

Article

Semiconductor–Conductor Transition Analysis by Low-Frequency Impedance in Ultrasonically Synthesized Al-Doped Sodium Tantalate

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Abstract

An aluminum-doped NaTaO₃ perovskite sample was prepared by the ultrasonic method, employing an immersed sonotrode, followed by thermal treatment at 600 °C for 6 h in air. X-ray diffraction analysis reveals a biphasic system with relatively low crystallinity, consisting of a dominant NaTaO₃ perovskite phase and a secondary Na₂Ta₄O₁₁ phase. Optical investigations indicate a reduced band gap energy of 3.77 eV compared to undoped NaTaO₃ (4 eV), suggesting enhanced absorption toward the infrared region and improved photocatalytic potential. Fourier Transform Infrared FTIR Spectroscopy highlights the emergence of a distinct absorption band at 670 cm^{−1}, attributed to Ta–O and Al–O stretching vibrations, evidencing successful incorporation of Al dopants. Complex impedance analysis over the frequency and temperature ranges of (20 Hz–2 MHz) and (29–100) °C identifies, for the first time, the semiconductor–conductor transition temperature at 58 °C. Nyquist analysis further supports the coexistence of grain and grain boundary contributions, modeled via equivalent R and CPE parallel circuits. Conductivity studies confirm obedience to Jonscher’s universal law, with a change in σ_{DC} slope near 54 °C, corroborating semiconductor–conductor transition behavior. Dielectric measurements similarly indicate a relaxation process linked to interfacial polarization, with a transition temperature of (~54 °C). Overall, the ultrasonic synthesis route uniquely enables a biphasic structure that facilitates the observation of a low-temperature semiconductor-to-conductor transition, absent in analogous single-phase materials obtained via sol–gel methods.

Keywords: sodium tantalate; complex impedance; DC and AC conductivity; complex dielectric permittivity; VRH theoretical model



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1. Introduction

The group of compounds with the generic formula ABO₃, where A and B are cations and oxygen O is an anion, is known as perovskite oxides. They have been extensively studied in the recent years due to their simple crystalline structure and various properties (electrical, magnetic, optical, etc.) [1,2], as well as their potential applications in different areas, such as: catalytic and photovoltaics [3,4], solar cells [5,6], electronic and memory devices [7,8] supercapacitors and sensors [9] and energy conversion [10]. The ability to

obtain perovskite materials with nanometer-scale particle sizes has recently encouraged researchers to identify new ceramic materials that could serve as energy sources, such as thermoelectric generators, and could represent an alternative to lead-based perovskite systems with thermoelectric properties [11,12]. In this regard, studies have shown that the most promising compounds in the category of functional ceramic materials [13] are based on niobium and tantalum oxides. In the category of tantalates, sodium tantalate (NaTaO_3) represents a feasible alternative to lead-based compounds, with it being considered a perovskite material with thermoelectric potential [14]. In addition to these properties, NaTaO_3 also exhibits photocatalytic properties [15]. Moreover, in Refs. [16,17], the authors show that by doping metal cations or anions onto the NaTaO_3 structure, the visible light response of the NaTaO_3 photocatalyst is improved due to the reduction in the optical band gap below 4 eV [15,18]. The theoretical study of the electronic and structural characteristics of the NaTaO_3 compound shows the presence of a structural phase transition with a decrease in temperature [19,20]. At the same time, doping with metal ions (Fe, Mn, Co, Cu, Ag) leads to modification of the electronic structure of the NaTaO_3 compound [21,22], which modifies its optical and electrical properties. In other references, the authors investigate the dielectric relaxation and conduction mechanism in calcium copper titanate codoped with Sr and Zn [23] and in strontium gadolinium niobate perovskite oxide [24]. On the other hand, in Ref. [25], the authors analyzed the electrical conductivity and electrical modulus based on impedance spectroscopy in the range (4 Hz–8 MHz). In our paper [26], the electrical band gap energy of the two samples was determined using electrical resistivity measurements at temperatures over the range (30–150) °C for undoped and Ag-doped NaTaO_3 materials synthesized by the sol–gel method.

The structural, electrical, optical, and magnetic properties of perovskite materials, as well as their possible applications, depend greatly on the synthesis process and doping of the perovskite material. Several methods are known for the synthesis of perovskite oxides, such as the sol–gel method [27], combustion synthesis [28], hydrothermal and solvothermal synthesis [29], microwave-assisted hydrothermal synthesis [30] and the co-precipitation method [31].

Recently, our group studied the AC conductivity of NaTaO_3 ceramic materials doped with Cu and Al metal ions and prepared by the sol–gel method [32]. Unlike this study, in that paper, we prepared a sample of sodium tantalate (NaTaO_3) doped with Al metal ions using the ultrasonic method with the sonotrode immersed in the reaction medium, followed by heat treatment at 600 °C, which was developed for the first time by our group [22]. The aim was to investigate comparatively the effect of the synthesis and Al doping method of the NaTaO_3 compound on the electrical conductivity (σ) and complex dielectric permittivity (ϵ) of the sample, based on complex impedance measurements in the range of 20 Hz–2 MHz and at different temperatures between (29 and 100) °C. The electrical conductivity was investigated based on the VRH and CBH models, and the dielectric behavior of the sample was explained in terms of Maxwell–Wagner interfacial relaxation. Nyquist plots ($-Z''$ versus Z') of the complex impedance were constructed for each temperature, and the results were analyzed by fitting the experimental data to an equivalent circuit given by the combination of two parallel circuits, $R//CPE$. For the first time, the transition temperature from semiconductor-like to conductor-like behavior of the sample was identified based on the temperature dependence of the time relaxation, DC conductivity, and dielectric constant of the sample. At the same time, the use of the ultrasonic method in the preparation of the aluminum-doped NaTaO_3 sample uniquely resulted in a biphasic system, highlighting an electrical transition at low temperature, unlike the single-phase counterpart derived from the sol–gel method [32].

2. Materials and Methods

2.1. Synthesis of Sample

For the synthesis of an Al-doped sodium tantalate (NaTaO_3) ceramic material, the following precursors were used: tantalum ethoxide ($\text{Ta}(\text{OC}_2\text{H}_5)_5$), sodium hydroxide (NaOH), and aluminum nitrate ($\text{Al}(\text{NO}_3)_3$), all of analytical grade. The synthesis procedure was as follows: First, 1 mL of tantalum ethoxide was added to 50 mL of bidistilled water under continuous magnetic stirring. Then, 0.0375 g of aluminum nitrate was introduced, and the precipitation of the material was carried out by adding aqueous sodium hydroxide solution (2 M NaOH) until the pH reached 13. The resulting solution was subjected to ultrasonic treatment for 15 min using a Sonics Vibra-Cell sonotrode immersed in the reaction medium, with the following operating parameters: 80% amplitude and pulse mode 10 s ON/5 s OFF. The obtained precipitate was filtered and washed with bidistilled water until neutral pH, then heat-treated at 600 °C for 6 h. Using this ultrasonic-assisted method, first developed by our group [22], an Al-doped NaTaO_3 powder was obtained (denoted as sample US-Al).

