnFe$_2$O$_4$, were successfully fabricated by a conventional sintering of nanosized powders (at 1373 K for 2 h) synthesized by soft mechanochemical route. The particle size and morphology of powders were studied using X-ray diffraction (XRD) and transmission electron microscopy (TEM). XRD analysis was carried out for the determination of phase purity, crystal structure and average crystallite size of sintered ferrites. Both mechanosynthesized ferrite samples show mean crystallite sizes in the nm–range. Over the frequency range of 100 Hz to 1 MHz, impedance spectra of prepared ferrite ceramics are investigated at and above room temperature. Changes in the impedance plane plots with temperature have been discussed and correlated to the microstructure of materials. An equivalent circuit model is applied to explore the electrical parameters (resistance and capacitance) associated with grains and grain boundaries. Complex impedance analysis indicates the dominance of grain boundary effects which control the overall electrical behaviour of studied ferrites. The decrease in grain boundary resistance with temperature suggests a thermally activated conduction mechanism.

Keywords: Ferrite ceramics; Impedance spectroscopy; Electrical properties; Grain boundaries.

1. Introduction

Spinel ferrites MFe$_2$O$_4$, where M$^{2+}$ is a divalent metal cation, are an important class of magnetic materials, which have been intensively investigated over the last few decades due to good combination of electrical and magnetic properties. These materials are technologically interesting because of high electrical resistivities and consequently low eddy current losses, as well as high dielectric constant, high initial permeability and moderate saturation magnetization [1]. Polycrystalline ferrites are widely used in electronic applications in a range of frequencies extending from microwave to radio frequency.

Most of the ferrospinel form cubic spinel–type structure with the distribution of
cations between two different coordinated interstitial lattice sites, tetrahedral (A) and octahedral [B] sublattices. The divalent metal cation $M^{2+}$ can occupy the either (A) or [B] sites or both sites of the spinel structures. Generally, the structure of spinel ferrites could be described with the $(M_1\lambda Fe_\lambda)\{M_\lambda Fe_{2-\lambda}\}O_4$ formula, where $\lambda$ is the so-called inversion degree defined as the fraction of tetrahedral (A) sites occupied by trivalent $Fe^{3+}$ cations and its value depends on the method of preparation [2]. Typically two extreme type of spinel structure can be found, normal and inverse spinel, but in practice a mixed spinel structure are often observed, especially in the case of nanocrystalline ferrites. If metal cation $M^{2+}$ occupies only the (A) sites, the spinel is normal ($\lambda$=0); but in the case when it occupies only the [B] sites, the spinel is inverse ($\lambda$=1). The mixed spinel structure has the cations distributed in a combined mode, $M^{2+}$ occupying both (A) and [B] sites. In addition, the magnetic, dielectric and electrical properties of spinel ferrites can be varied systematically by changing the identity of the divalent $M^{2+}$ cations (M= Ni, Zn, Mn, Mg, Co etc.) without changing the spinel crystal structure [3].

Among spinel ferrites, the magnesium (MgFe$_2$O$_4$) and manganese (MnFe$_2$O$_4$) ferrites are soft magnetic semiconducting materials characterized by low dielectric losses and high resistivity due to their dielectric nature. Previous experimental studies have shown that MnFe$_2$O$_4$ belongs to the partially inverse spinel, where in the bulk form 20% of the Mn$^{2+}$ cations residing on the octahedral sublattice, while a higher inversion up to 60% were reported in nanostructured manganese ferrite [4]. Also, it has been reported that the spinel structure of MgFe$_2$O$_4$ bulk material is nearly inverse with 90% of Mg$^{2+}$ cations distributed over [B] sites, and that inversion factor decreases to about 70% for the magnesium ferrite nanoparticles [5]. Electrical as well as magnetic properties of spinel ferrites strongly depend on the cation distribution at the different sites as well as method of preparation, sintering temperature, particle size, doping of additives etc. [6]. Currently, the synthesis of reproducible ferrites is being carried out by various techniques in order to improve their physical properties and widen the scope of their applications. Several methods such as co–precipitation [4, 6, 7], sol–gel [8, 9], combustion [5, 8], ball milling [10, 11] etc. have been used for successful preparation of magnesium and manganese ferrites in recent years.

