Correlation of structural and electrical properties in heterovalent substituted Sr-hexaferrites

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Abstract
M-type hexagonal ferrites with heterovalent substitutions were synthesized using co-precipitation method. The nominal compositions used were SrFe_{12-2x}Cr_{x}Zn_{x}O_{19} (x = 0.0–1.0). X-ray diffractograms of the samples confirmed the hexagonal structure of strontium hexaferrite. The crystallite size was found to be in the range of 42–67 nm. The lattice expands in a non-monotonous way with the extent of dopant concentration. Scanning electron microscopy showed particles like morphology with the estimated particle sizes in the range of 8–11 μm. By varying the frequency (from 1 kHz to 3 MHz) and temperature (from RT to 300 °C), the dielectric properties were investigated. Varying both frequency and temperature, the dielectric constant and loss tangent were found to be decreased with increase in frequency. Also, dielectric constant and loss tangent were found to be increased with increase in temperature. At 1 kHz frequency and temperature 300 °C composition (x = 1.0) showed maximum value of dielectric constant. Such a composition can be used for capacitors where high energy storage is required. AC conductivity of all the prepared samples was also found to be increased with increasing the temperature and applied frequency. This phenomenon is exactly in accordance with Jonscher’s power law. The value of exponent ‘n’ used in Jonscher’s power law has been calculated and is found to be decreasing with temperature. Samples followed Correlated Barrier Hoping (CBH) model. The real and imaginary parts of complex impedance were also measured, and these are in accordance with the ac conductivity graphs. Cole-Cole plots were drawn. The plots helped to suggest an equivalent circuit to understand the grain and grain boundary effect in the prepared samples. It was observed that for all the prepared samples grain boundary effect is dominant at 00 °C. Nonsymmetrical and depressed semicircle arcs were also produced that indicate the non-Debye type relaxation processes in the studied compositions.

Introduction
Magnetoplumbite (M-type) hexagonal ferrites with general chemical formula MnFe_{12}O_{19} (M = Pb, Ba, Sr) are magnetic materials and among M-type ferrites, strontium hexaferrites are significantly important due to their high chemical stability [1], good thermal durability and excellent magnetic properties [2, 3]. They are used in energy storage devices [4], generators, medical equipment [5], wave absorbers [6] and many other electronic devices. Strontium hexaferrites crystal structure consists of different eleven symmetry sites. Each site has 64 ions per unit cell. The Fe ions are occupied by three octahedral sites (12 k, 2a, 4f_{2}), one tetrahedral site (4f_{1}) and one bi-pyramidal site (2b) [7]. It has been reported that magnetic and electrical properties of strontium hexaferrites can be enhanced by improving its microstructure features, type of substitution of divalent-tetravalent ions (Ni–Ti, Co–Ti, Bi–Co etc) trivalent metal ions (Al, Cr, Bi and La etc) and rare earth metal ions such as La, Nd, Gd, Tb, Dy etc [8]. It is also reported that the transport properties of hexaferrites can be improved by changing the sintering environment [9], method of preparation and with doping concentration [10]. The dielectric behavior provides valuable information about the localized charge carriers but it also varies with frequency and temperature of operation. Its behavior is much helpful in studying the mechanisms responsible for charge
transport phenomena and dielectric behavior. Moreover, a number of methods are used to synthesis hexaferrites however, co-precipitation method is considered to be a favorable wet chemical method to get phase pure and controlled grain size [11, 12]. Magnetic and electrical properties of different hexaferrites have been extensively studied by many researchers worldwide however, investigations of the substitution effect on the electrical and dielectric properties of strontium hexaferite with respect to temperature are very rare. It is reported by one group that dielectric constants and dielectric losses were decreased with increase in Cr–Zn concentration while investigating structural, dielectric and magnetic properties of Cr–Zn doped strontium hexa-ferrites for high frequency applications [13]. As much of our knowledge, the temperature dependent structural and electrical properties of Cr–Zn doped hexaferrites have not been reported in literature yet. In the present work, efforts have been made to investigate the doping effects of Cr$^{3+}$–Zn$^{2+}$ pair on electrical and dielectric properties of Strontium hexaferites such as AC conductivity ($\sigma_{AC}$), dielectric constant ($\varepsilon'$), dielectric loss tangent (tan δ) with respect to different temperatures. The value of exponent n, used in Jonscher’s power law showed CBH model. In order to distinguish the conduction processes, Cole–Cole plots (or Nyquist plots) were drawn for the different compositions of SrFe$_{12-2x}$Cr$_x$Zn$_x$O$_{19}$ (x = 0.0, 0.2, 0.4, 0.6, 0.8, 1.0) at different temperatures.