2.2. Characterization

The X-ray diffraction pattern of the prepared US-Al sample was recorded using an Advance Bruker-AXS D8 diffractometer (Bruker Optics Inc, Billerica, MA, USA) with $\text{CuK}\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$, and Zr filter on the diffracted beam, 40 kV and 40 mA), set to operate in constant scan mode in the range $10^\circ \leq 2\theta \leq 85^\circ$. The width and position of the diffraction peaks were determined using the evaluation software in the DIFFRACPLUS 12.0, software Version 16.0 package. A scanning electron microscope (SEM), model FEI Inspect S, from FEI Company PANalytical (Almelo, Netherlands), was used for the morphological analysis of the sample. This microscope can also perform elemental analysis and generate elemental maps of a sample using its energy-dispersive X-ray (EDX) analysis capability. Diffuse reflection spectra were recorded by ultraviolet–visible light diffusion spectrometry (UV–VIS) to estimate the band gap energy of the obtained material, using a UV-VIS Spectrophotometer type DRUV-VIS Lambda 950 Perkin-Elmer (Waltham, MA, USA), which operates in the wavelength range of (200–2500) nm. Using a Shimadzu Prestige-21 spectrometer, Kyoto, Japan, in the range of 400–4000 cm^{-1} , the Fourier Transform Infrared Spectroscopy (FTIR) spectrum was obtained for the US-Al sample. The identification of the absorption bands was carried out using existing data from the literature [33]. Based on the complex impedance measurements in the frequency range 20 Hz–2 MHz and at different temperatures between (30 and 100) °C, the electrical resistivity/conductivity was determined. The measurements were performed using an LCR meter Agilent E-4980-A type (Keysight Technologies, Santa Clara, CA, USA), in conjunction with a laboratory experimental setup [34], like ASTM D150-98 [35].

3. Results and Discussion

3.1. Structural and Morphological Characterization

3.1.1. XRD Analysis

The X-ray diffraction pattern of the sodium tantalate sample doped with Al metal ions (sample US-Al) is shown in Figure 1a.

As can be seen in Figure 1a, the XRD pattern of the NaTaO_3 sample doped with Al ions, obtained by the ultrasonic method (US-Al), exhibits relatively low crystallinity, with two phases present: the main perovskite phase NaTaO_3 and a secondary phase identified as $\text{Na}_2\text{Ta}_4\text{O}_{11}$ (natrotantite), according to JCPDS card no. 38-0463. From the X-ray diffraction data, the average crystallite size was calculated using the Scherrer equation based on the broadening of diffraction peaks, specifically the full width at half maximum

(FWHM, β), after correcting for the instrumental contribution and subtracting the $K\alpha_2$ component. For the investigated sample (US-Al), crystallite size estimation was performed on several diffraction maxima exhibiting sufficient broadening to allow accurate FWHM determination, namely, the (006), (012), (104), (119), and (115) reflections. Crystallite size analysis revealed that the $\text{Na}_2\text{Ta}_4\text{O}_{11}$ phase has an average size of 26.6 nm, while the NaTaO_3 phase has a noticeably larger size of 39.1 nm, suggesting distinct nucleation and growth mechanisms for the two crystalline phases.

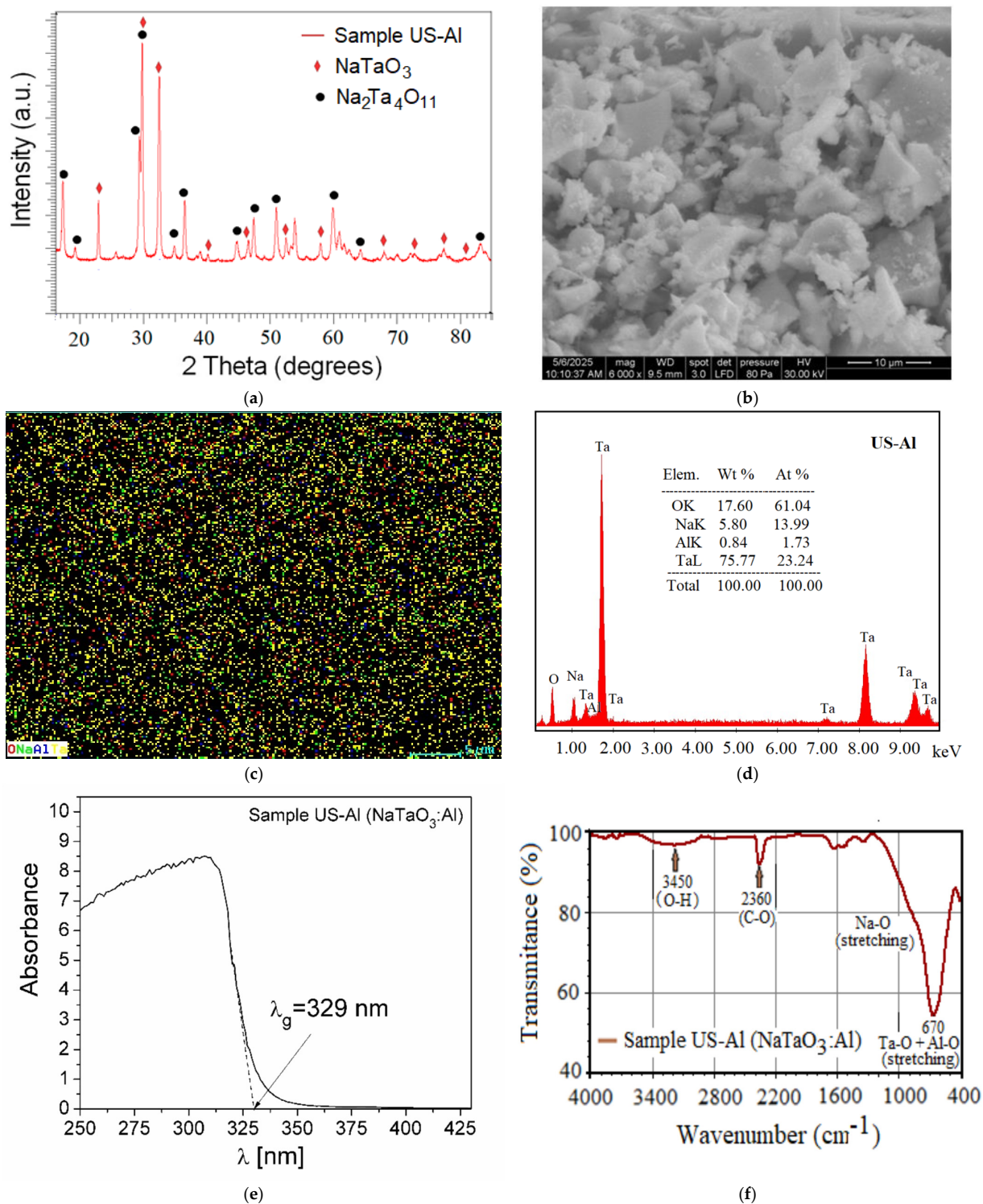


Figure 1. The XRD pattern for the US-Al sample (a); an SEM image (b); element mapping (c); EDX spectrum (d); the diffuse reflection spectrum (e) and FT-IR spectrum for the US-Al sample (f).

3.1.2. Scanning Electron Microscopy

In Figure 1b–d, the SEM images, the EDX spectrum, element quantification, and mapping of the Al-doped NaTaO₃ ceramic material synthesized by the ultrasonic method are presented. The SEM micrograph of the Al-doped NaTaO₃ sample (Figure 1b) reveals a highly agglomerated microstructure composed of irregular, plate-like particles with rough surfaces and sharp fracture edges. The grain size distribution appears relatively broad, ranging from submicron dimensions to several micrometers, as estimated from the scale bar. In Figure 1c, the elemental mapping analysis demonstrates the presence and uniform spatial distribution of Na, Ta, O, and Al across the analyzed area. No evidence of elemental segregation or Al-rich clusters is observed at the micrometer scale. The homogeneous distribution of aluminum suggests its successful incorporation into the NaTaO₃ matrix rather than the formation of secondary phases. This compositional uniformity is consistent with the absence of distinct morphological contrasts observed in the corresponding SEM image. The EDX spectrum (Figure 1d) further verifies the elemental composition of the sample, displaying characteristic peaks corresponding to Na, Ta, O, and Al. Quantitative analysis indicates that the measured elemental ratios are close to the nominal composition within the experimental uncertainty inherent to EDX measurements. The detection of aluminum, together with its uniform distribution revealed by elemental mapping, confirms the effective doping of the NaTaO₃ perovskite structure.