In the present paper, we have reported the electrical properties of MgFe$_2$O$_4$ and MnFe$_2$O$_4$ ferrites prepared by a conventional sintering of nanosized powders synthesized by soft mechanochemical processing that is environmentally friendly, does not require expensive starting materials or extremely high temperature [12]. In order to understand the conduction mechanism, electrical properties associated with grains and grain boundaries in these synthetized nanocrystalline ceramic materials were studied as a function of frequency and temperature using complex impedance spectroscopy technique.

2. Complex impedance formalism

Complex impedance spectroscopy [13] is a non–destructive and widely used testing method for analysing the electrical processes occurring in materials on the application of small AC signal as input perturbation. To understand the overall properties of electroceramics, electrical information associated with its microstructures is very important. Impedance spectroscopy has the capability to resolve the effect of grains and grain boundaries, two main components which comprise the microstructure of ceramics. In this technique, the real and imaginary parts of complex electrical parameters (impedance, modulus or dielectric permittivity) are measured as a function of frequency and temperature. Generally, the complex impedance can be expressed by using the classical Debye expression [14]

$$Z^*(\omega) = \frac{Z_0}{1 + j\omega\tau} = Z'(\omega) - jZ''(\omega),$$

(1)
where \( \omega \) and \( \tau \) are the angular frequency and the relaxation time, respectively. Here, \( Z' \) and \( Z'' \) represent the real component of impedance and imaginary component of impedance, respectively.

The charge transport behavior of the electroceramics and its relation to the different microstructures (phases) are studied by means of the impedance spectra. The impedance spectrum, known as Cole–Cole plot, is usually represented as \( Z'' \) against \( Z' \). The Cole–Cole plot of polycrystalline material commonly gives two semicircles depending upon the electrical properties of investigated material. The first semicircle at low frequency represents the resistance of grain boundary, while the second one obtained for high frequency domain corresponds to the resistance of grain or bulk properties [15]. It is well–known that the behaviour of characteristic impedance spectra is usually interpreted in terms of equivalent circuit model. From the microstructural point of view, each of ceramic components (i.e. grains and grain boundaries) may be represented by a R–C element (resistor R and capacitor C in parallel combinations), and the simplest appropriate equivalent circuit is a series array of parallel R–C elements [16], see Fig 1a. In accordance with this approach known as brick layer model, total complex impedance is given by

\[
Z''(\omega) = \left( \frac{1}{R_{gb} + j\omega C_{gb}} \right)^{-1} + \left( \frac{1}{R_g + j\omega C_g} \right)^{-1},
\]

where \( R_g \) and \( R_{gb} \) represent the grain and grain boundary resistance, while \( C_g \) and \( C_{gb} \) are the grain and the grain boundary capacitance, respectively.

Fig. 1. Equivalent electrical circuits of two–phase electroceramic materials according to brick layer model: (a) two parallel R–C elements; (b) two parallel R–CPE elements.

In many electroceramics, the resistance of grain boundaries exceeds that of the grains [17]. This can be ascribed to the fact that the atomic arrangement near the grain boundary region is disordered, resulting in a serious increase in electron scattering. The higher boundary resistance also arises from other factors such as a decrease in \( Fe^{2+} \) ion content in this region [18] Also, the capacitance of grain boundary is usually larger than that of the grain, which is explained on basis that the capacitances are inversely proportional to the thickness of the media. In such system, these two components of ceramic microstructure indicate two distinct relaxation phenomena with sufficiently different relaxation times: \( \tau_g = R_g C_g \) and \( \tau_{gb} = R_{gb} C_{gb} \) [19]. Additionally, the relaxation time of the grain boundaries \( \tau_{gb} \) is much larger than that of the grains \( \tau_g \), since the semicircle representing the grain boundaries lies on the lower frequency side. In general, all these quantities can be treated as fitting parameters whose values can be evaluated from the best fit to the impedance spectrum. The fitting is relatively
unambiguous since the elements of equivalent circuits dominate in different frequency regions.