Experimental procedure

Cr–Zn doped M-type SrFe$_{12-2x}$Cr$_x$Zn$_x$O$_{19}$ with nominal composition (x = 0.0, 0.2, 0.4, 0.6, 0.8, 1.0) were prepared by using co-precipitation method. Appropriate quantities of highly pure precursors of Fe(NO$_3$)$_3$9H$_2$O, Cr(NO$_3$)$_3$9H$_2$O, Sr(NO$_3$)$_2$ and Zn(NO$_3$)$_2$6H$_2$O were weighed, separately dissolved in de-ionized water and then magnetically stirred. To attain the homogeneity, all individual solutions were mixed together and heated at 70°C on a hot plate. To maintain the pH level at 12, one molar solution of NaOH was added. In order to get the precipitates, the mixed solution was further heated up and magnetically stirred for one hour. Finally the precipitates produced were washed out using de-ionized water followed by drying in an oven at 105°C. The precipitation was done at 910°C for 20 min. Pellets were prepared using uniaxial press upon applying a compressed load of 100 Pa for ten minutes. The produced pellets were finally sintered at 920°C for ten minutes with temperature gradient of 10°C min$^{-1}$. The phase analysis of the produced samples was measured by using x-ray diffractometer (Model: PANalytical; Xpert Pro). The microstructure of the samples was measured by using Scanning electron Microscope. Dielectric measurements of ferrite samples were done using a Precision Component Analyzer (Model: Wayne Kerr; 6440B).

Results and discussion

Structural analysis of all the sintered samples of SrFe$_{12-2x}$Cr$_x$Zn$_x$O$_{19}$ showed hexagonal structure. Figure 1 shows indexed XRD pattern of Cr–Zn doped strontium hexaferites. A very small fraction of α-Fe$_2$O$_3$ phase was also observed in the prepared compositions and that is indicated by ‘*’ in the figure. The Miller indices (hkl) were obtained by comparing with JCPDS card number 00-033-1340 and 01-072-0739. Generally, lattice constants are calculated by hexagonal formula as given in equation (1).

$$\frac{1}{d^2} = 4 \left( \frac{h^2 + hk + kl}{a^2} \right) + \frac{l^2}{c^2}$$

(1)

By using Scherrer formula [11] crystallite size of the prepared samples was calculated. The mathematical formulism is given in the equation (2).

$$D = \frac{k\lambda}{\beta \cos \theta}$$

(2)

Whereas $k = 0.9$, $\beta$ is the full width at half maximum. $\lambda = 1.54$ Å. It is the X-ray wavelength and $\theta$ is the Bragg’s angle. The crystallite size was in the range 42–67 nm. The Lattice constant (a), (c), x-ray density ($\rho_a$), measured density ($\rho_m$), porosity (P), crystallite size of maximum intensity peak ($D_{max \text{ int}}$), volume of a unit cell (V) all are shown in the table 1. Lattice parameters showed nonlinear dependence on Cr-Zn substitution. The lattice constant of prepared samples were found to be decreased up to a certain composition i.e. $x = 0.6$. The reasons can be deformation in the ferrite lattice [14]. The volume of the unit cell is also decreased in it accordingly. After $x = 0.6$ compositions, the lattice constant starts increasing. The ionic radii of Fe$^{3+}$ (0.67 Å) is higher than the ionic radii of Cr$^{3+}$ (0.63 Å). Due to the replacement of charges between Fe$^{3+}$ and Cr$^{3+}$ the crystal structure of strontium hexaferites is contracted [15].