3.1.3. UV-VIS Spectroscopic Analysis

Figure 1e shows the UV-VIS absorption spectrum of the Al-doped sodium tantalate (NaTaO₃) sample. From Figure 1e, the wavelength end of the absorption range was determined, λ_g , corresponding to sample NaTaO₃ doped with aluminum ions, as the intersection point between the tangent to the linear portion of the absorption spectrum and the wavelength axis; the obtained value is $\lambda_g = 329$ nm. The optical bandgap energy (E_g) of the sample can be computed [36,37] with the relation:

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

Considering Equation (1) and the λ_g value determined from Figure 1e, the value obtained for the optical bandgap energy is $E_g = 3.77$ eV for sample NaTaO₃ doped with aluminum ions. The obtained result shows that by doping the sodium tantalate ceramic material (NaTaO₃) with Al metal ions, the value of the optical band gap energy decreases to 3.77 eV from 4 eV [18] (for undoped NaTaO₃). This decrease in E_g energy compared to that of undoped NaTaO₃ indicates an increase in the mobility of the carriers and also an improvement in the absorption in the infrared domain, making the NaTaO₃ material doped with Al metal cations useful for photocatalysis, as also shown by other authors [38,39].

3.1.4. FT-IR Analysis

Figure 1f shows the FTIR spectrum of the Al-doped sodium tantalate (NaTaO₃) sample (sample US-Al). As shown in [40,41], the FTIR absorption spectrum of a NaTaO₃ sample exhibits a main absorption band centered around $(1100\text{--}1200)$ cm^{−1}, attributed to stretching vibrations of the Na-O functional group, as well as three distinct (weaker) absorption bands centered between $(500$ and $700)$ cm^{−1}, indicating vibrational excitation. The presence of Al dopant ions in the structure of NaTaO₃ prepared by the ultrasonic method (US-Al, Figure 1f) causes a significant decrease, up to disappearance, in the share of the main absorption band (Na-O), accompanied by the appearance of a strong absorption band at 670 cm^{−1}, attributed to the excitation of vibrations (stretching) of the Ta-O and Al-O groups, with a much higher share of the latter. An explanation for the appearance of this band could be a symmetric

combination of the Ta-O and Al-O stretching modes in the TaO₆ octahedra [40,41]. At the same time, from Figure 1f, it is observed that the characteristic peak corresponding to C–O stretching is present at 2360 cm^{−1}, like the value corresponding to the undoped NaTaO₃ sample [40]. By doping the NaTaO₃ sample with Al metal ions (US-Al sample, Figure 1f), it is observed that the IR band corresponding to the O–H vibrations of the water adsorbed on the surface shifts to lower values (3150 cm^{−1}) compared to the IR band corresponding to an undoped NaTaO₃ sample [40,41], which is present between (3200 and 3300) cm^{−1}.

3.2. Impedance Spectra and Equivalent Circuit

Figure 2a,b show the frequency dependence of the real Z' and imaginary Z'' components of the complex impedance, in the range 20 Hz–2 MHz, at different temperatures T , between (29 and 100) °C, as well as the Nyquist diagrams (Figure 2c) of the sample at different temperatures.

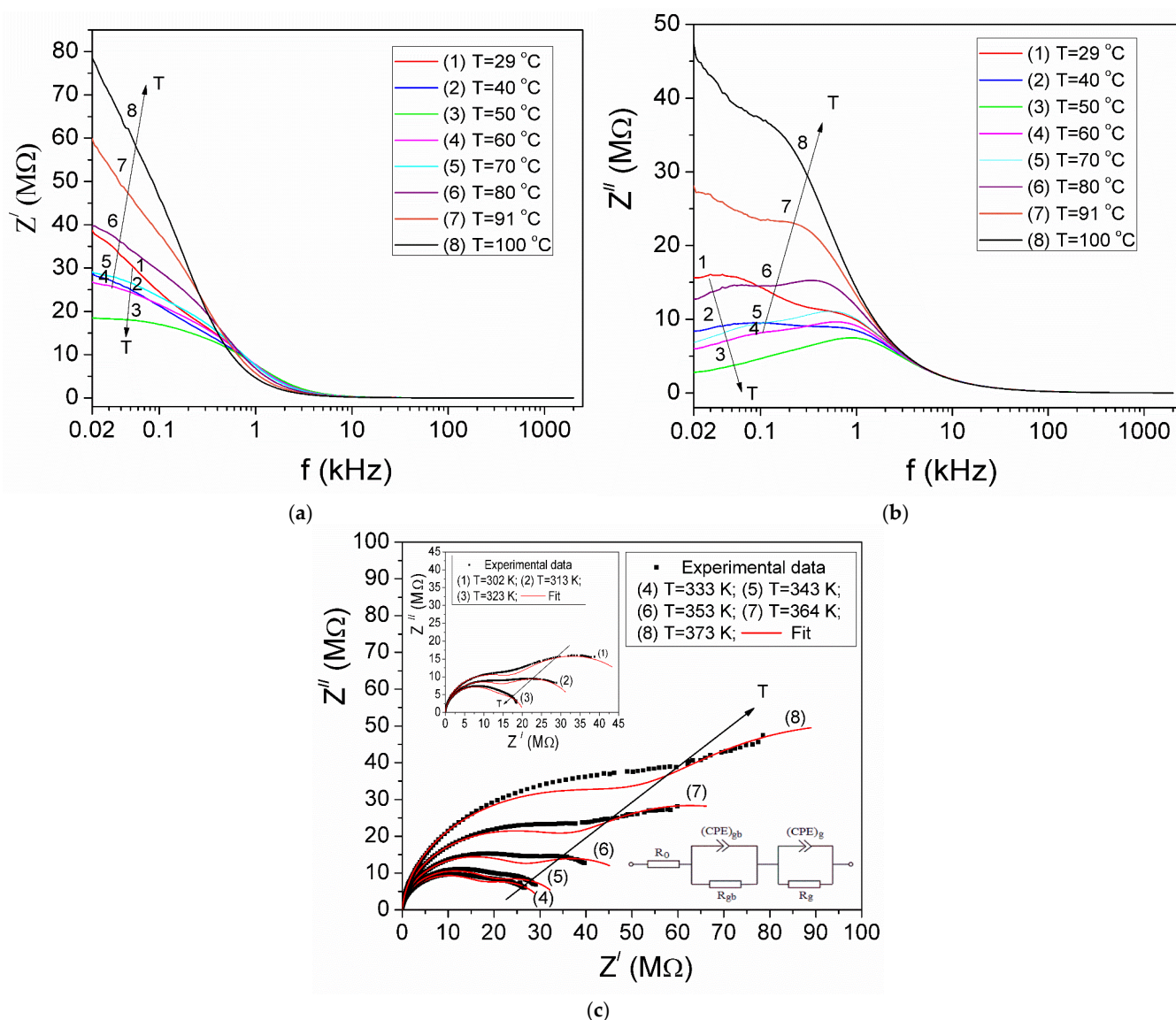


Figure 2. Frequency dependence of the Z' (a) and Z'' (b) components of the complex impedance at different temperatures of the sample US-Al. (c) Nyquist diagrams of the US-Al sample at different temperatures. The inset shows the equivalent circuit used to fit the data, where R_0 , R_g and R_{gb} are the contributions due to the electrode interface (R_0), the grain (R_g) and the grain boundary (R_{gb}), and $(CPE)_g$ and $(CPE)_{gb}$ represent the constant phase element (CPE) corresponding to the grain $(CPE)_g$ and grain boundary $(CPE)_{gb}$, respectively.

