

Article

Experimental Investigations of the AC-Conductivity in NaTaO₃ Ceramic Materials Doped with Cu and Al Metal Ions

Iosif Malaescu ^{1,2} , Paula Sfirloaga ³  and Catalin Nicolae Marin ^{1,*}

¹ Faculty of Physics, West University of Timisoara, Bd. V. Parvan No. 4, 300223 Timisoara, Romania; iosif.malaescu@e-uvvt.ro

² Institute for Advanced Environmental Research, West University of Timisoara (ICAM-WUT), Oituz Str., No. 4, 300086 Timisoara, Romania

³ National Institute of Research and Development for Electrochemistry and Condensed Matter, 144 Dr. A. P. Podanu, 300569 Timisoara, Romania; paulasfirloaga@gmail.com

* Correspondence: catalin.marin@e-uvvt.ro

Abstract: Two ceramic samples of sodium tantalate (NaTaO₃), doped with metal ions of copper (Cu; sample S1) or aluminum (Al; sample S2), were obtained by the sol-gel method. Complex impedance measurements in the frequency range (200 Hz–2 MHz) and at temperatures between 30 °C and 90 °C allowed identification of a transition temperature from semiconductor-type behavior to conductor-type behavior for each sample (52 °C for sample S1 and 54 °C for sample S2). In the temperature range with semiconductor behavior, the activation energy of each sample was determined. Based on the Mott's variable-range hopping (VRH) model, the density of localized states at the Fermi level, $N(E_F)$, the hopping distance (R) and the hopping energy (W) were determined, for the first time, on NaTaO₃ samples doped with Cu or Al metal ions. The increase in $N(E_F)$ of sample S2 compared to $N(E_F)$ of sample S1 was explained by the decrease in the hopping distance of charge carriers in sample S2 compared to that in sample S1. Additionally, using the correlated barrier hopping (CBH) model, the energy band gap (W_m) and the hopping (crossover) frequency (ω_h) at various temperatures were determined. Knowledge of these electrical properties is very important for explaining the electrical conduction mechanisms in metal ion-doped compounds, with perovskite structure being of interest for the use of these materials in the conversion of thermoelectric energy, photocatalytic applications, electronics or other applications.

Keywords: sodium tantalate; perovskites; complex impedance measurements; DC and AC conductivity; VRH and CBH theoretical models



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1. Introduction

An important category of ceramic materials that has been extensively studied in recent years includes perovskite oxides with the general chemical formula $A^{2+}B^{4+}(X^{2-})_3$, where A and B are cations and X is an anion, typically oxygen. These materials can be used in various applications such as energy conversion and storage [1,2], photocatalytic applications [3], photovoltaic and solar cells [4], memory devices, sensors, detectors and multilayer capacitors [5,6] or tunable microwave devices [7]. In the development of electronic and microelectronic devices, a series of lead compounds (such as lead titanate, lead zirconate titanate and lead magnesium niobate) has been extensively used [8]. The broad range of applications of perovskites is due to their physicochemical properties, which can be controlled by varying the chemical composition, particle size or technological parameters of material manufacture [9,10]. Using the resistive switching property of perovskites, it was possible to investigate the possibility of making new electronic devices, such as memristors [11], artificial synapses [12], as well as applications in storage, sensing and display information.

At present, other ceramic materials have been achieved, such as tantalates, with different perovskite structures and enhanced features compared to compounds containing

lead, which can also be used in applications. Tantalates of $ATaO_3$ -type ($A = Li, Na, \text{ and } K$) [13] have important photocatalytic properties [14,15] and $NaTaO_3$ is currently considered a potential candidate for thermoelectric materials [16,17]. Other authors [18,19] show that $NaTaO_3$ has ferroelectric characteristics. The electronic structure of $NaTaO_3$, can be changed by doping with metallic ions (such as Ag, Fe, Al, Cu, etc.), which leads to the modification of its electrical and optical properties [20,21]. At the same time, in Ref. [22], the authors studied the effect of doping with Fe ions on the structural, dielectric and magnetic properties of the $SrTiO_3$ perovskite material. The results obtained by investigating the dielectric parameters in the frequency range from 30 Hz to 1 MHz [22] showed that the values of the dissipation factor are high at low frequencies. This behavior is due to the grain boundaries of high resistance, which are more important than the grains [23]. Few papers are known regarding the determination of the electrical properties, such as electrical conductivity, of these perovskite compounds.

In this paper, the effect of doping sodium tantalate ($NaTaO_3$) with Cu or Al metal ions on the frequency and temperature dependence of the electrical conductivity $\sigma(f,T)$ is investigated by means of complex impedance measurements in the frequency range of 200 Hz–2 MHz and at different temperatures from 30 °C to 90 °C. Using these measurements, the aim of the paper is to determine the static (DC) and dynamic (AC) electrical conductivity of the synthesized materials, their transition temperature from semiconducting to conducting behavior, and to determine the semiconducting properties, based on the variable range hopping (VRH) and correlated barrier hopping (CBH) theoretical models.

Knowledge of the electrical conductivity of these ceramic materials, in addition to their morpho-structural analysis, is very useful to obtain information regarding the possibility of using these materials in different fields of applications [11,24].

2. Materials and Methods

2.1. Synthesis

Using the sol-gel method, two samples were synthesized: a sample of sodium tantalate ($NaTaO_3$) doped with Cu metal ions ($NaTaO_3:Cu$), hereafter named sample S1 (SG-Cu) and another sample doped with Al ions ($NaTaO_3:Al$), hereafter referred to as sample S2 (SG-Al). The amount of dopant ions of Cu and Al was 1%, in a weight ratio. For the synthesis of samples, purely analytical chemicals (99.98%) were used (purchased from Aldrich, St. Louis, MO, USA). Tantalum ethoxide ($Ta(OC_2H_5)_5$) (1 mL), solution of ethyl alcohol (1:1) and distilled water were used. The precipitation was carried out at room temperature, with sodium hydroxide solution (1M NaOH), at pH = 7, under continuous magnetic stirring. For the synthesis of $NaTaO_3$ doped with Cu or Al, 0.04 g of $Cu(NO_3)_2$ and 0.04 g $Al(NO_3)_3$, respectively, were added to the suspension, under continuous stirring. The resulting mixtures were heated for 2 h at a temperature of 90 °C and the products thus obtained, in the form of a gel, were then dried in an oven at 110 °C for 4 h. Finally, for crystallization and obtaining the perovskite phases, $NaTaO_3$ samples doped with Cu and Al were subjected to heat treatment for 6 h at a temperature of 600 °C, with a heating rate of 5 °C/min. The materials thus obtained were in powder form.

2.2. Characterization Techniques

The phase analysis of the prepared samples was carried out based on the X-ray diffraction pattern, and recorded with a Bruker-AXS D8 (Billerica, MA, USA) Advance diffractometer with $CuK\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$, Zr filter on the diffracted beam, 40 kV and 40 mA). The diffractometer was operated in constant scan mode over the range of $10^\circ \leq 2\theta \leq 85^\circ$, with zero point determined from an external quartz standard. The position and width of the diffraction maxima were determined using the evaluation package DIFFRACPLUS software 12.0. The morphological analysis of the samples was performed with an FEI Inspect S microscope model scanning electron microscopy (SEM), which also has the capability of performing the elemental analysis and mapping of samples using the Energy Dispersive X-ray Analysis (EDX) facility. The optical studies were carried out

using the UV-VIS Spectrophotometer type DRUV-VIS Lambda 950 Perkin-Elmer (Waltham, MA, USA), which works in the wavelength range of 250–2500 nm allowing both reflection and diffuse transmission measurements. The electrical conductivity of prepared samples was determined based on the complex impedance measurements in the frequency range 200 Hz–2 MHz and at different temperatures between 30 and 90 °C, using an LCR meter (Agilent (Santa Clara, CA, USA) E-4980-A type, in conjunction with a laboratory experimental setup [25], similar to ASTM D150-98 [26]). This LCR meter can perform measurements in the frequency range 20 Hz–2 MHz, but the choice of measurement range depends on the device under test (DUT). For reliable measurements, the quality factor of DUT must be greater than 10. Thus, for our measuring cell, the measuring frequency range is 200 Hz–2 MHz.

3. Results

3.1. X-ray Diffraction Analysis

Figure 1 shows the X-ray diffraction patterns of the sodium tantalate samples doped with Cu and Al metal ions.