It is important to note that a proposed circuit, two parallel RC elements connected in series, refers to the ideal Debye–like behaviour in ceramic materials. But in practice, the most electroceramics exhibit non–ideal Debye–like behaviour, so that a constant phase element (CPE) is used in the equivalent circuit to illustrate more fully the deviation from an ideal capacitor [15], as shown Fig 1b. The CPE has impedance defined as [20]

\[ Z_{\text{CPE}}(\omega) = \left[ A(j\omega)^n \right]^{-1}, \quad (3) \]

where parameter A is independent of frequency, and n is the measure of non–ideal behavior having value between 1 and 0 (0 ≤ n ≤ 1). The CPE describes an ideal capacitor with C = A for n = 1 and an ideal resistor with R = 1/A for n = 0. Based on that, total complex impedance reads as follows [16]

\[ Z^*(\omega) = \left( \frac{1}{R_{gb}} + A_{gb}(j\omega)^{n_{gb}} \right)^{-1} + \left( \frac{1}{R_g} + A_g(j\omega)^{n_g} \right)^{-1}. \quad (4) \]

Using the above equation, the equivalent circuit elements can be fitted from the experimental data of impedance spectrum for various materials.

3. Experimental details

3.1. Preparation

The MgFe\textsubscript{2}O\textsubscript{4} and MnFe\textsubscript{2}O\textsubscript{4} ferrite nanoparticles were prepared by means of soft mechanochemical route using a mixture of:

- magnesium(II)–hydroxide (Mg(OH)\textsubscript{2}, Merck 95% purity) and hematite (α–Fe\textsubscript{2}O\textsubscript{3}, Merck 99% purity), and
- manganese(II)–hydroxide (Mn(OH)\textsubscript{2}, Merck 95% purity) and hematite (α–Fe\textsubscript{2}O\textsubscript{3}, Merck 99% purity)

in equimolar ratio, respectively. The starting hydroxides were amorphous, while hematite was crystalline. Mechanochemical synthesis was carried out with planetary ball mill (Fritsch Pulverisette 5) in air atmosphere. Experimentally determined milling times were 15 h and 25 h for MgFe\textsubscript{2}O\textsubscript{4} and MnFe\textsubscript{2}O\textsubscript{4}, respectively, depending upon the progress of formation of spinel ferrite phase. Thus obtained nanosize powders were pressed into disk–shaped pallets using a cold isostatic press. The pellets were then sintered at 1373 K for 2 h (Lenton–UK oven) without pre–calcinations step. Heating rate was 10°C/min, with nature cooling in air atmosphere. The thickness and diameter of sintered samples used for further characterizations are 2.0 mm and 10.0 mm, respectively.

3.2. Characterization

The particle size and morphology of soft mechanochemical synthetized powders were investigated using a 200 kV transmission electron microscope (TEM, Model JEOL JEM–2100 UHR). The phase purity and crystal structure of the powder and sintered samples have been characterized using X–ray diffractometer (Model Philips PW 1050) at room temperature. The XRD data was collected using CoKα radiation (λ = 1.78897 Å) at 40 kV and 20 mA (PW 1730 generator) in a wide range of Bragg’s angles 2θ (15° ≤ 2θ ≤ 80°) with a scanning step size of 0.05° in a 10 s per step of counting time. The lattice parameter and crystallite (grain) size of the sintered ferrites were calculated from XRD data.