As shown in Figure 2a, the real component Z' decreases with increasing frequency (f) at each constant temperature (T). At low frequencies (up to approximately 1 kHz), the Z' values decrease as the temperature rises from 29 °C to 50 °C; however, beyond 50 °C, Z' begins to increase as the temperature reaches 100 °C. At high frequencies (above 10 kHz), the Z' curves overlap for all temperatures. Figure 2b shows that at each constant temperature, the imaginary component Z'' exhibits two maxima: one at low frequencies (on the order of tens of Hz) and the second at higher frequencies (on the order of hundreds of Hz). The peak frequencies, $f_{\max,1(Z)}$ and $f_{\max,2(Z)}$, shift toward higher values as the temperature increases from 29 °C to approximately 50 °C, while the Z'' values decrease. Conversely, as the temperature increases from 50 °C to 100 °C, these two frequencies shift back toward lower values while the Z'' values increase.

The presence of two maxima in the Al-doped NaTaO₃ sample synthesized via the ultrasonic method (US-Al) can be correlated with the two phases identified by XRD analysis: the primary perovskite phase (NaTaO₃) and a secondary phase (Na₂Ta₄O₁₁, natrotantite) (see Figure 1a). This secondary phase can generate charge traps and regions of varying conductivity; the boundary between these two phases acts as a barrier, influencing the low-frequency dielectric response through Maxwell–Wagner interfacial relaxation [42]. These results differ from those reported in a recent study of an Al-doped NaTaO₃ sample prepared by the sol–gel method, which exhibited only a single perovskite phase [32]. For that sample [32], the Z'' component presented only one maximum at frequencies near 1 kHz. Consequently, we can consider that the second maximum observed in Figure 2b at frequency $f_{\max,2(Z)}$ can be attributed to the main NaTaO₃ perovskite phase.

Figure 2c shows the Nyquist plots ($-Z''$ versus Z') of the Al-doped NaTaO₃ (US-Al) sample at various temperatures within the frequency range of 20 Hz–2 MHz. As observed in Figure 2c, all impedance spectra exhibit two semicircular arcs, indicating non-Debye relaxation [43]. This behavior may be attributed to the two phases present in the sample, as well as the contributions from grains and grain boundaries [41,44]. Additionally, the decrease in arc radii with increasing temperature indicates semiconducting behavior, which is particularly evident between 29 °C and approximately 60 °C (see inset of Figure 2c).

Furthermore, as shown in Figure 2b, the presence of the maxima of imaginary component (Z''), $f_{\max,1(Z)}$ and $f_{\max,2(Z)}$, correlates with the existence of an electrical relaxation process in the Al-doped sodium tantalate sample [45,46], due to the presence of charge carriers in the sample [44,47]. Using the experimental values for $f_{\max,1(Z)}$ and $f_{\max,2(Z)}$ in Figure 2b and Debye's theory [48], $2\pi f_{\max} \tau_{(Z)} = 1$, the relaxation times $\tau_{1(Z)}$ and $\tau_{2(Z)}$ —corresponding to the relaxation processes of each phase (see Figure 1)—can be determined for each temperature (T). Assuming an Arrhenius-type dependence for the relaxation time $\tau_{(Z)}$:

$$\tau_{(Z)} = \tau_0 \exp\left(-\frac{\Delta E_{\text{relax}}}{kT}\right) \quad (2)$$

In Figure 3a, the dependence $\ln \tau_{(Z)}(1/T)$ is shown.

In Equation (2), τ_0 represents the pre-exponential factor of the relaxation time; k is the Boltzmann constant, and ΔE_{relax} is the barrier energy between the localized states [47,49].

The linear fits of the experimental dependencies, $\ln \tau_{1(Z)}$ vs. (T^{-1}) and $\ln \tau_{2(Z)}$ vs. (T^{-1}) , shown in Figure 3a, present two different slopes, corresponding to the two temperature ranges: below and above 53 °C for τ_1 and below and above 58 °C for τ_2 . These variations in τ with temperature can be associated with two distinct electrical conductivity regimes of the sample [50]. Furthermore, the activation energies (ΔE_{relax}) for these temperature ranges were determined from the Arrhenius fits (Equation (2)). For τ_1 (secondary phase), the values obtained were $\Delta E_{\text{relax},(1)} = 0.376$ eV (below 53 °C) and $\Delta E_{\text{relax},(2)} = 0.512$ eV (above 53 °C). For τ_2 (main perovskite phase), the values were $\Delta E_{\text{relax},(1)} = 0.217$ eV (below 58 °C)

and $\Delta E_{\text{relax},(2)} = 0.583$ eV (above 58 °C). Consequently, for the Al-doped NaTaO₃ sample prepared by the ultrasonic method (US-Al), the temperature range of 50–60 °C—where the change in activation energy occurs—represents a transition from semiconductor-type to conductor-type behavior. We will justify this result in what follows.