There are total five interstitial sites, three octahedral (B) and one bipyramidal (C) and one tetrahedral (A). It is reported in the literature that Cr$^{3+}$ preferably goes to octahedral (B) sites, however Zn$^{2+}$ goes to tetrahedral (A) or bipyramidal (C) sites where it replaces Fe$^{3+}$ ions. There are total 24 iron atoms distributed in these five
distinct interstitial sites. It has been reported that on replacing a cation in hexaferrites with another cation of different valance, the charge neutrality is maintained by exchange of Fe$^{3+}$ to Fe$^{2+}$ ions and vice versa. In the present case, Zn$^{2+}$ ions are replacing Fe$^{3+}$ ions and charge neutrality is adjusted with transfer of Fe$^{2+}$ ions from octahedral (B) site to Fe$^{3+}$ [13]. Moreover, the ionic radii of Zn$^{2+}$ (0.74 Å) is greater than the ionic radii of iron. It might be one of the reasons for the expansion of volume of the unit cell for composition $x = 0.8$ and $x = 1.0$.

It was also observed that the porosity of prepared samples is also decreased which indicates that the doped samples accepted the process of densification of the hexaferrite matrix. Due to heating process unavoidable pores are produced. These pores decreased the value of measured density as compared to that of x-ray density.

Figure 2 shows the SEM micrographs of powder samples of doped strontium hexaferrites. It clearly confirms the hexagonal structure for all the samples. The estimated particle size of powder sample was in the range of 8–11 μm. The calculated values of particle size of samples are mentioned in table 2.

**Electrical analysis**

**Dielectric constant and dielectric loss properties**

There are basic four types of polarizations. The interfacial (or Maxwell-Wagner polarization), dipole, ionic and electronic polarization. These polarizations mainly depend upon the applied frequency. At lower frequency, interfacial polarization becomes more dominant however at higher frequency ionic and electronic polarization plays important role. Dielectric materials are considered to be heterogeneous in nature due to interfaces and high resistivity. Such non-homogenous system is well defined by Maxwell-Wagner model [16, 17]. Such a behavior is already observed in hexaferrites [11]. Figure 3 shows the variation in dielectric constant ($\varepsilon'\varepsilon''$) for all the compositions prepared by co-precipitation method. It can be seen from the figure that on increasing the temperature, dielectric constant increases for all the compositions. At 1 kHz frequency and temperature 300°C composition ($x = 1.0$) showed maximum value of dielectric constant. Relaxation peaks can be observed for the prepared samples. These relaxation peaks are produced due to thermal energy provided to the holes and

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**Table 1.** The Lattice constant (a, c), x-ray density ($\rho_m$), porosity (P), maximum intensity crystallite size ($D_{\text{max int}}$), volume of unit cell (V) of sintered SrFe$_{12-2x}$Cr$_x$Zn$_x$O$_{19}$ samples.

| SrFe$_{12-2x}$Cr$_x$Zn$_x$O$_{19}$ | $x = 0.0$ | $x = 0.2$ | $x = 0.4$ | $x = 0.6$ | $x = 0.8$ | $x = 1.0$ |
|-------------------------------|----------|----------|----------|----------|----------|----------|
| Lattice constant a(Å)          | 6.0(1)   | 5.89(3)  | 5.78(1)  | 5.78(1)  | 5.87(1)  | 5.86(2)  |
| Lattice constant c(Å)          | 22.99(8) | 22.997(8)| 22.962(1)| 22.98(1) | 23.039(3)| 23.047(4)|
| Measured density $\rho_m$ (g cm$^{-3}$) | 2.36  | 2.35  | 2.25  | 2.61  | 2.80  | 3.37  |
| x-ray density $\rho_x$ (g cm$^{-3}$) | 5.79  | 4.90  | 5.35  | 5.32  | 5.50  | 5.75  |
| Porosity (%)                   | 59       | 52      | 58      | 51     | 49     | 41     |
| Maximum intensity peak crystallite size $D_{\text{max int}}$(nm) | 67     | 56     | 48     | 42     | 48     | 56     |
| Volume of unit cell (Å$^3$)    | 720.5(9)| 692.76(4)| 666.43(1)| 664.84(1)| 687.93(1)| 685.58(3)|
electrons. The thermal energy then shifts these peaks towards higher frequency and higher temperatures. In general, dielectric constant of the ferrites depends on temperature and the applied frequency. The dielectric properties are significantly influenced by the polarization phenomena. The plots in figure show that upon increasing the frequency, the polarization is reduced and as a result the dielectric constant of prepared samples is also decreased.