In order to study the electrical properties of MgFe\textsubscript{2}O\textsubscript{4} and MnFe\textsubscript{2}O\textsubscript{4} ferrite ceramics by means of complex impedance spectroscopy, both the flat and parallel surfaces of the
samples were polished and contacts were made by high quality silver paste on opposite sides of the pallets. The electrical parameters of sintered ferrites, parallel resistance ($R_p$) and parallel capacitance ($C_p$), were measured in a wide frequency range (100 Hz – 1 MHz) at different temperatures (298 K – 398 K) using a computer-controlled Impedance Analyzer HP–4194A with a laboratory set of temperature control equipment. The experimentally measured parallel values of the resistance and the capacitance were converted and displayed in the form of complex impedance as follows:

$$Z^\ast(\omega) = Z'(\omega) - jZ^\prime\prime(\omega) = \left(\frac{1}{R_p} + j\omega C_p\right)^{-1}.$$

The details of this measurement procedure are same as presented in [15]. For fitting and analysis of the impedance data, EIS Spectrum Analyzer software [21] was used with about 2% of fitting errors.

4. Results and discussion
4.1. Structural analysis

XRD patterns of synthesized MgFe$_2$O$_4$ and MnFe$_2$O$_4$ ferrite nanopowders are shown in Fig. 2. It is evident that the cubic spinel structure was formed during the soft mechanochemical processing. The appearance of a peak at $2\theta = 35.50^\circ$, as well as of peaks at $2\theta = 18.24^\circ$, $30.14^\circ$, $43.13^\circ$, $53.63^\circ$, $57.00^\circ$ and $62.66^\circ$, clearly points to the formation of the new phase of MgFe$_2$O$_4$ (JCPDS card 88–1943) (Fig. 2 left). The peaks are well indexed to the crystal plane of spinel ferrite (k h l) (311) for the most intense peak at $2\theta = 35.50^\circ$, and (k h l) (111), (220), (400), (422), (511) and (440) for other peaks, respectively. The peaks at $2\theta = 18.28^\circ$, $29.96^\circ$, $35.25^\circ$, $42.92^\circ$, $53.22^\circ$, $56.46^\circ$, $62.13^\circ$ and $73.74^\circ$, clearly confirm the spinel phase MnFe$_2$O$_4$ (JCPDS card 74–2403) (Fig. 2 right). The peaks are well indexed to the crystal plane of spinel ferrite (k h l) (111), (220), (311), (400), (422), (333), (440) and (533), respectively. The diffraction peaks are fairly broad, which is caused by nanometer size of the crystallites. The average crystallite sizes were calculated from the most intense peak (311) using Scherer’s formula [20]. The average particle sizes have been found to be 12 nm and 16 nm for magnesium and manganese ferrites, respectively.

Fig. 2. The experimental XRD patterns of MgFe$_2$O$_4$ and MnFe$_2$O$_4$ ferrite nanopowders, recorded at room temperature. Insets show TEM images of ferrite nanoparticles.
The representative TEM images of powders obtained after appropriate milling time are depicted in the inset of Fig. 2. It can be seen that the particles have an almost spherical morphology with size distribution between 5 – 15 nm and 10 – 25 nm for MgFe$_2$O$_4$ and MnFe$_2$O$_4$, respectively, which is in close agreement with the crystallite size obtained from XRD data. TEM micrographs confirm nanoscale nature of as–prepared ferrite particles which tend to agglomerate because of the dipolar field of each crystallite.

Fig. 3. shows the room temperature X–ray diffraction spectra of MgFe$_2$O$_4$ and MnFe$_2$O$_4$ ferrite ceramics sintered at 1373 K for 2 h. The relative intensities of all diffraction peaks match well with the reported cubic spinel ferrite and sharp XRD peaks indicate the polycrystalline nature of sintered samples.

The values of crystallite size and lattice parameters deduced from experimental X–ray data are summarized in Tab. I. The lattice constants were calculated using data of inter–planar spacing and corresponding Miller indices (hkl) [15]. Also, Tab. I shows the values of the sintered (bulk) density of the samples which were measured by applying Archimedes’ principle at room temperature.