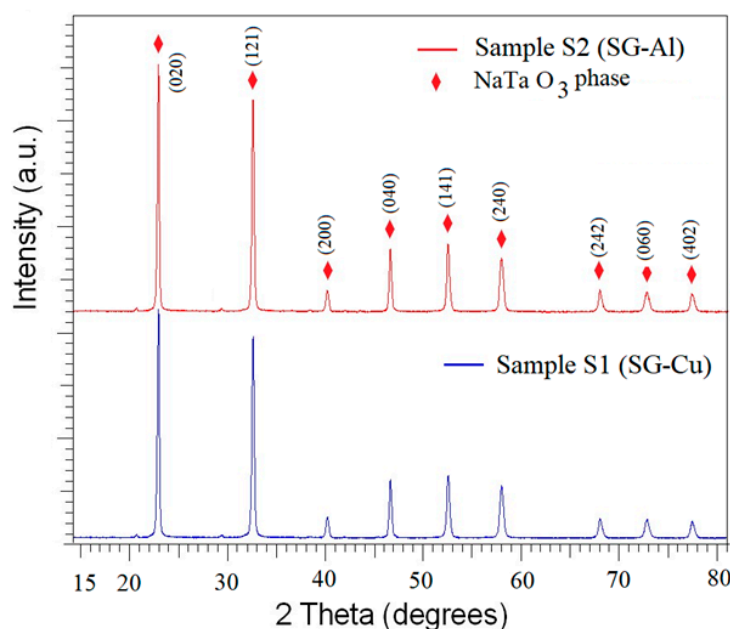


Figure 1. The XRD patterns for the S1 (SG-Cu) and S2 (SG-Al) samples.

From Figure 1 it is observed that the XRD patterns of the samples are similar and they present the diffraction peaks corresponding to the well crystallized single phase NaTaO₃ (JCPDS 73-0878). The X-ray diffraction data (Figure 1) allowed a good determination for the full width at half maximum (FWHM), β , of several diffraction maxima that present sufficient width: (020), (121), (040), (141) and (240), thus being able to calculate the average crystallite diameter of samples using the Debye–Scherrer equation. The results are listed in Table 1.

Table 1. Structure parameters obtained from X-ray diffraction analysis for the investigated samples.

Sample	Mean Diameter (nm)	Lattice Parameters
S1 (SG-Cu)	39.4	$a = 5.5140 \text{ \AA}$; $b = 7.7510 \text{ \AA}$; $c = 5.4950 \text{ \AA}$
S2 (SG-Al)	37.7	$a = 5.5138 \text{ \AA}$; $b = 7.7508 \text{ \AA}$; $c = 5.4940 \text{ \AA}$
NaTaO ₃	-	$a = 5.5130 \text{ \AA}$; $b = 7.7503 \text{ \AA}$; $c = 5.4941 \text{ \AA}$

The X-ray powder diffraction spectra of SG-Al and SG-Cu samples were indexed using the EXPO2009 computer program (Version 2009) [27], leading to the lattice parameters

shown in Table 1, the results are consistent with the space group Pcmn (space group number 62), belonging to the orthorhombic crystallographic family. At the same time, in Table 1 we presented the values of the lattice parameters for undoped NaTaO_3 , obtained with the VESTA program. It can be observed that by introducing Cu and Al dopant ions, the lattice parameters change, which proves the presence of dopants in the crystalline lattice. The values obtained for the lattice parameters are very close to those obtained by other authors [28].

3.2. Scanning Electron Microscopy

The SEM images, the EDX spectrum, elements quantification and mapping of the Al or Cu doped NaTaO_3 ceramic materials synthesized by sol-gel method, are shown in Figure 2.

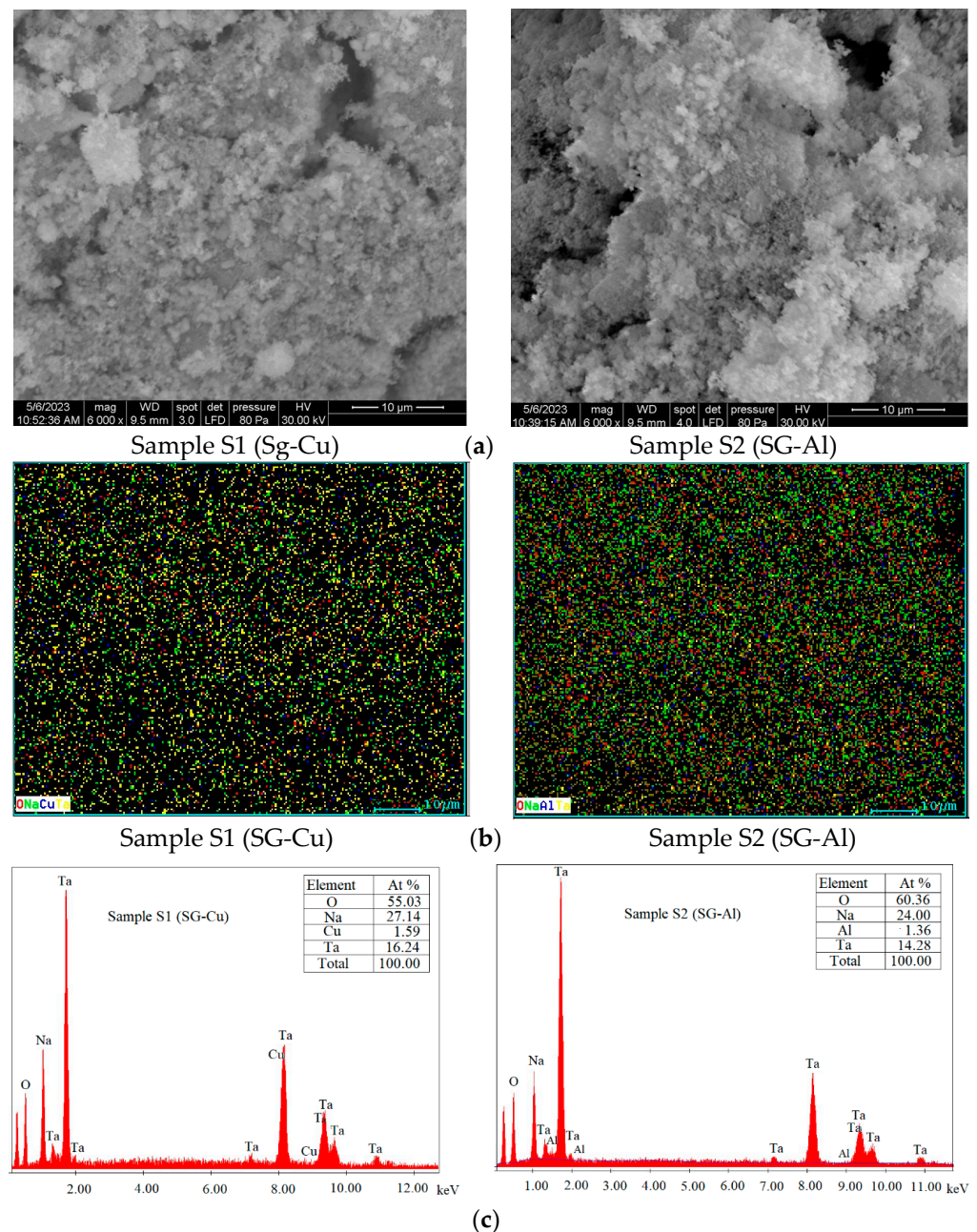


Figure 2. SEM images (a), element's mapping (b), and EDX spectrum (c), for samples S1 and S2.

From the SEM images of samples (Figure 2a), one can observe that for both samples the particles have a spherical shape, with dimensions of hundreds of nanometers, and are

strongly agglomerated. Also, element's mapping (Figure 2b) revealed uniform distribution of all present elements: O, Na, Ta and metal ions used as dopant (Cu or Al).

3.3. UV-VIS Spectroscopic Analysis

Figure 3 shows the UV-VIS absorption spectra of the samples. The wavelength λ_g , at the end of the absorption range was determined as the intersection between the wavelength axis and the tangent line drawn on the absorption spectrum. As can be seen from Figure 3, the λ_g values of the two samples are $\lambda_g = 326$ nm for sample S1 (Sg-Cu) and $\lambda_g = 322$ nm for sample S2 (SG-Al). Using the Kubelka–Munk method [29], the values of λ_g from Figure 3 and Equation (1), the values of the band gap energy (E_g) of the investigated samples, were computed [30].

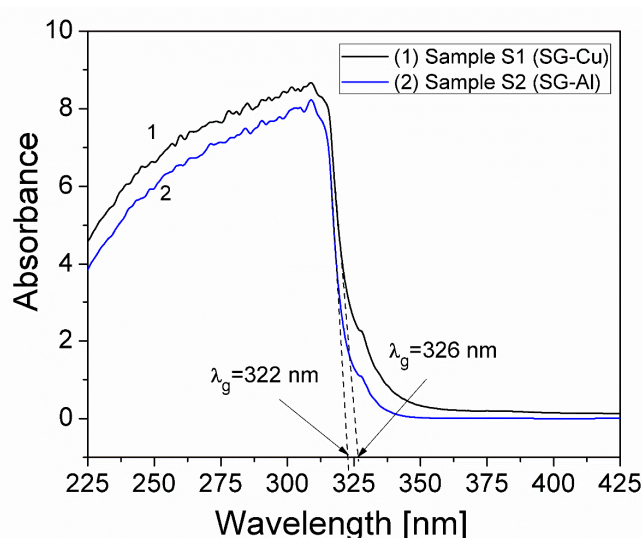


Figure 3. Diffuse reflectance spectra of the prepared samples: sample S1 (SG-Cu) and sample S2 (SG-Al).