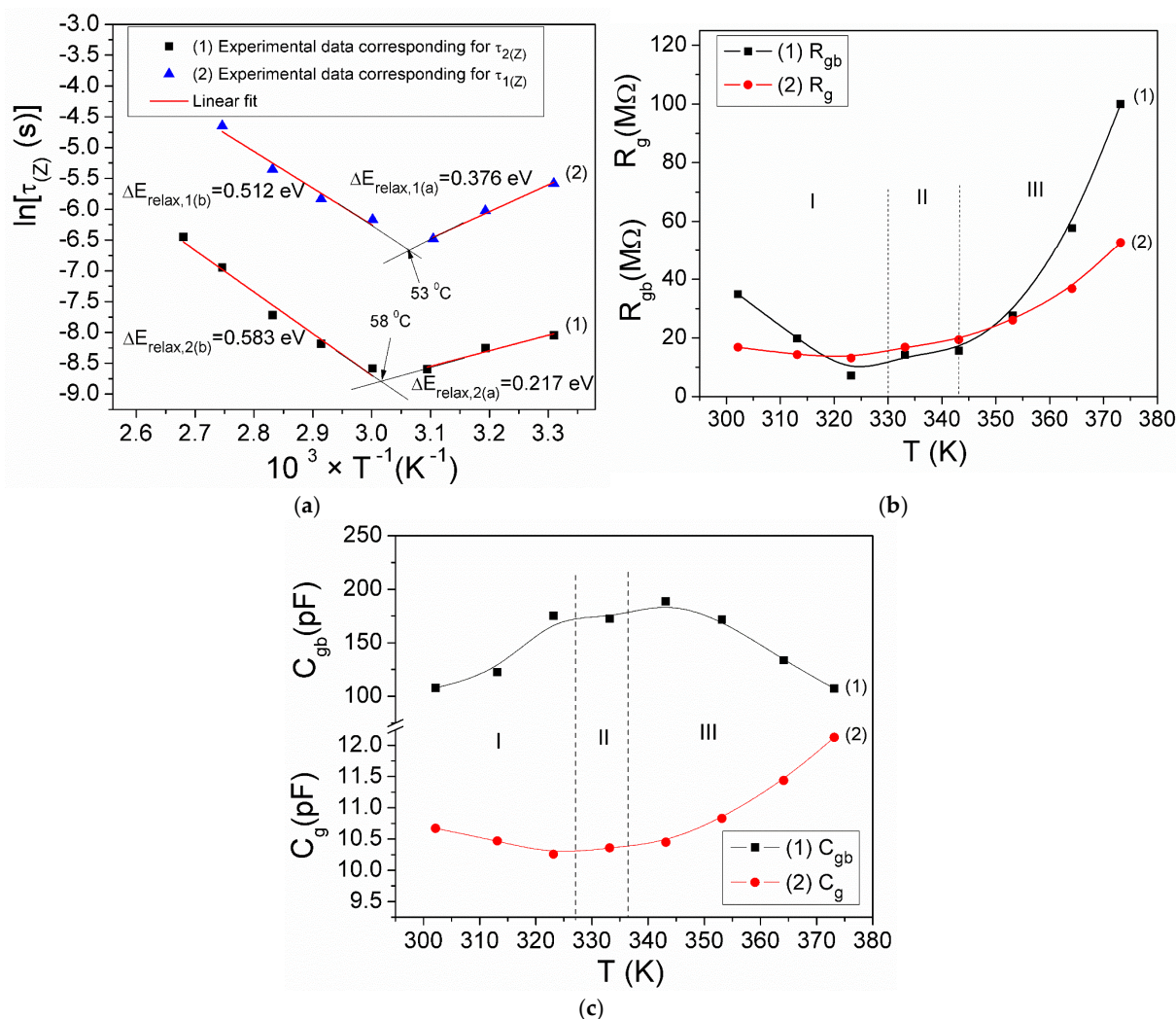


Figure 3. $\ln\tau_z$ (T^{-1}) plots for aluminum-doped sodium tantalate material (a); the temperature dependence of the resistances R_g and R_{gb} (b) and of the capacitances C_g and C_{gb} (c) extracted from fitting the data. Region II is the phase transition region.

An analysis of the impedance spectra in Figure 2c was performed by fitting/modeling the experimental data with an equivalent electrical circuit [43,51]. This circuit consists of two parallel circuits in series, representing the contributions of the grains and grain boundaries, along with a negligible series resistance (R_o) corresponding to the electrode interface, which is not accounted for in conventional RC models [44]. The grain circuit comprises a parallel combination of R_g and $(\text{CPE})_g$, while the grain boundary circuit consists of R_{gb} and $(\text{CPE})_{gb}$ (see Figure 2c). Here, CPE denotes a constant phase element, indicating a deviation from the ideal Debye model [52,53]. The impedance of the CPE, for the grains and grain boundaries [44,54], at each temperature is given by the following equation [55]:

$$Z_{\text{CPE}} = Q^{-1}(i \cdot 2\pi f)^{-n}, \quad (i = \sqrt{-1}) \quad (3)$$

where Q and n are parameters of the impedance of CPE. It should be noted that the exponent n varies between 0 and 1 when the CPE changes from ideal resistor behavior ($n = 0$) to

perfect capacitor behavior ($n = 1$). In Table 1, the values of the parameters R_0 , R_g , R_{gb} , Q , and n are indicated, obtained by fitting the experimental data of the Nyquist plots, $Z''(Z')$ of the sample for each temperature from Figure 2c, with the equivalent electrical circuit from the inset of Figure 2c.

Table 1. The parameters obtained by fitting the experimental data of the Nyquist plots at different temperatures, using the equivalent circuit from the inset of Figure 2c.

T (°C)	R_0 (Ω)	R_{gb} (M Ω)	R_g (M Ω)	Q_{gb}	n_{gb}	Q_g	n_g
29	1.0434×10^{-12}	34.943	16.793	1.9062×10^{-10}	0.89787	1.35×10^{-11}	0.97274
40	1.0322×10^{-12}	19.741	14.271	2.3125×10^{-10}	0.89462	1.351×10^{-11}	0.97106
50	9.8438×10^{-13}	7.1522	13.108	3.3726×10^{-10}	0.9021	1.3437×10^{-11}	0.96735
60	1.1649×10^{-12}	14.176	16.808	3.1193×10^{-10}	0.90139	1.3938×10^{-11}	0.96559
70	1.1444×10^{-12}	15.582	19.372	3.3134×10^{-10}	0.90332	1.4235×10^{-11}	0.96363
80	1.0571×10^{-12}	27.586	26.008	2.7085×10^{-10}	0.91491	1.496×10^{-11}	0.96047
91	1.0491×10^{-12}	57.732	36.84	1.8453×10^{-10}	0.93365	1.6008×10^{-11}	0.95674
100	8.9193×10^{-13}	100	52.739	1.3167×10^{-10}	0.95467	1.7301×10^{-11}	0.9517

From Table 1, it is observed that the n_g values of the grains are closer to unity (between 0.95 and 0.97) than those of the grain boundaries, n_{gb} (0.89–0.95), which indicates that the grains behave like ideal capacitors more than the grain boundaries. This result could be associated with the heterogeneity in the material studied, as observed in the SEM-EDX analysis (Figure 1b–d). Also, from Table 1, the thermal variation throughout the temperature range of R_g and R_{gb} values, extracted from Nyquist plot fitting, was obtained, as presented in Figure 3b,c.

As shown in Figure 3b, the temperature dependence of both the grain resistance (R_g) and the grain boundary resistance (R_{gb}) can be divided into three distinct regions. In region I, R_g and R_{gb} decrease as the temperature rises up to approximately 325–330 K. This is followed by a plateau in region II (330–340 K), where R_g and R_{gb} remain approximately constant. Finally, in region III, R_g and R_{gb} increase with further temperature rises. This thermal behavior suggests that the plateau (region II) represents a phase transition, during which the Al-doped sodium tantalate perovskite transitions from a semiconductor phase to a conductor phase, consistent with the trends shown in Figure 3a. Similar phase transitions have been reported by other authors [54–56] for various perovskite materials.

As shown in paper [57], the capacitance of constant phase element CPE can be computed with the relation:

$$C = \left(R^{1-n} Q \right)^{1/n} \quad (4)$$

where the value of $n < 1$ (see Table 1) represents the deviation from the ideal Debye behavior, for which $n = 1$. From Table 1, by knowing the corresponding R , Q , and n values and using relation (4), we determined the grain boundary capacitance (C_{gb}) and the grain capacitance (C_g) for each temperature, and in Figure 3c the temperature dependence of these capacitances is shown.

As can be observed in Figure 3c, the temperature dependence of both the grain capacitance (C_g) and the grain boundary capacitance (C_{gb}) can be divided into three regions, similar to the temperature dependence of the R_g and R_{gb} resistances in Figure 3b. In the first region (I), C_g decreases while C_{gb} increases with temperature up to approximately 325 K. This is followed by a plateau in region II (325–336) K, where both C_g and C_{gb} remain nearly constant. Finally, in region III, C_g increases and C_{gb} decreases with rising temperature (see Figure 3c). As a result, the plateau regions (region II) in Figure 3b,c are nearly identical and represent the semiconductor-to-conductor phase transition of the

aluminum-doped NaTaO₃ perovskite material. This transition temperature range aligns with the complex impedance measurements shown in Figure 3a and the data presented further below.