Tab. I Some structural parameters of MgFe$_2$O$_4$ and MnFe$_2$O$_4$ ferrite samples sintered at 1373 K for 2h.

<table>
<thead>
<tr>
<th>ferrite sample</th>
<th>lattice constant (Å)</th>
<th>crystallite size (nm)</th>
<th>sintered density (g/cm$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MgFe$_2$O$_4$</td>
<td>8.371</td>
<td>41</td>
<td>4.53</td>
</tr>
<tr>
<td>MnFe$_2$O$_4$</td>
<td>8.421</td>
<td>49</td>
<td>4.21</td>
</tr>
</tbody>
</table>

4.2. Complex impedance analysis

The frequency dependence of imaginary part of impedance $Z''$, usually called as loss spectrum, at selected temperatures is shown in Fig. 4. In the case of both samples can be seen that the $– Z''$ value increases initially, reaches the maximum value ($Z''_{\text{max}}$) at particular frequency and then it decreases continuously with increasing frequency for given temperatures. The appearance of only one peak in loss spectra at characteristic frequency ($\omega_{\text{max}} = 2\pi f_{\text{max}}$) suggests an existence of single relaxation process in MgFe$_2$O$_4$ and MnFe$_2$O$_4$ ferrites under study. Observed relaxation peaks are due to the existence of the space charge relaxation that dominates when the material is composed of grain and grain boundaries [22]. In both samples, these peaks shift towards the higher frequency side possibly due to an
increase in the rate of hopping of charge carriers with the rise of temperature. The shifting of peaks in $Z''$ also indicates to the decreasing relaxation time of mobile charge carriers with the increase of temperature [16]. A lower relaxation time implies faster movement of mobile carriers and vice versa. Also, it is evident that the magnitude of $Z''$ maxima decreases with increase in temperature, indicating increasing loss in the resistive property of prepared samples [23].

![Image](image.png)

**Fig. 4.** Variation of imaginary parts of impedance with frequency for MgFe$_2$O$_4$ and MnFe$_2$O$_4$ ferrites at and above room temperature.

Generally, the impedance loss spectra were used to evaluate the relaxation time $\tau$ of the electrical phenomena occurring in ferrites using the following relation [24]

$$\tau = \omega^{-1}_{\text{max}} = \frac{1}{2\pi f_{\text{max}}},$$  

(6)

where $f_{\text{max}}$ is characteristic frequency maximum known as relaxation frequency which depends on temperature. Variation of corresponding values $\tau$ with inverse of absolute temperature is given in Fig 5. A decrease in relaxation time with temperature can be noted for both ferrites. Also, these graphics follow the Arrhenius dependence [4]

$$\tau = \tau_0 \exp\left(\frac{\Delta E}{k_B T}\right),$$  

(7)

where $\tau_0$ is the pre–exponential factor, $\Delta E$ represents the activation energy and $k_B$ is Boltzmann’s constant. From the Arrhenius plots, the activation energies are found to be 0.361 eV and 0.406 eV for MgFe$_2$O$_4$ and MnFe$_2$O$_4$ samples, respectively. Since $\tau = (2P)^{-1}$, where $P$ is the hopping probability, the decrease in relaxation time with temperature results in an increase in hopping probability of charge carriers with temperature [4]. The pre–exponential factor $\tau_0$ has been calculated from the results of fitting and its values are $6.35 \times 10^{-11}$ s and $1.49 \times 10^{-13}$ s for MgFe$_2$O$_4$ and MnFe$_2$O$_4$, respectively.