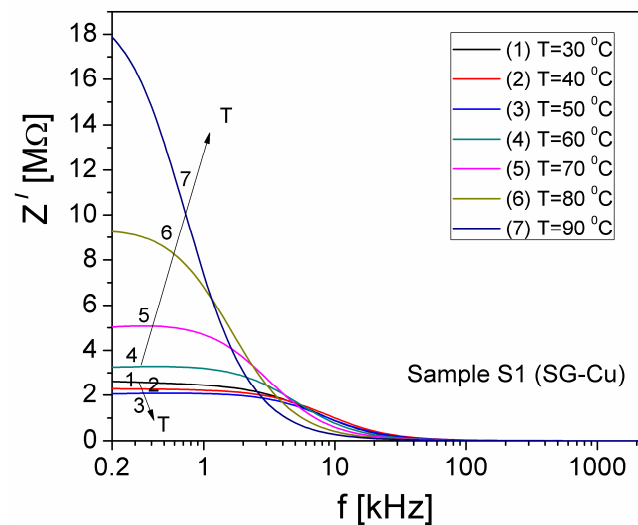
$$E_g(\text{eV}) = \frac{1240}{\lambda_g(\text{nm})} \quad (1)$$

The obtained values are $E_g = 3.80$ eV for sample S1 (SG-Cu) and $E_g = 3.85$ eV for sample S2 (SG-Al), which is smaller than the value reported in Ref. [30] (i.e., 4 eV) for a NaTaO_3 obtained by the hydrothermal method. Therefore, the presence of the dopants (Cu or Al) in the structure of NaTaO_3 leads to a decrease in the band gap energy, E_g , which can be useful for photocatalysis. The obvious decrease of the band gap energy (E_g) compared to that of pure NaTaO_3 , demonstrates an improved absorption in the visible (infrared) light range, and an increase in carrier mobility for each doped structure [31]. Studies have shown [31,32] that NaTaO_3 is the first semiconductor photocatalyst that produces infrared absorption when doped with metal cations, which leads to a decrease in the band gap energy. At the same time, doping with metal ions reduces the rate of electron-hole recombination and consequently the population of charge carriers in the equilibrium state increases [32].

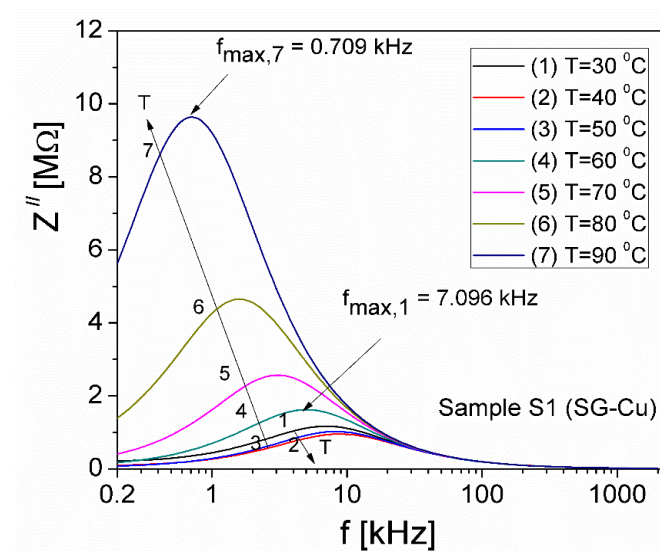
3.4. Electrical Properties

3.4.1. Complex Impedance

The frequency and temperature dependencies of the Z' and Z'' components of the complex impedance of the samples, in the frequency range 200 Hz–2 MHz and at different temperatures between 30 and 90 °C are shown in Figure 4 for sample S1 (SG-Cu) and in Figure 5 for sample S2 (SG-Al).



(a)



(b)

Figure 4. Frequency and temperature dependence of the real component Z' (a), and the imaginary component Z'' (b), of the complex impedance of sample S1 (SG-Cu).

As can be seen from Figures 4a and 5a, for a constant temperature T , the real component Z' of the complex impedance decreases with increasing frequency, for both samples. At low frequencies, between 200 Hz and 600 Hz, by increasing the temperature up to about 40–50 °C, it is observed that Z' decreases slightly, then starts to increase significantly, up to values of 18 MΩ for sample S1, SG-Cu and up to 33 MΩ for sample S2, SG-Al, by increasing the temperature in the range 50–90 °C.

From Figures 4b and 5b one can observe that the imaginary component Z'' presents a maximum at frequency $f_{max,Z}$ for both samples and for all investigated temperatures. For both samples, $f_{max,Z}$ shifts towards higher values when the temperature increases up to approximately 50–60 °C, after which the temperature continues to increase up to 90 °C, $f_{max,Z}$ moves towards smaller values.

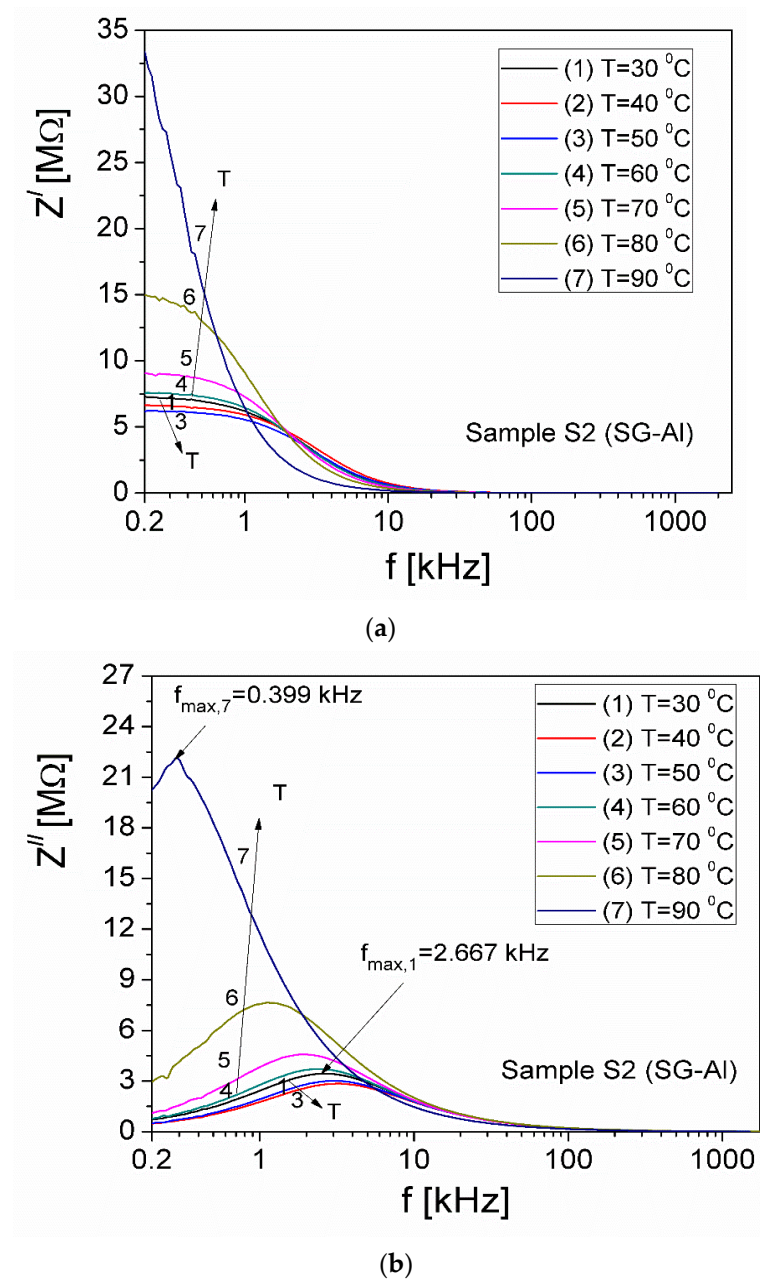


Figure 5. Frequency and temperature dependence of the real component Z' (a), and the imaginary component Z'' (b) of the complex impedance of sample S2 (SG-Al).

This behavior of Z'' may be due to the perovskite-type structure of the two samples, highlighting the existence of an electrical relaxation process [33], which is due to the presence of charge carriers in the investigated samples [34,35]. The relaxation process is determined by the hopping of the charge carriers between localized states, according to the VRH model of Mott and Davis [34]. Based on the experimental values, $f_{max,Z}$ from Figures 4b and 5b, and on the Debye equation, $\tau_Z = 1/2\pi f_{max,Z}$ [36], we computed the relaxation times τ_Z , of the two samples at each temperature T . The variation of the relaxation time τ_Z , over the measurement temperature intervals, agrees with an Arrhenius type law:

$$\tau_Z = \tau_0 \exp\left(-\frac{E_{activ.}}{kT}\right) \quad (2)$$

where, τ_0 represents the pre-exponential factor of the relaxation time; k is the Boltzmann's constant and $E_{activ.}$ is the activation energy or the barrier energy between the localized states [34,37].