Figure 4 shows dielectric loss tangent (tanδ) for all the prepared Cr$^{3+}$–Zn$^{2+}$ compositions as a function of frequency and temperature. The plots of the figure show that the dielectric loss tangent decreases with increase in frequency for all the compositions. The hopping of electrons between Fe$^{2+}$ and Fe$^{3+}$ sites plays a pivotal role for

| Table 2. Particle size of powder samples (μm) of SrFe$_{12-2x}$Cr$_x$Zn$_x$O$_{19}$ samples. |
|---------------------------------------------------------------|
| SrFe$_{12-2x}$Cr$_x$Zn$_x$O$_{19}$       x = 0.0 | x = 0.2 | x = 0.4 | x = 0.6 | x = 0.8 |
| Particle size of powder samples (μm)     | 8      | 11      | 8       | 10      | 9       |
it. On increasing the frequency, electrons could not make successful hops and as a result loss tangent is decreased. It is also observed that on increasing Cr–Zn substitution the dielectric loss is increased.

Figure 5 shows variations in AC conductivity with respect to frequency and temperature. AC conductivity depends on hopping of electrons between octahedral and tetrahedral sites. It is evident from the plots that conductivity of the prepared samples increases on increasing frequency and temperature. At lower frequency, there is a plateau region. However as the frequency and the temperature increases, the successful hopping between octahedral (B) to octahedral (B) site also increased. This increased the conductivity of the material. Generally the conductivity can be defined as the sum of the hopping and the band conduction parts and the relation can be given as [18]

$$\sigma_{tot} = \sigma_o(T) + \sigma(\omega, T)$$

(3)

where \(\sigma_o(T)\) is the dc conductivity due to band conduction and \(\sigma(\omega, T)\) is ac conductivity due to hopping process.

Figure 3. Variation in dielectric constant ($\varepsilon'$) as a function of frequency and temperature.
The empirical formula for ac conductivity will be then written as

$$\sigma_{AC}(\omega, T) = B \omega^n$$

where $\omega$ is the frequency, $B$ and $n$ are constant and mainly depend on temperature and compositions. With the help of $n$, different theoretical models for conduction mechanism in the material can be proposed such that if the value of $n$ found to be decreased with temperature, Correlated barrier hopping (CBH) model can be employed [19]. In case, where $n$ depends on both temperature and frequency, and $n$ decreases with temperature with broad minima and then again starts increasing with increase in temperature, Overlapping Large Polaron Tunneling (OLPT) model can be proposed [20]. However, if $n$ is found to be increased with increase in temperature, the Non-overlapping Small Polaron Tunneling (NSPT) model can be proposed [21]. If $n$ is found to be independent of the temperature, then Quantum Mechanical Tunneling (QMT) model can be suggested [22]. In the present case, the value of $n$ can be calculated for each composition by using the relation in equation (4). The plot drawn between $\ln \sigma_{AC}$ versus $\ln \omega$ gives straight line. This has been shown in figure 6. The slope of the equation gives the value of exponent $n$. The value of $n$ for our prepared samples ranges between 0.04–0.8 as described in table 3. It is found that the value is decreasing with increasing the temperature. We assume that CBH model is most relevant.
to our case. In the table 3 values of n have been calculated for each composition at different temperatures. The above mentioned calculated value of n is in accordance with Jonscher’s power law [18].

**Impedance analysis**

In order to understand the electrical properties of the material, complex impedance measurement were taken with respect to frequency and temperature. The real and imaginary parts of impedance can be calculated using formula $z' = z \cos \theta$ and $z'' = z \sin \theta$ [23, 24]. The complex part consists of $z = z' + iz''$ where, $z'$ is the real part and it is related to the pure resistance of the material and $z''$ is imaginary part of complex impedance and it is related to the capacitance C of the material, where $\omega = \frac{1}{\sqrt{LC}}$ [25]. Figure 7 shows the variation of real part of complex impedance with respect to frequency and temperature. It was observed that for all the compositions, the value of the impedance is decreased with increasing frequency and temperature. At low frequencies, the high value of $Z'$ was observed which may be due to all types of polarizations. By increasing temperature, $Z'$ was observed to be decreased. This showed negative temperature co-efficient of resistance (NTCR) type behavior in our prepared materials. There are two regions observed in the graphs, dispersive region and the plateau region.