3.3. DC Conductivity and Complex Dielectric Permittivity

An important way to study the macroscopic movement of charge carriers through the investigated material is to evaluate its electrical conductivity using the Z' and Z'' values of the complex impedance and the relation:

$$\sigma = \frac{Z'}{Z'^2 + Z''^2} \frac{d}{A} \quad (5)$$

where d and A are the thickness of the sample and its cross-sectional area. In Figure 4a, the frequency dependence of the conductivity σ of the aluminum-doped sodium tantalate (US-Al sample) over the frequency range (20 Hz–2 MHz) and at different temperatures between (29 and 100) °C is shown.

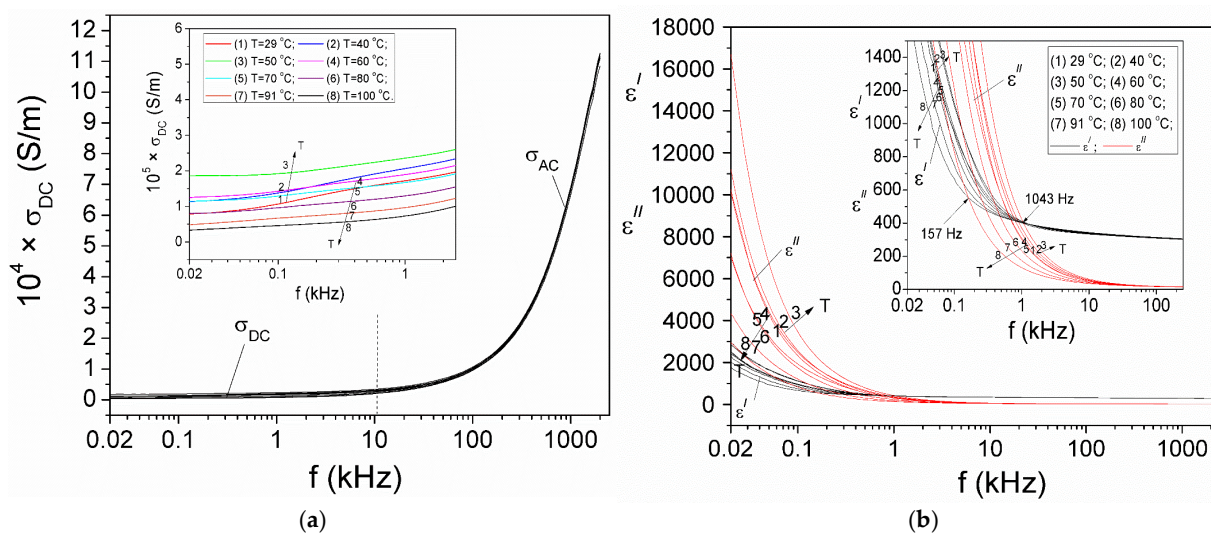


Figure 4. (a) The frequency dependence of the electrical conductivity of the US-Al sample at different temperatures (the inset shows the variation in the DC component of the conductivity with frequency and temperature, in the low-frequency range between 20 Hz and 1200 Hz). (b) The frequency dependence of the real (ϵ') and imaginary (ϵ'') components of the complex permittivity of the US-Al sample at different temperatures (the inset shows the variation in ϵ' and ϵ'' with frequency and temperature, around the frequencies at which the two components intersect).

The low-frequency plateau observed at all investigated temperatures (see Figure 4a) is attributed to the contribution of static conductivity (σ_{DC}), while the high-frequency dispersion is associated with dynamic conductivity (σ_{AC}) [58]. The identification of the appropriate conductivity mechanism from the investigated sample (US-Al) can be done based on the analysis of the frequency spectra in Figure 4a and Jonscher's universal power law [59,60]:

$$\sigma(\omega, T) = \sigma_{DC}(\omega, T) + \sigma_{AC}(\omega, T) \quad (6)$$

and

$$\sigma_{AC}(\omega, T) = A_0(T)\omega^n(T) \quad (7)$$

In these relations, ω represents the angular frequency, and the coefficient A_0 and exponent n are constants that depend on the temperature and the intrinsic properties of the sample material under study [60,61].

Based on the Z' and Z'' values of the complex impedance in Figure 2a,b, the real (ϵ') and imaginary (ϵ'') components of the complex dielectric permittivity at different frequencies between 20 Hz and 2 MHz and different temperatures between (29 and 100) °C for the investigated sample were determined with relations (8) and (9). The results are shown in Figure 4b.

$$\epsilon' = \frac{1}{\omega \epsilon_0} \frac{Z''}{Z'^2 + Z''^2} \frac{d}{A} \quad (8)$$

$$\epsilon'' = \frac{1}{\omega \epsilon_0} \frac{Z'}{Z'^2 + Z''^2} \frac{d}{A} \quad (9)$$

where ϵ_0 is the dielectric permittivity of the free space.

As shown in Figure 4b, both the real (ϵ') and imaginary (ϵ'') components of the complex dielectric permittivity gradually decrease with frequency across the entire range for all temperatures. At the low-frequency limit (20 Hz), ϵ' values are high—ranging from 1600 to 2800, depending on temperature—before decreasing as frequency increases. This behavior aligns with Koop's phenomenological theory [62,63] and Maxwell–Wagner interfacial polarization, a process involving electron exchange between ions of the same molecule [64]. An electric field applied to the material acts on the positive and negative charges, leading to the creation of a large number of dipoles [63,65]. The high dielectric constant at low frequencies is explained by the fact that the dipoles can easily follow the quasi-static field at these frequencies. By increasing frequency, an increasingly smaller number of dipoles can follow the oscillations of the electric field so that the dielectric constant ϵ' decreases. At the same time, at a certain frequency, the value of ϵ' can increase by increasing the temperature, which suggests the presence of thermally activated charge carriers. Also, from Figure 4b, it is observed that the values of the ϵ'' component are much higher than those of the ϵ' component, varying between 3000 and 16,300 depending on the temperature. This behavior at low frequencies shows that the conduction losses are higher than those due to dielectric relaxation [66]. Increasing the frequency, the values of the ϵ'' component decrease for all investigated temperatures, so that at frequencies between 157 Hz and 1043 Hz, they become smaller than those of the ϵ' component, as observed in the inset of Figure 4b. This indicates that for frequencies above those between 157 Hz and 1043 Hz, dielectric relaxation losses become important [66].

Considering the Debye equation [66] for the imaginary component ϵ'' , the following relationship can be written:

$$\epsilon''(\omega) = \epsilon''_{\text{cond}}(\omega) + \epsilon''_{\text{rel}}(\omega) = \frac{\sigma_{\text{DC}}}{2\pi f \epsilon_0} + \epsilon''_{\text{rel}}(\omega) \quad (10)$$

In Equation (10), $\sigma_{\text{DC}}/2\pi f \epsilon_0$ represents the conduction losses (ϵ''_{cond}) and (ϵ''_{rel}) the dielectric relaxation losses. Considering the σ_{DC} values of the static conductivity (Figure 4a) and the total values of the ϵ'' component (Figure 4b) at all investigated temperatures, the dielectric relaxation losses (ϵ''_{rel}) were determined from Equation (10) by eliminating the conduction losses. The results are presented in Figure 5.

As can be seen in Figure 5, ϵ''_{rel} exhibits a maximum at frequency f_m for each temperature T , which indicates the existence of a relaxation process attributed to interfacial relaxation [59–61].