Figure 6 shows the plot of the experimental dependence, $\ln(\tau_Z)$, on $(10^3/T)$ for the investigated samples. The linear fit of the curves in Figure 6 provides two different slopes for each sample, which correspond to the two temperature ranges: below 52 °C and above 52 °C (for sample S1) and below 54 °C and above 54 °C (for sample S2), respectively. These temperature dependencies of τ_Z could be associated with two ranges of different electrical conductivity of the samples [38]. From the fitting with a straight line, of equation $y = a + bx$, where a is the intercept and b is the slope of the line, we determined the values of the activation energy, $E_{activ.}$, of samples S1 and S2, corresponding to the two temperature ranges. The obtained values are 0.046 eV (below 52 °C) and 0.602 eV (above 52 °C) for sample S1 (SG-Cu) and 0.020 eV (below 54 °C) and 0.461 eV (above 54 °C) for sample S2 (SG-Al), respectively. As a result, it can be stated that for the samples of NaTaO₃ doped with Cu and Al ions respectively, we highlighted for the first time a transition from a semiconductor-like behavior to a conductor-like behavior, at a temperature of 52 °C for sample S1 (SG-Cu) and a temperature of 54 °C for sample S2 (SG-Al) respectively, where the change in the activation energy, $E_{activ.}$, also appears (see Figure 6).

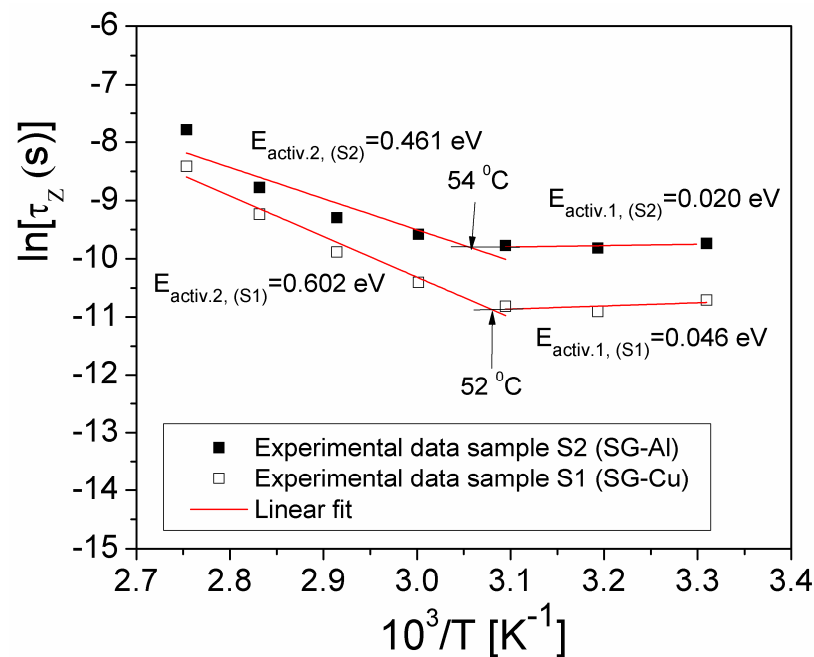


Figure 6. The $\ln(\tau_Z)(T^{-1})$ plots for sample S1 (SG-Cu) and sample S2 (SG-Al).

3.4.2. DC and AC Conductivity

Using the experimental values of the Z' and Z'' components of the complex impedance of the samples (Figures 4 and 5), we computed the electrical conductivity σ , with the relation:

$$\sigma = \left(\frac{d}{A} \right) \cdot \frac{Z'}{Z'^2 + Z''^2} \quad (3)$$

where d is the thickness of sample and A is its cross-sectional area.

The frequency dependence of the conductivity, σ , of the investigated samples over the frequency range (200 Hz–2 MHz) and at different temperatures between 30 and 90 °C, is shown in Figure 7a for sample S1 (SG-Cu) and Figure 7b for sample S2 (SG-Al), respectively.

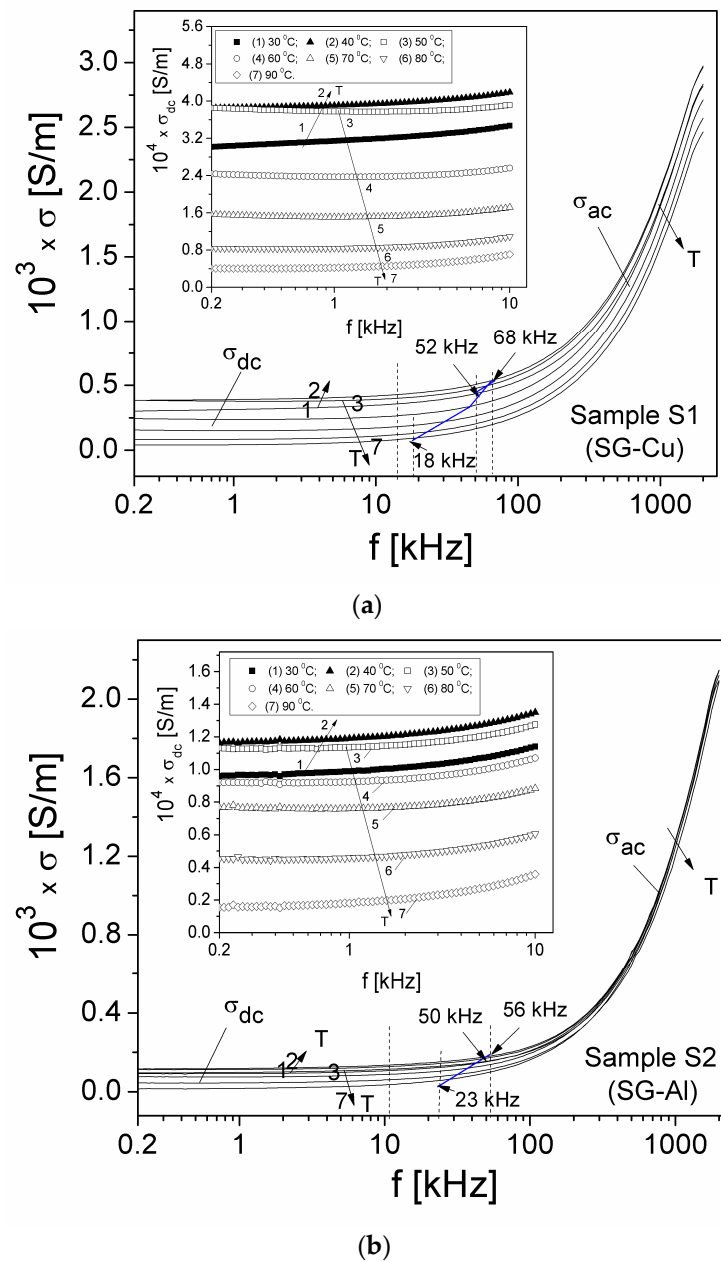


Figure 7. The frequency dependence of the electrical conductivity of the samples: (a) S1 (SG-Cu), and (b) sample S2 (SG-Al) at different temperatures.

As can be seen from Figure 7, for each sample, the conductivity consists of two components according to Jonscher's theoretical model [39], where the total conductivity is given by the equation:

$$\sigma(\omega, T) = \sigma_{dc}(\omega, T) + \sigma_{ac}(\omega, T) \quad (4)$$

The component denoted by σ_{dc} is called static conductivity (or DC conductivity) and the second component denoted by σ_{ac} is called dynamic conductivity (or AC conductivity). At the same time, from Figure 7a,b it can be observed that σ_{dc} remains nearly constant for each temperature, T , up to the frequency of approximately 10 kHz, after which a rapid increase follows for both samples. In the inset of Figure 7a,b, the frequency dependence of σ_{dc} , over the range 200 Hz–10 kHz and at different temperatures between 30 and 90 °C is presented. Based on the values obtained for the static conductivity, σ_{dc} , from Figure 7a,b, at every temperature, T , at which the measurements were performed, we plotted the temperature dependence of the static conductivity, $\sigma_{dc}(T)$, for both samples, as shown in Figure 8.

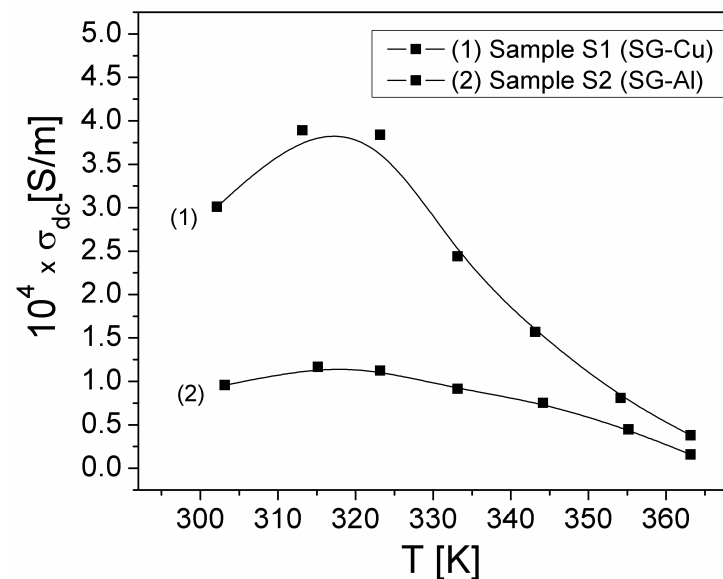


Figure 8. Temperature dependence of the static conductivity σ_{dc} of the samples.