![Figure 5. Variations in AC conductivity with respect to frequency and temperature.](image-url)
These regions are observed due to the relaxation effect of grain and grain boundaries at high and low frequencies. In high frequency region for all temperature the merging of plots observed. This was might be due to the space charge polarization effect which finally decreased the barrier potential in our prepared samples. Such a behavior is already reported in magnetite [26]. The independent plateau region showed the dc resistance of the material. The motion of the charge carriers is responsible for the shape of real impedance curve. It is observed that there is a linear fall at room temperature and at 100 °C for compositions x = 0.0, 0.6, and 1.0. It is in accordance with the ac conductivity graphs of prepared samples. We see that as the conductivity increases linearly, the resistivity of the material also decreases linearly. Such a behavior is already reported in strontium hexaferrites [11].

The successful hoping of charge carriers plays a key role in long range movements. In fact, long range movements of charge carriers are caused by their successful hoping mechanism. During these successful hoping, the neighboring charged particles stay relaxed to their positions which leads to generation of dc resistivity and that in return results dc resistivity, in impedance plots [27]. Similarly, localized movements are caused by the unsuccessful hoping of charged carriers. During these unsuccessful hoping, charged carriers themselves, keep on relaxing at their own positions [28]. It leads in generation of ac conductivity which in turn imparts a dispersive trend to the impedance plots. These dispersive regions in the plots are associated with conducting behavior of charge carriers in the material and that is why they almost attain the shape of ac conductivity [27,28].

Inhomogeneous material consists of grain and grain boundaries. Intrinsic and extrinsic properties of the material can be explained on the basis of conduction phenomenon. Conduction at the interfaces of the material is considered to be as extrinsic property of the material, whereas the conduction in grains is referred as intrinsic property [29]. In figure 8, the variation of the imaginary part (z”) of the impedance is shown. It was observed that by increasing the frequency and temperature the value of reactive (imaginary) part decreases for all the compositions. At a characteristic frequency, also known as relaxation frequency, relaxation peaks were observed for 0.6 and 0.8 compositions which seem to be moving towards higher frequency regions. The characteristic or peak frequency is an intrinsic property of the material which does not dependent on the material’s geometric
factors. This peak frequency tend to shift towards high-frequency region which is also applicable to our case for 
\(x = 0.6\) and \(x = 0.8\) compositions.

**Cole-cole plots**
In order to distinguish the conduction processes, Cole-Cole plots (or Nyquist plots) were drawn between \(z''(\omega)\) versus \(z'(\omega)\) for the different compositions of \(\text{SrFe}_{12-2x}\text{Cr}_x\text{Zn}_x\text{O}_{19}\) \((x = 0.0, 0.2, 0.4, 0.6, 0.8, 1.0)\) at different temperatures. The Cole-Cole plots are shown in figure 9. Due to different relaxation behavior, the arcs and semicircles were produced at different temperatures. Conduction mechanism can be explained by the growth of these arcs. It was seen that for all the compositions at RT and at \(100^\circ\text{C}\) unsymmetrical and depressed semicircle arcs are produced. It indicates the non-Debye type relaxation process in hexaferrites [27]. In Nyquist plots, bulk resistance \(R_b\) of the material can be determined by the intercepts of the semicircle on real axis \((Z')\). It is observed that upon increasing the temperature, the diameter of the semicircles is decreased which tend to shift towards the origin. The decrease in the bulk resistance \(R_b\) with temperature suggests NTCR behavior in the material [30]. As the temperature was increased, the value of the radius of the arcs was also decreased which
suggests the semiconducting nature of the material. According to our assumption, the larger semicircle arcs at room temperature showed grain boundary effect. It implies that as the temperature gets rise, accumulation and activation of thermally generated charge carriers takes place at different interfacial discontinuities. Each semicircle conforms to a unique relaxation process. In case, three semicircles arcs are appeared, the one at higher frequency region conforms to grain effect, the intermediate arc describes grain boundary effect and semicircle arc related to lower frequency zone is due to surface effect. The arc radii are the measure of resistances attributed by different regions of material. For the case of a perfect semicircle having center at abscissa, it indicates a Debye-type conduction process.