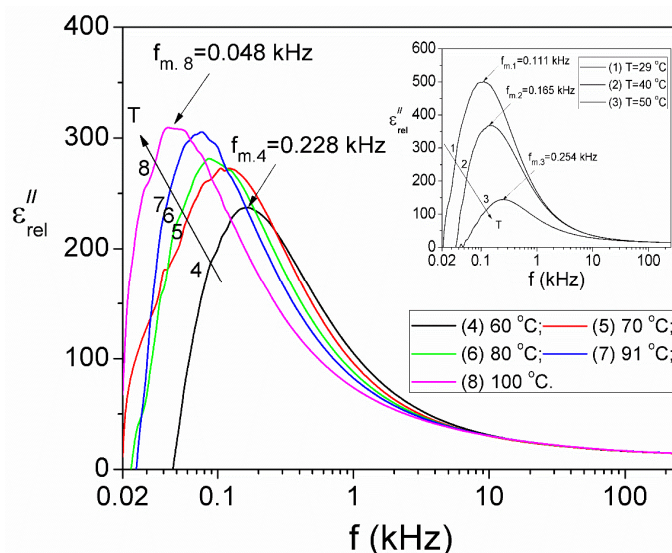


Figure 5. Frequency dependence of the imaginary component (ϵ''_{rel}) of the complex dielectric permittivity, due to dielectric relaxation, at different temperatures for the aluminum-doped sodium tantalate sample.

3.4. Identifying the Transition Temperature

Considering the inset of Figure 4a, which shows the frequency and temperature dependence of the static conductivity σ_{DC} from low frequencies (20 Hz–1200 Hz), by fitting with a straight line, the values of σ_{DC} were determined at each temperature T . In Figure 6a, the dependence of $\sigma_{DC}(T)$ of the aluminum-doped sodium tantalate sample (US-Al) is shown.

As can be seen in Figure 6a, by increasing the temperature to approximately (50–60) °C, the conductivity σ_{DC} increases, then it starts to decrease by increasing the temperature to 100 °C. As a result, in the low-temperature range (below 60 °C), the increase in the conductivity σ_{DC} indicates semiconductor-like behavior of the analyzed sample. In other words, it can be said that near the temperature of (50–60) °C, there is a transition from a semiconductor-like behavior to a conductor-like behavior of the sample (above 60 °C) when σ_{DC} decreases [32]. The behavior of the Al-doped NaTaO₃ sample in Figure 6a in the temperature range up to (50–60) °C is in agreement with Mott's VRH theory [47].

As a result, considering the VRH model, by doping the NaTaO₃ sample with Al ions, with increasing temperature, the hopping distance of charge carriers between localized states decreases [32], which determines the increase in the static conductivity of the investigated sample with temperature. At the same time, it can be said that the phase boundary acts as a barrier between the low-conductivity and high-conductivity regions.

According to the VRH model, conductivity σ_{DC} is given by the relation:

$$\sigma_{DC} = \sigma_0 \exp\left(-\frac{B}{T^{1/4}}\right) \quad (11)$$

where σ_0 is the pre-exponential factor of conductivity and B is given by the relation:

$$B = \left(\frac{4E_{A,cond}}{kT^{3/4}}\right) \quad (12)$$

In Equation (12), $E_{A,cond}$ represents the thermal activation energy of static conductivity. In Figure 6b, the dependence $\ln(\sigma_{DC})(1/T^{1/4})$ is presented according to the VRH model (Equation (11)). By fitting the experimental dependence in Figure 6b with a straight line, it is found that the conductivity slope changes at a temperature of 56 °C, which is the transition

temperature from semiconductor-type behavior to a conductor-type behavior. This result is like the experimental dependence, $\ln\tau_{(Z)}(1/T)$, in Figure 3a, which shows two different slopes at (50–60) °C, associated with two ranges of different electrical conductivities of the sample.

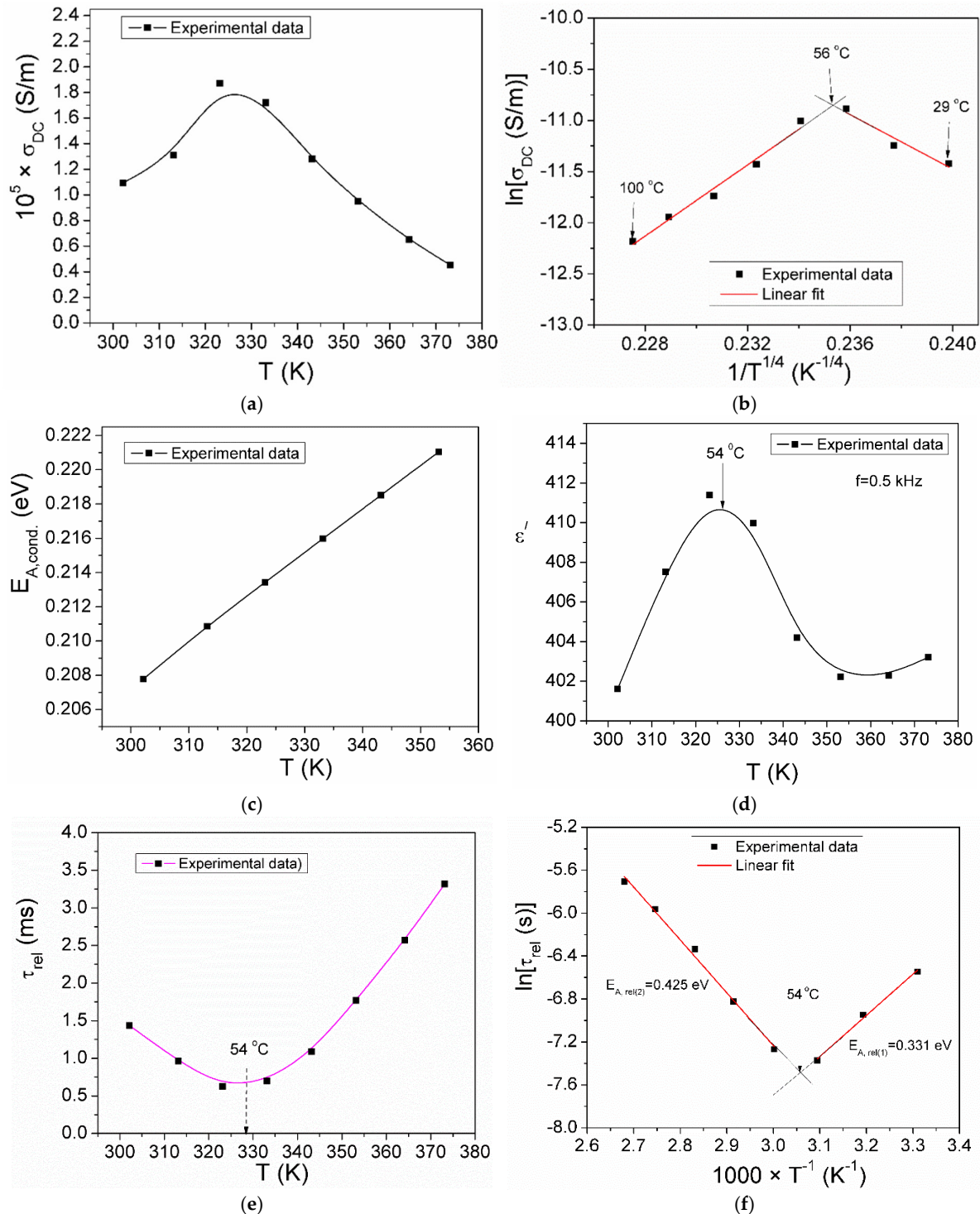


Figure 6. (a) The temperature dependence $\sigma_{DC}(T)$ of the static conductivity of the US-Al sample; (b) the dependence $\ln(\sigma_{DC})/(1/T^{1/4})$ for the investigated sample; (c) variation in the thermal activation energy of electrical conduction with temperature, $E_{A,cond.}(T)$, for the US-Al sample; (d) temperature dependence of the real component ϵ' of complex permittivity at fixed frequency 0.5 kHz; (e) temperature dependence of the relaxation time (τ_{rel}) due to the dielectric relaxation and (f) the $\ln(\tau_{rel})/(1/T)$ plots for the US-Al sample.