From Figure 8, it is observed that $\sigma_{dc}(T)$, increases when the temperature increases from 30 °C to approximately 50 °C, then decreases with the increase in temperature from 50 °C to 90 °C, this behavior agrees with the VRH theory of Mott [34]. The increase in conductivity, σ_{dc} , in the region of low temperatures, between 30 and 50 °C, corresponds to semiconductor type behavior. This increase can be attributed to the increase in the drift mobility of electric charge carriers, which are thermally activated when the temperature increases [33]. As a result, it can be highlighted that, around the temperature of 50 °C, there is a transition from a semiconductor-type behavior to a conductor-type behavior in the investigated samples. At temperatures above 50–60 °C, the static conductivity decreases with the increase in temperature, indicating that the samples behave like conductors [40].

For both investigated samples, over the temperature range 30–50 °C, where they have a semiconductor-type behavior, based on Mott's VRH model [34], the temperature dependence of the static conductivity, σ_{dc} is given by the equation:

$$\sigma_{dc} = \sigma_0 \exp\left(-\frac{D}{T^{1/4}}\right) \quad (5)$$

In Equation (5), σ_0 is the pre-exponential factor with dimensions of $\Omega^{-1}\text{m}^{-1}$ and D is given by the relation:

$$D = \frac{4E_{cond}}{kT^{3/4}} \quad (6)$$

Figure 9 shows the experimental dependence $\ln(\sigma_{dc})(1/T^{1/4})$, for the two samples at low frequencies. The linear fit of the curves in Figure 9, corresponding to both sample S1 (SG-Cu) and sample S2 (SG-Al), shows that the slope of the conductivity changes at $T = 54$ °C (for sample S1) and at $T = 56$ °C (for sample S2). This result is in agreement with the variation of the relaxation time, τ_Z with temperature, shown in Figure 6, which was associated with two ranges of different electrical conductivity of the samples. Thus, the result obtained in Figure 9, highlights once again the fact that the temperatures of 54 °C for sample S1 (SG-Cu) and 56 °C for sample S2 (SG-Al), represent the transition temperature from a semiconductor-type behavior to a conductor-type behavior, also taking place a change of the activation energy of conduction, $E_{a,cond}$, of the samples, as we also obtained in Figure 6. At the same time, by increasing the temperature to approximately 50–60 °C, the doping ions of Cu and Al, respectively, induce in the NaTaO₃ structure, a transition from the semiconductor-like to conductor-like behavior of the TaO compounds [41]. Considering the paper [41], we can say that in the case of NaTaO₃ samples doped with Cu ions (sample

S1) and respectively with Al ions (sample S2), Cu-Ta and Al-Ta bonds are formed on the TaO surface, which introduce holes as the main charge carrier and, consequently, the Fermi level is shifted into the valence band, where electronic states are now populated. As a result, it can be said that in the analyzed samples a metallic transition takes place at a temperature located between 50 and 60 °C, called transition temperature, thus highlighted for the first time in perovskite compounds of this type.

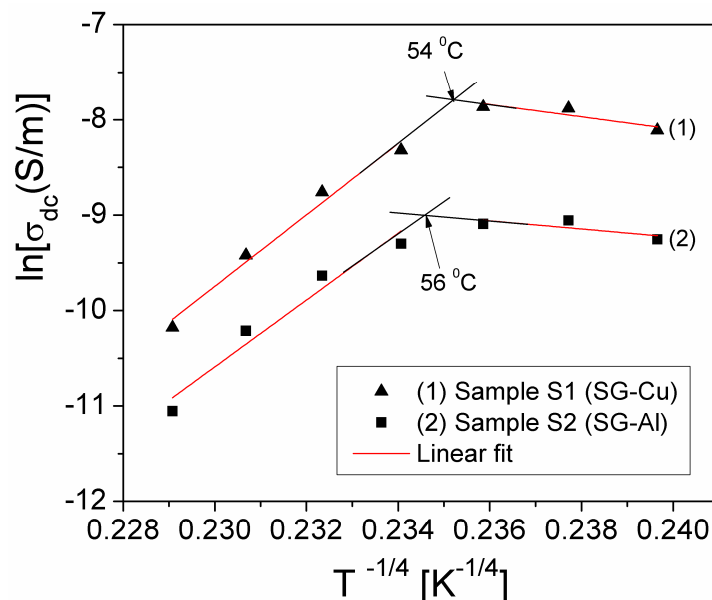


Figure 9. Plot of dependence, $\ln(\sigma_{dc})(1/T^{1/4})$, for the investigated samples at low frequencies.

From the straight-line fitting of the experimental dependence, $\ln(\sigma_{dc})(1/T^{1/4})$, from Figure 9, for each sample, we determined the slope D, corresponding to the interval (30–54 °C), for sample S1, and for the temperature range (30–56 °C) for sample S2, respectively, in which the samples show a semiconductor-like behavior. Then with the relation Equation (6) we computed the activation energy of conduction, $E_{a,cond}(T)$, for each sample. Figure 10 shows the temperature dependence $E_{a,cond}(T)$, for the S1 (SG-Cu) and S2 (SG-Al) samples.

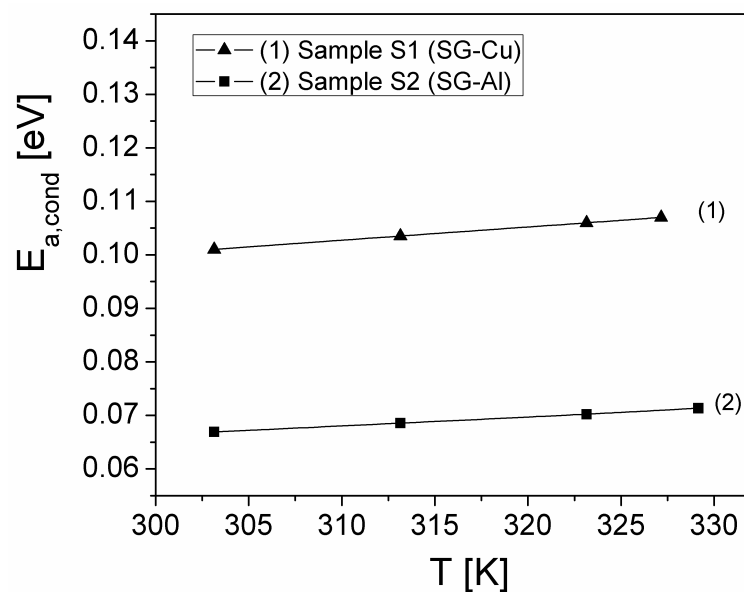


Figure 10. The experimental dependence $E_{a,cond}(T)$ for the investigated samples.

From Figure 10, it can be seen that $E_{a,cond}$ increases linearly with temperature, for both samples, from 0.101 eV to 0.107 eV for sample S1 (SG-Cu) and from 0.067 eV to 0.071 eV for sample S2 (SG-Al). The mechanism of the electrical conductivity in the investigated samples at low frequencies can be explained by the hopping process of the charge carriers between the localized states [42] over the temperature range between 30 °C and approximately 54–56 °C, based on the Mott VRH model. The obtained values are similar to the values obtained in Ref. [22] for the activation energy of the conduction process for NaTaO₃ samples doped with Fe or Ag ions.

As we showed in Ref. [43], taking into account Mott's VRH model and the values obtained for the activation energy of the conduction process, $E_{a,cond}(T)$, we established Equation (7), which allows the determination of the density of localized states at the Fermi level, $N(E_F)$:

$$N(E_F) = \frac{\lambda(akT)^3}{(4E_{a,cond})^4} \quad (7)$$

where $\lambda \approx 16.6$, is a non-dimensional constant and $a \approx 10^9 \text{ m}^{-1}$ is the degree of localization [34]. Using Equation (7) and the values of $E_{a,cond}(T)$, from Figure 10, we have computed $N(E_F)$ at temperatures below 50–60 °C for both samples. The following values were obtained for the first time for NaTaO₃ ceramic samples doped with Cu or Al metal ions: $N(E_F) = 4.45 \times 10^{19} \text{ cm}^{-3} \cdot \text{eV}^{-1}$ for sample S1 (SG-Cu) and $N(E_F) = 23.12 \times 10^{19} \text{ cm}^{-3} \cdot \text{eV}^{-1}$ for sample S2 (SG-Al), and these do not depend on temperature for the investigated temperature range. Similar results, where $N(E_F)$ remains constant throughout the investigated temperature range, were also obtained for other materials, such as: vivianite [40] or crednerite [44] materials.

From the values obtained for the density of localized states at the Fermi level of the two samples, it is observed that $N(E_F)$ for sample S2 (SG-Al) is higher than $N(E_F)$ for sample S1 (SG-Cu). The result is in agreement with the decrease of DC conductivity, σ_{dc} of sample S2 compared to σ_{dc} of sample S1 (see Figure 8), i.e., the decrease of the activation energy for electrical conduction, $E_{a,cond}$ (see Figure 10) leads to an increase of the density of states at the Fermi level $N(E_F)$. We consider that this increase in $N(E_F)$ could be correlated with a decrease in the hopping distance between the localized states of the charge carriers. To justify this statement, we determined two other Mott's parameters of the VRH model: the hopping distance R and the hopping energy W , with these equations [34]:

$$R = \left(\frac{9}{8akTN(E_F)} \right)^{1/4} \quad (8)$$

$$W = \frac{3}{4\pi R^3 N(E_F)} \quad (9)$$

Using the $N(E_F)$ values of samples S1 and S2, and the Equations (8) and (9), we determined the hopping distance, R , and the hopping energy, W , at different temperatures for the investigated samples. The values obtained are indicated in Table 2.