Figure 10 shows the Cole-Cole (Nyquist plots) for the prepared samples at temperature 300 °C. The electrical behavior of these samples has been carried out with the help of Zview software. The RC circuits were obtained to understand the microscopic pictures of the material. For sample composition x = 0.4, the fitted model was (RQ)(RC) however for the rest of the compositions (x = 0.0, 0.2, 0.6, 0.8, 1.0) it followed (RQ)(RQ), whereas R is the resistance, C is the capacitance and Q is the Constant Phase Element (CPE). This element is mostly used to tune the non-ideal capacitive behavior in the circuits. The values obtained from fitting

Figure 8. Variation of imaginary part of complex impedance with respect to frequency and temperature.
parameters at 300 °C temperature are also given in table 4. It is observed that for all the prepared samples grain boundary effect is dominant at 300 °C.

Conclusions

M-type SrFe$_{12-2x}$Cr$_x$Zn$_x$O$_{19}$ ($x = 0.0, 0.2, 0.4, 0.6, 0.8, 1.0$) were prepared by co-precipitation method. XRD analysis confirmed hexagonal structure for strontium hexaferrite. Dielectric properties as a function of frequency and temperature were measured. The frequency range was from 1 kHz to 3 MHz and the temperature range was RT to 300 °C. According to SEM micrographs, the estimated particle size of powder samples was in the range of 8–11 μm. Dielectric constant and loss tangent were increased with temperature and decreased with increase in frequency. AC conductivity of all the prepared samples was increased upon increasing the frequency and temperature. The value of $n$ ranges between 0.04–0.8. The prepared samples were not strictly composition dependent but other factors like porosity, site occupancy etc are also playing important role. Each composition behaved different with respect to frequency and temperature. At 1 kHz frequency and temperature 300 °C composition ($x = 1.0$) showed maximum value of dielectric constant. Such a composition can be used for
capacitors where high energy storage is required. The real and imaginary parts of complex impedance are also measured. It is observed in real part of impedance that for compositions $x = 0.0, 0.6, \text{ and } 1.0$ there is a linear fall at room temperature (RT) and at 100 °C. It is in accordance with the ac conductivity graphs of prepared samples.

Figure 10. The Cole-Cole (Nyquist plots) for the prepared samples at temperature 300 °C.

Table 4. The values obtained from fitting parameters at 300 °C temperature using Zview software for SrFe$_{12-2x}$Cr$_x$Zn$_x$O$_{19}$ ($x = 0.0, 0.2, 0.4, 0.6, 0.8, 1.0$).

| Different parameters | x = 0.0 | x = 0.2 | x = 0.4 | x = 0.6 | x = 0.8 | x = 1.0 |
|----------------------|---------|---------|---------|---------|---------|---------|
| R1                   | 164790  | $2.022 \times 10^6$ | 29195   | 43027   | 19327   | 4984    |
| R2                   | $2.0 \times 10^6$ | 16785   | 16483   | 135040  | 29321   | 21231   |
| Q1                   | $2.282 \times 10^{-11}$ | $6.289 \times 10^{-11}$ | $1.834 \times 10^{-9}$ | $4.614 \times 10^{-10}$ | $2.06 \times 10^{-10}$ | $1.040 \times 10^{-9}$ |
| Q2                   | $2.639 \times 10^{-11}$ | $9.791 \times 10^{-12}$ | —       | $3.765 \times 10^{-12}$ | $1.0 \times 10^{-20}$ | $2.444 \times 10^{-11}$ |
| C1                   | —       | —       | —       | $2.542 \times 10^{-11}$ | —       | —       |
| $\tau$               | $2.40 \times 10^{-6}$ | $2.60 \times 10^{-5}$ | $8.58 \times 10^{-7}$ | $6.91 \times 10^{-7}$ | $5.10 \times 10^{-7}$ | $1.51 \times 10^{-7}$ |
The Cole-Cole plots are also drawn to understand the conduction mechanism for hexagonal ferrites. At room temperature and at 100 °C unsymmetrical and depressed semicircle arcs were produced for all the samples that indicate the presence of non-Debye type relaxation process. The electrical behavior of these samples were studied using Zview software. The RC circuits were obtained to understand the microscopic pictures of the material. It can be seen that for sample composition x = 0.4, the fitted model was (RQ)(RC) however for the rest of the compositions (x = 0.0, 0.2, 0.6, 0.8, 1.0), it followed (RQ)(RQ) model. It is observed that for all the prepared samples grain boundary effect is dominant at 300 °C.

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