Knowing the slope B of the fit corresponding to the temperature range (29–56) °C ($B = 132.96394$) in which the sample behaves like a semiconductor (see Figure 6b), from Equation (12), the thermal activation energy of conduction, $E_{A,cond}$, over this temperature range was determined. As a result, Figure 6c shows the temperature dependence $E_{A,cond}(T)$ for the US-Al sample in the temperature range below 60 °C, in which the investigated sample behaves like a semiconductor.

From Figure 6c, it is observed that $E_{A,cond}$ increases linearly with increasing temperature in the range in which the sample behaves like a semiconductor, which means that the electrical conduction in the sample can be explained by the process of charge carriers hopping between the localized states [47], according to Mott's VRH model. The value of $E_{A,cond}$ for the aluminum-doped sodium tantalate sample increases from approximately 0.208 eV to 0.222 eV, correlating very well with the result obtained from electrical relaxation measurements for the barrier energy between the localized states, $\Delta E_{relax,2(a)} = 0.217$ eV, as shown in Figure 3a, Section 3.2. This value corresponds to the temperature range of (29–60) °C, in which the sample behaves as a semiconductor, and is correlated with the main perovskite phase NaTaO_3 of the investigated sample, being similar to the values indicated by other authors for similar perovskite samples with semiconductor behavior [32,44,61]. It should be noted that this value (0.217 eV) represents the average value of the thermal conduction activation energy, $E_{A,cond}$, corresponding to the investigated temperature range of (29–56) °C in Figure 6c, which justifies the interpretation of the fact that ΔE_{relax} represents the barrier energy necessary for charge carrier hopping between localized states.

The identification of the semiconductor-to-conductor transition temperature of the aluminum-doped NaTaO_3 sample can also be supported by the analysis of the dielectric permittivity with temperature. In this regard, Figure 6d presents the temperature variation in the real component ϵ' at $f = 0.5$ kHz, corresponding to the low-frequency plateau (see Figure 4b) attributed to the static conductivity contribution.

From Figure 6d, it is observed that by increasing the temperature, ϵ' increases and reaches a maximum at a temperature of 54 °C, after which it starts to decrease with a further increase in temperature towards 100 °C. The temperature of 54 °C at which ϵ' changes its direction of variation can be attributed to the transition temperature [67] from semiconductor-type behavior to conductive behavior of the sample. The value obtained is very close to the value obtained both from static conductivity measurements (56 °C) (see Figure 6b) and complex impedance measurements (58 °C) (see Figure 3a). On the other hand, as previously shown in Figure 5, the imaginary component ϵ''_{rel} , due to dielectric relaxation, has a maximum at frequency f_m and at each temperature T , which indicates the existence of a relaxation process attributed to interfacial relaxation [59–61], characterized by the relaxation time τ_{rel} . From the experimental values of f_m (Figure 5) and the Debye equation [48], the relaxation time τ_{rel} was determined, and in Figure 6e, the temperature dependence of τ_{rel} for the investigated sample, US-Al, is presented.

The variation in the relaxation time τ_{rel} with temperature in Figure 6e indicates a decrease up to a temperature close to 54 °C, followed by an increase in τ_{rel} with increasing temperature up to 100 °C, which indicates that τ_{rel} has an Arrhenius-type behavior of the form of Equation (2) at temperatures up to 54 °C. The linear fit of the experimental dependence $\ln\tau_{rel}(1/T)$, considering Arrhenius-type dependence (Figure 6f), presents two different slopes, corresponding to two temperature ranges, namely, below 54 °C and above 54 °C, which are associated with different electrical conductivities of the sample in these temperature ranges. Also, from the fit slope, the activation energy of the dielectric relaxation process $E_{A,rel}$ for the two temperature ranges can be determined. The values obtained are $(E_{A,rel})_1 = 0.331$ eV and $(E_{A,rel})_2 = 0.425$ eV, in agreement with the value obtained by other authors for similar perovskite samples [68,69]. Consequently, the temperature of 54 °C,

where changes in the activation energy of relaxation (type of conductivity) occur, represents the transition temperature from semiconductor-type behavior to conductor-type behavior of the sample. The value obtained for the transition temperature (54 °C) is very close to that obtained from complex impedance measurements (58 °C) (see Figure 3a), or from static electrical conductivity measurements (56 °C) (see Figure 6b), the real component of permittivity and dielectric relaxation time (54 °C) (see Figure 6d,f).

4. Conclusions

Aluminum-doped NaTaO₃ perovskite (US-Al), synthesized by an ultrasonic method, exhibits relatively low crystallinity and comprises two phases: the main perovskite phase, NaTaO₃, and a secondary phase identified as Na₂Ta₄O₁₁ (natrotantite). The optical bandgap of the US-Al sample is 3.77 eV, which is lower than that of the undoped NaTaO₃ perovskite (4 eV), suggesting enhanced infrared absorption and improved photocatalytic potential. For the first time in an FT-IR spectrum, a strong absorption band was identified at 670 cm^{−1}, attributed to Ta-O and Al-O stretching vibrations induced by Al incorporation.

In the frequency range (20 Hz–2 MHz) and at different temperatures between (29 and 100) °C, the imaginary component Z'' presents two maxima, indicating the hopping of charge carriers between localized states with relaxation times τ_1 and τ_2 . The analysis of $\tau(T)$ yields barrier energies between the localized states of 0.217 eV (below 58 °C) and 0.583 eV (above 58 °C), corresponding to two conductivity regimes. Therefore, a transition at ~58 °C is highlighted, marking a change from semiconducting to conducting behavior of the sample.

The fitting of the experimental data $Z''(Z')$ for each temperature shows two semi-circular arcs associated with the coexisting phases and the contributions of the grains and grain boundaries. The fitting was performed using two parallel equivalent circuits (R//CPE) corresponding to the grains and grain boundaries, respectively, and confirms this interpretation. These temperature-dependent grain (R_g) and grain boundary (R_{gb}) resistances show a plateau between 330 and 340 K, consistent with a phase transition region that encompasses the identified transition temperature.

The conductivity spectrum $\sigma(f)$ follows the universal Jonscher law at all temperatures. The static conductivity $\sigma_{DC}(T)$ shows a change in slope near 56 °C, further supporting the semiconductor–conductor transition. The dielectric analysis of the ϵ' and ϵ'' components, after eliminating conduction losses, indicates interfacial polarization and dielectric relaxation with relaxation time τ_{rel} . The temperature dependence of the ϵ' component at 0.5 kHz shows a change in the direction of variation in ϵ' at ~54 °C. At the same time, the $\tau_{rel}(T)$ analysis highlights that at the same temperature (~54 °C), a change in the activation energy of the dielectric relaxation process occurs, corresponding to two conductivity regimes. As a result, the temperature of 54 °C can be attributed to the semiconductor–conductor transition temperature of the sample, in close agreement with values obtained from conductivity (56 °C) and impedance (58 °C) analyses.

Overall, the ultrasonic synthesis route uniquely produces a two-phase system that enables the observation of a low-temperature semiconductor-to-conductor transition, a phenomenon not present in single-phase materials prepared by the sol–gel method.

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Abbreviations

The following abbreviations are used in this manuscript:

US-Al Ultrasonic Aluminum
VRH Variable Range Hopping

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