Table 2. The Mott parameters of samples at different temperatures.

T [°C]	Sample S1 (SG-Cu)		Sample S2 (SG-Al)	
	R[nm]	W[eV]	R[nm]	W[eV]
30	2.86	0.366	1.90	0.242
40	2.84	0.375	1.88	0.248
50	2.82	0.384	1.87	0.254
54	2.81	0.388	1.86	0.256

As can be seen from Table 2, for both samples, the hopping distance, R , decreases with increasing temperature and the hopping energy, W , increases with increasing temperature. Also, the hopping distance, R , and the hopping energy, W , corresponding to sample S2 (SG-

Al) are smaller than R and W corresponding to sample S1 (SG-Cu) for the same temperature, T. This result confirms our previous statement regarding the increase in density states at the Fermi level $N(E_F)$ of sample S2 (SG-Al) compared to $N(E_F)$ of sample S1 (SG-Cu), due to the decrease of the hopping distance R(S2) compared to R(S1).

In the dispersion region corresponding to high frequencies (Figure 7a,b), the dynamic conductivity, $\sigma_{ac}(\omega, T)$, respects the equation [39]:

$$\sigma_{ac}(\omega, T) = A_0(T)\omega^{n(T)} \quad (10)$$

where n is a temperature-dependent exponent ($0 < n < 1$) and A_0 is a pre-exponential factor that depends on temperature [39]. By taking the logarithm of Equation (10), a linear dependence between $\ln\sigma_{ac}$ and $\ln\omega$ is obtained. Figure 11a,b shows the experimental dependence of $\ln(\sigma_{ac})$ on $\ln(\omega)$ for samples S1 (SG-Cu) and S2 (SG-Al) at each temperature T.

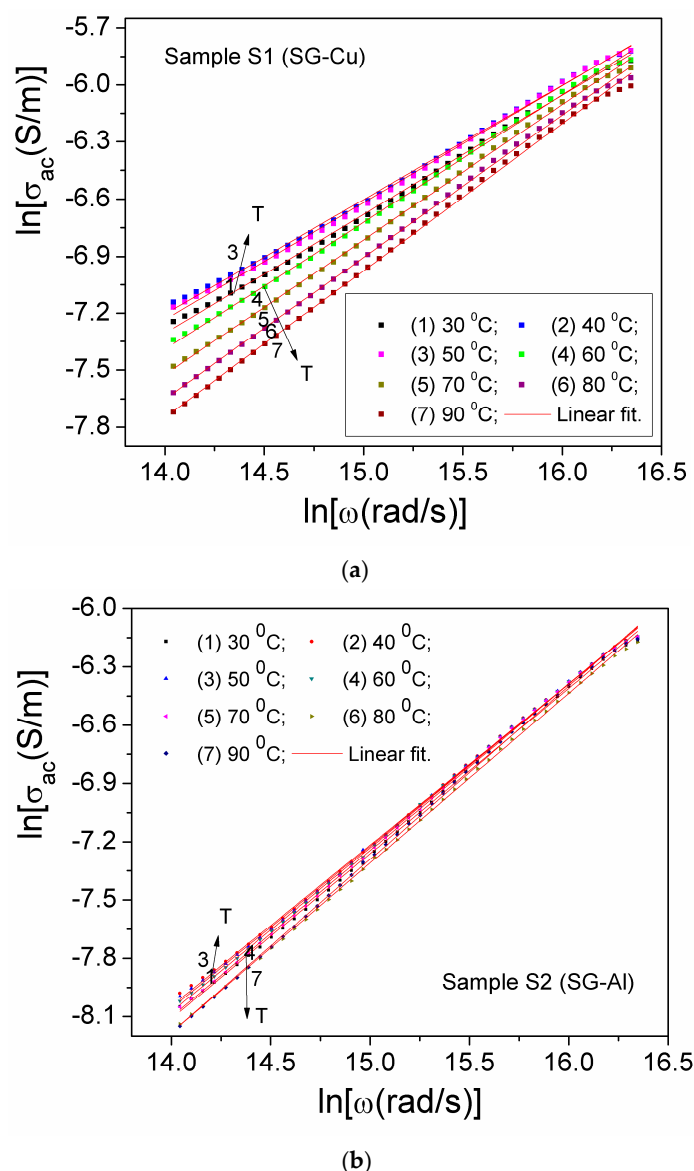


Figure 11. The $\ln\sigma_{ac}(\ln\omega)$ dependence at different values of temperature, T: (a) for sample S1(SG-Cu) and (b) for sample S2 (SG-Al).

By fitting the experimental dependence $\ln(\sigma_{ac})(\ln(\omega))$, from Figure 11 with a straight line, the exponent n and the parameter A_0 , corresponding to each sample at every temperature, T, can be determined and the obtained values are listed in Table 3.

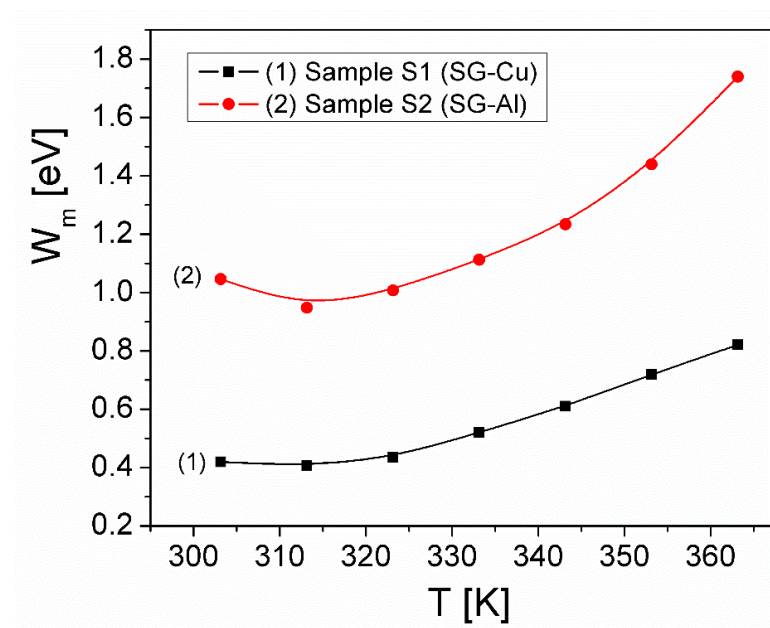
Table 3. Electrical parameters of samples, determined from AC-conductivity measurements.

T [°C]	<i>n</i>	Sample S1 (SG-Cu)		<i>n</i>	Sample S2 (SG-Al)	
		<i>A</i> ₀ [S/ms ^{<i>n</i>}]	ω_h [s ^{−1}]		<i>A</i> ₀ [S/ms ^{<i>n</i>}]	ω_h [s ^{−1}]
30	0.626	10.54×10^{-8}	3.31×10^5	0.850	2.03×10^{-9}	3.15×10^5
40	0.601	16.42×10^{-8}	4.12×10^5	0.829	2.91×10^{-9}	3.57×10^5
50	0.615	13.21×10^{-8}	4.28×10^5	0.834	2.68×10^{-9}	3.49×10^5
60	0.669	5.26×10^{-8}	3.02×10^5	0.845	2.27×10^{-9}	2.82×10^5
70	0.709	2.62×10^{-8}	2.13×10^5	0.856	1.89×10^{-9}	2.37×10^5
80	0.746	1.39×10^{-8}	1.12×10^5	0.873	1.36×10^{-9}	1.49×10^5
90	0.771	0.80×10^{-8}	1.11×10^5	0.892	1.06×10^{-9}	1.48×10^5

From Table 3, it can be observed that in the case of sample S2 of NaTaO₃ doped with Al ions, the exponent *n* has relatively high values (over 0.7), at all temperatures *T* over the investigated range, while in the case of sample S1 of NaTaO₃ doped with Cu ions, high values for the exponent *n* (over 0.7) are obtained only when the temperature increases above 70 °C. The relatively high values obtained for the exponent *n* (over 0.7), show that in the investigated samples the phenomenon of electrical conduction at high frequencies (*f* > 100 kHz) is due to a hopping process of the charge carriers from samples between the nearest neighboring states, according to the CBH (correlated barrier hopping) model [35,45], which in the case of sample S1 of NaTaO₃ doped with Cu ions is present only at high temperatures, above 70 °C. At the same time, this result can also be correlated with the fact that the activation energy for conduction *E*_{*a,cond*} obtained for sample S1 (SG-Cu) is higher than the activation energy for conduction *E*_{*a,cond*} obtained for sample S2 (SG-Al), as shown in Figure 10. The exponent *n* in a first approximation, based on CBH model can be written as:

$$n = 1 - \frac{6kT}{W_m} \quad (11)$$

In Equation (11) *W_m* represents the maximum energy barrier height (or band gap energy) [45,46] and can be calculated using Equation (11) and the *n* values of the two samples (Table 3) corresponding to each temperature *T*. The temperature dependence of the maximum energy barrier height, *W_m*(*T*), is shown in Figure 12 for the two samples.