In order to correlate the electrical properties of synthetized ferrites with their microstructures, the impedance spectra were studied in the wide frequency range from 100 Hz to 1 MHz at several temperatures (298 K – 398 K). Fig. 6 shows the measured complex impedance plane plots for MgFe$_2$O$_4$ and MnFe$_2$O$_4$ at selected temperatures. It is evident that Cole–Cole plots clearly show evidence of the existence of only one semicircle, which indicates that the contribution to electrical conductivity arises mainly from the grain boundary and that contribution from the grain is not resolved for both samples within the measuring range up to 1MHz. In the cases of nanocrystalline ferrites, the similar behaviour of impedance spectra with single semicircle was observed by other authors [25, 26]. These results suggest
that the grain boundary volume in nanostructured ferrites is high because of the small grain sizes and hence conduction takes place predominantly through the grain boundary [27]. In addition, it is obvious that obtained impedance spectra of MgFe$_2$O$_4$ and MnFe$_2$O$_4$ are temperature dependent, since the diameters of semicircles exhibit decreasing trends with increasing temperature. This indicates decrease in relaxation time and increase in conductivity with rise in temperature.

![Fig. 5. Variation of relaxation times with inverse of temperature.](image1)

![Fig. 6. Impedance spectra of MgFe$_2$O$_4$ and MnFe$_2$O$_4$ ferrites at and above room temperature.](image2)

It is usual to interpret the impedance response in terms of equivalent circuit model. The shape of the impedance plane plots for ferrites under study suggests that the impedance data can be approximately modelled by an equivalent circuit based on only one parallel R–CPE element shown in the inset of the Fig. 6. Here, R$_{gb}$ and CPE$_{gb}$ are the resistances and the constant phase elements for grain boundaries, respectively. The parameters R$_{gb}$, C$_{gb}$ and n$_{gb}$ have been derived by fitting the impedance spectra (within 0.5 – 2% fitting errors).

The variation of the fitted parameters is depicted as a function of temperature in Fig. 7. Evaluated R$_{gb}$ for MgFe$_2$O$_4$ and MnFe$_2$O$_4$ show a strong similarity in their temperature dependence and their values decrease as the temperature increased. This kind of behaviour
suggests that the both ferrites possess a negative temperature coefficient of resistance (NTCR) [16], implying a thermal activated conductivity of these materials. Also, this substantial drop in the $R_{gb}$ provides a clear and unambiguous evidence of grain boundary conduction resulting from reduction in the barrier facilitating thereby mobility of charge carriers with rise in temperature [24]. Further, Fig. 7 right implies the slight variations in parameters $C_{gb}$ and $n_{gb}$ with temperature. The $n_{gb}$ has a slow decreasing trend in the measured temperature range and its values are between 0.835 and 0.817 for MgFe$_2$O$_4$, i.e. between 0.775 and 0.735 for MnFe$_2$O$_4$. This trend signifies that the grain boundary capacitance deviates from ideal behaviour as the temperature is increased. The observed decrease in the capacitance of the grain boundaries with increasing temperature may be due to the delocalization of the charge carriers from traps followed by the accumulation of these charges at the grain boundaries, thereby increasing its capacitance as well as space charge polarization effects [28]. Similar results for this behaviour of the grain boundaries capacitance were found by others authors [2, 28].

Fig. 7. Variation of grain boundary resistance $R_{gb}$ with temperature (left), and variation of grain boundary capacitance $C_{gb}$ and deviation parameter $n_{gb}$ with temperature (right) for nanocrystalline MgFe$_2$O$_4$ and MnFe$_2$O$_4$ ferrites.

In the present study, the DC conductivity for grain boundary contribution is calculated using the expression [24]

$$\sigma_{DC} = \frac{d}{R_{gb} A}, \quad (8)$$

where $R_{gb}$ is the resistance values of the grain boundary which is evaluated from impedance spectra of nanocrystalline ferrites under study. Parameters $d$ and $A$ represent the thickness of sample and area of the electrode deposited on the sample, respectively. Fig. 8 shows the temperature dependence of $\sigma_{DC}$. It can be clearly noticed that the nature of the variation is linear and well obeys the Arrhenius relation that is defined by equation [20]:

$$\sigma_{DC} = \sigma_0 \exp(-\frac{E_a}{k_B T}), \quad (9)$$

where $\sigma_0$, $E_a$ and $k_B$ are the pre–exponential factor, the activation energy of the carriers for conduction and Boltzmann’s constant, respectively. In the cases of both ferrites, the DC conductivity increases with rise in temperature showing a typical semiconductor behaviour with negative temperature coefficient of resistance (NTCR) characteristics [29]. Using this Arrhenius plot of grain boundary conductivity, the corresponding activation energies for the thermally activated charge carriers in MgFe$_2$O$_4$ and MnFe$_2$O$_4$ were determined from the slope.
of the log\(\sigma_{DC}\) versus 1000/T curves and were found to be 379 meV and 392 meV, respectively.

![Graph showing variation of DC conductivity of grain boundary with inverse of temperature.](image)

**Fig. 8.** Variation of DC conductivity of grain boundary with inverse of temperature.

Comparison of evaluated DC conductivity for grain boundary contribution and activation energy shows a good agreement in the sense that the sample with lower conductivity has higher value of activation energy and vice versa. The grain boundary conductivity in sintered nanocrystalline MgFe\(_2\)O\(_4\) and MnFe\(_2\)O\(_4\) was found to be \(7.83 \times 10^{-6}\) \(\Omega^{-1} m^{-1}\) and \(2.82 \times 10^{-6}\) \(\Omega^{-1} m^{-1}\) at room temperature, respectively.

### 5. Conclusions

In summary, nanocrystalline MgFe\(_2\)O\(_4\) and MnFe\(_2\)O\(_4\) spinel ferrites have been synthesis using a soft mechanochemical route and then the powders were sintered at 1373 K for 2 h. The X–ray diffraction analysis confirmed the cubic spinel structure of sintered samples and the average crystallite sizes in the nm–range were determined on the basis of XRD data. Complex impedance spectroscopy was used to characterize the electrical properties of these materials. In the studied frequency range, the impedance spectra of sintered ferrites show the presence of only one semicircle at and above room temperatures. The analysis of impedance data using an equivalent circuit indicates that the electrical properties of the synthetized ferrites are mainly attributed to the processes that are associated with the grain boundaries. The decrease in value of grain boundaries’ resistance on increasing temperature suggests the negative temperature coefficient of resistance behavior usually shown by semiconducting materials. In the case of both ferrites, the imaginary part of impedance clearly confirms only one relaxation processes due to grain boundaries. It was found that the relaxation time decreases with temperature and follows Arrhenius dependence.

### Acknowledgments

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6. References

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Садржај: Два феритна керамичка материјала, MgFe₂O₄ и MnFe₂O₄, успешно су фабрикована помоћу конвенционалног синтетовања нанопрахова (на 1373 K за 2 сата) синтетисаних soft механохемијским путем. Величина честице и морфологија прахова је испитана коришћењем рендгенске дифракције (XRD) и трансмисионог електронског микроскопа (TEM). XRD анализа је употребљена за одређивање чистоће фазе, кристалне структуре и просечне величине кристала синтетованих ферита. Оба механо–хемијском реакцијом синтетисана феритна узорка поседују средњу величину кристала у тт–опсезу. У широком опсезу фреквенција од 100 Hz до 1 M Hz, импедансни спектри феритних керамика су испитани на и изнад собне температуре. Промене на импедансним дијаграмима са порастом температуре су дискутоване и доведене у корелацију са микроструктуром материјала. Примењен је модел еквивалентног електричног кола ради одређивања електричних параметара (отпорност и капацитивност) везаних за зрна и границе зрна. Анализа комплексне импедансе указала је на доминацију ефекта граница зрна који контролише електричне особине испитиваних ферита. Смањење отпорности границе зрна са температуром указује на механизам провођења активиран температуром.

Кључне речи: феритна керамика; импедансна спектроскопија; електрична својства; границе зрна