**Figure 12.** The temperature dependence of the maximum energy barrier height, *W_m*(*T*), of the S1 (SG-Cu) and S2 (SG-Al) samples.

From Figure 12 it can be seen that the addition of metal ions of Al or Cu in the structure of the NaTaO₃ perovskite compound leads to a decrease in the energy of the band gap, up to temperatures of 40–50 °C, after which W_m starts to increase with the increase in temperature, up to 90 °C, tending to values of 0.75 eV for sample S1 (SG-Cu) and 1.74 eV for sample S2, approaching the bandgap energy value of the un-doped NaTaO₃ material, which has values between 1 and 3 eV [17,47]. According to Jonscher's theoretical model [39], the total conductivity given by Equation (4), can also be written in the following form:

$$\sigma(\omega, T) = \sigma_{dc} \left[1 + \left(\frac{\omega}{\omega_h} \right)^n \right] \quad (12)$$

Based on Equations (10) and (12), the expression of ω_h results immediately. It represents the hopping frequency [37,39], i.e., the frequency of transition from the DC regime to the dispersive regime AC, of the electrical conductivity.

$$\omega_h = \left(\frac{\sigma_{dc}}{A_0} \right)^{1/n} \quad (13)$$

The obtained values for ω_h are shown in Table 3. In Figure 7a,b, the hopping frequencies ($f_h = \omega_h/2\pi$), are indicated at which the transition from the DC regime to the AC regime occurs. The results show that by increasing the temperature from 30 °C to 50 °C, f_h , increases slightly for both samples (see Figure 7), after which it decreases until at 18 kHz (sample S1) and 23 kHz (sample S2), when the temperature increases up to 90 °C. It is known that the mechanisms of electrical conduction in materials can be investigated based on the electrical modulus, $M = M' - iM''$, where the real (M') and imaginary (M'') components can be determined with the relations [48]:

$$M' = \left(\frac{\omega \varepsilon_0 A}{d} \right) Z'' \quad (14)$$

$$M'' = \left(\frac{\omega \varepsilon_0 A}{d} \right) Z' \quad (15)$$

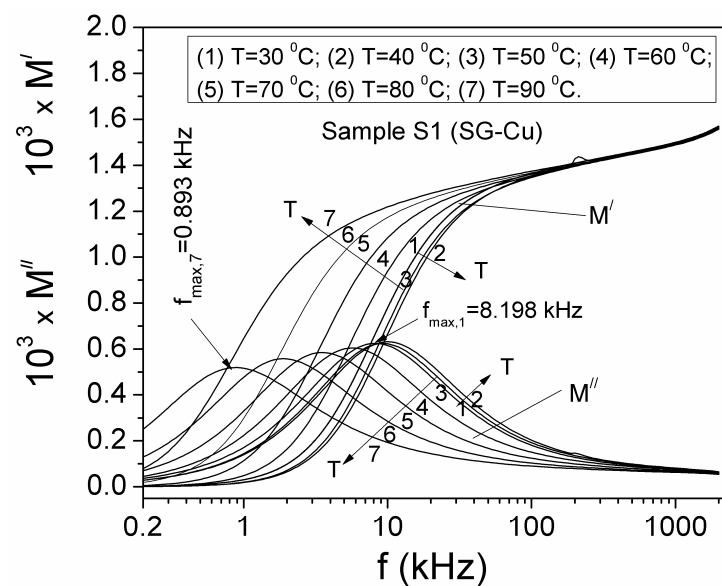
In these relationships, ε_0 is the free space dielectric permittivity. Using Equations (14) and (15) and the Z' and Z'' values from Figures 4 and 5, we determined the components M' and M'' of the electrical modulus for the two samples. Figure 13 shows the frequency dependence of the M' and M'' components at all investigated temperatures, T , for both sample S1 (SG-Cu) (Figure 13a) and sample S2 (SG-Al) (Figure 13b).

From Figure 13 it can be seen that for each temperature, by increasing the electric field frequency, the M' component increases continuously and tends to a constant value of approximately 1.5 (for sample S1) and 1.8 (for sample S2). At the same time, for a constant frequency, the value of M' decreases slightly with the increase in temperature up to approximately 50 °C, after which it starts to increase when the temperature increases from 50 °C to 90 °C. This behavior can be attributed to the temperature-dependent relaxation process in the investigated samples [49]. The imaginary component M'' presents a maximum for each temperature, corresponding to a frequency, $f_{max,(M)}$ (see Figure 13a,b). Using the values $f_{max,(M)}$ and Debye equation, we have computed the relaxation times $\tau_{M'}$, corresponding of each temperature. The obtained values are listed in Table 4.

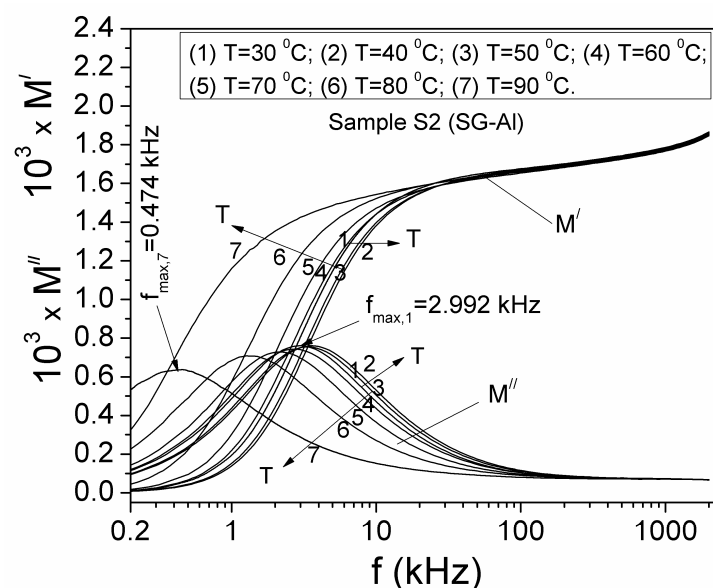
It is known [48,50] that in the case of ideal samples placed in a capacitive cell connected to the terminals of an RLC-meter of variable frequency, the equivalent electrical circuit of the cell is a parallel circuit formed by the capacity C and the resistance R , of the sample material between the capacitor plates. On the other hand, to estimate the electrical properties of a material with dielectric losses, the equivalent electrical model includes the RQC circuit, associated with the grain intake: the grain resistance (R_g), the grain capacity (C_g) and the constant phase element (CPE), which is introduced due to the non-ideal capacitive behavior of the material in the measuring cell [51]. Besides the deviation from the ideal

Debye behavior [50], the presence of the constant phase element (CPE) in the equivalent circuit of the investigated material is correlated with the Jonscher characteristic of universal power law (see Equations (4) and (10)) which plays an important role in the investigation of the electrical properties of the material [20]. As shown in Refs. [51,52], the constant phase element “Q” can be calculated from Equation (16), where the parameter n is the same as that obtained from AC-conductivity measurements ($n < 1$), which are indicated in Table 3. For an ideal resistor and ideal capacitor (with a Debye-type behavior), $n = 1$ (for ideal capacitor) and $n = 0$ (for ideal resistor) [20].

$$C = (R^{1-n}Q)^{1/n} \quad (16)$$



(a)



(b)

Figure 13. The frequency dependence of the M' and M'' components of the electrical modulus of the samples: (a) S1 (SG-Cu), and (b) S2 (SG-Al) at different temperature T values.

Table 4. Electrical parameters of equivalent circuits for samples S1 (SG-Cu) and S2 (SG-Al).

Compounds	T[°C]	$\tau_M[\mu s]$	$R_g[M\Omega]$	$C_g[pF]$	$Q_g[nFs^{n-1}]$
Sample S1	30	19.42	2.20	8.81	0.510
	40	15.88	1.82	8.71	0.716
	50	17.31	1.96	8.82	0.601
	60	26.67	2.93	9.10	0.297
	70	47.43	5.04	9.42	0.171
	80	79.61	8.06	9.87	0.109
	90	178.25	16.83	10.59	0.076
Sample S2	30	53.21	6.74	6.75	0.030
	40	44.77	5.67	5.67	0.033
	50	47.42	5.97	5.97	0.033
	60	59.70	7.39	7.39	0.034
	70	70.95	9.12	9.12	0.035
	80	119.13	15.91	15.91	0.046
	90	335.80	35.81	35.80	0.073

At the same time, the relaxation time τ_M can be defined in terms of resistance (R) and capacity (C) with the relation, $\tau_M = RC$, so that the Debye equation is written in the form, $2\pi f_{max}RC = 1$. The estimation of the R and C values of the equivalent electric circuit can be accomplished both by graphically representing the temperature dependence $M''(f)$ for two values of the temperature T (the minimum value $T_1 = 303$ K and the maximum, $T_7 = 363$ K), and by the equivalent parallel (RQC) circuit (Figure 14).

The elements R and C of the parallel circuit influence the shape of the $M''(f)$ peaks (Figure 13) and the amplitude of the peak in the $M''(f)$ curves is proportional to the capacity C, i.e., $M''_{max} = C_0/2C$ where C_0 is cell capacity without sample ($C_0 = 0.0115$ pF). Knowing the M''_{max} (Figure 13) and τ_M values (Table 4), both the elements C_g and R_g of the equivalent parallel circuit, corresponding to the samples, at each temperature, T, were determined. The obtained values are shown in Table 4. The constant phase element “Q” can be computed from Equation (16) taking into account the values of R_g , C_g (Table 4) and n (Table 3) corresponding to samples S1 (SG-Cu) and S2 (SG-Al). The values obtained for the phase element constant “ Q_g ” are indicated in Table 4.

As seen from Table 4, by increasing the temperature T from 30 °C to 90 °C, the grain capacity C_g of the equivalent electric circuit of the two samples increases from 8.8 pF to 10.6 pF (for sample S1 (SG-Cu)), respectively from 7.8 pF at 9.4 pF (for sample S2 (SG-Al)). This behavior is observed in most ferrites [33,53,54]. By increasing the temperature, the thermal activation of the hopping of charge carriers in the octahedral sites takes place, this process is responsible for the electrical conduction in the samples [33]. As a result, the dielectric polarization increases, and therefore, the dielectric permittivity, which causes an increase in the capacity, C by increasing the temperature. Also, from Table 4, it is observed that by increasing the temperature from 30 °C to 90 °C, the grain resistance R_g of the equivalent electrical circuit increases from 2.20 M Ω to 16.8 M Ω (for sample S1 (SG-Cu)), respectively from 6.7 M Ω at 35 M Ω (for sample S2 (SG-Al)), this behavior is observed by other authors as well [55,56]. The increase in the resistance R_g of the equivalent electrical circuit of samples can be attributed to the increase in the mobility of charge carriers and their hopping rate with temperature [55]. At the same time, such materials that show a high resistance to temperature increase is characterized by low dielectric losses [55,56].

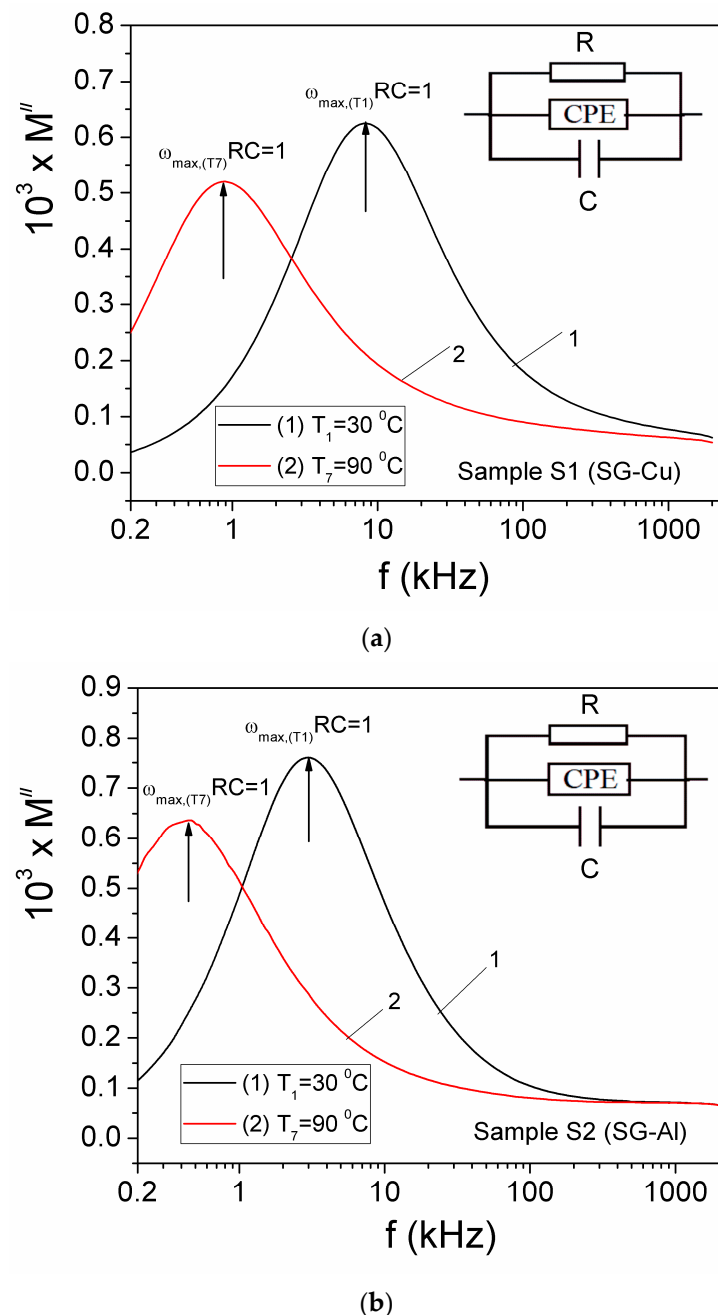


Figure 14. The frequency dependence of M'' component of samples at two values T of temperature. The inset shows the equivalent electric circuit as an example. (a) sample S1 (SG-Cu) and (b) sample S2 (SG-Al).

4. Conclusions

Two NaTaO_3 ceramic samples in powder form, doped with Cu metal ions (sample S1) or with Al ions (sample S2), were prepared by the sol-gel method. X-ray diffraction analysis shows that both samples S1 (SG-Cu) and S2 (SG-Al) are well crystallized, without secondary phases and crystallize in the space group Pcmn (space group number 62), which belongs to the orthorhombic crystallographic family. The UV-VIS measurements show that the presence of Cu or Al ions in the NaTaO_3 structure leads to a decrease in the optical band-gap energy (E_g) of the samples, obtaining values of 3.80 eV (for sample S1 (SG-Cu)) and 3.85 eV (for sample S2 (SG-Al)), which are lower than the value reported for a sample of NaTaO_3 (4 eV). Complex impedance measurements, in the frequency range 200 Hz–2 MHz, and at different temperatures between 30 and 90 °C, in both samples,

showed the presence of a maximum of the imaginary component Z'' of the complex impedance at each temperature T , thus, indicating the existence of an electrical relaxation process due to the hopping of charge carriers between localized states, according to Mott's VRH model. These measurements allowed, for the first time, the determination of the activation energy, E_{activ} , which represents the barrier energy between the localized states, associated with two zones of different conductivity. The following values were obtained for the two samples: 0.046 eV (below 52 °C) and 0.602 eV (above 52 °C) for sample S1 (SG-Cu) and 0.020 eV (below 54 °C) and 0.461 eV (above 54 °C) for sample S2 (SG-Al), respectively. Based on the obtained results, we highlighted for the first time a transition temperature from a semiconductor-type behavior to a conductor-type behavior, for the two samples: 52 °C (for sample S1 (SG-Cu)), respectively 54 °C (for sample S2 (SG-Al)), the temperature at which the change in the barrier energy between the localized states, E_{activ} , of the samples also occurs. Using the complex impedance measurements, the spectrum of the electrical conductivity $\sigma(f,T)$ of samples, was determined, and the results show that at all investigated temperatures, the conductivity σ for the samples follows Jonscher's universal law. The temperature dependence of the static conductivity $\sigma_{dc}(T)$ was determined and the results show that the slope of the static conductivity changes at the temperature of 54 °C (for sample S1) and 56 °C (for sample S2), another highlighting of the fact that the samples change their behavior from a semiconductor-type to a conductor-type. Using the Mott's VRH model, the density of localized states at the Fermi level, $N(E_F)$, was determined for the first time on NaTaO₃ ceramic samples doped with Cu or Al metal ions, obtaining the following values: $N(E_F) = 4.45 \times 10^{19} \text{ cm}^{-3} \cdot \text{eV}^{-1}$ (for sample S1 (SG-Cu)) and $N(E_F) = 23.12 \times 10^{19} \text{ cm}^{-3} \cdot \text{eV}^{-1}$ (for sample S2 (SG-Al)). These values do not depend on temperature for the investigated temperature range. The increase in the density of states at the Fermi level $N(E_F)$ of sample S2 (SG-Al) compared to $N(E_F)$ of sample S1 was explained by the decrease in the hopping distance, R , of charge carriers in sample S2 compared to that of sample S1. In the high frequencies region (over to 200 kHz), the band gap energy (W_m) of the both samples and the transition frequency ω_h , from the DC regime to AC regime, at all investigated temperatures were determined, for the first time, based on the VRH and CBH models, for samples S1 (SG-Cu) and S2 (SG-Al). Taking into account the Jonscher characteristic of universal power law, an original method to evaluate the elements of the equivalent electrical circuit of the investigated samples, at different temperatures, was proposed: the grain resistance (R_g), the grain capacity (C_g) and the constant phase element (CPE). Knowing the electrical properties of NaTaO₃ ceramic materials doped with Cu or Al metal ions at different frequencies and temperatures is useful for clarifying the electrical conduction mechanisms in these materials, as well as for their use in electronic applications, thermoelectric energy conversion or applications photocatalytic.

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