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Electrophysical properties of the Rochelle salt / zeolite A nanocomposite (Part 2)

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Abstract. This paper studies the electrical conductivity, permittivity, and dielectric losses of a nanocomposite obtained by impregnating a NaA zeolite host matrix with Rochelle salt (potassium sodium tartrate tetrahydrate, $\text{KNaC}_4\text{H}_4\text{O}_6 \times 4\text{H}_2\text{O}$) as a guest substance over the frequency range from 0.1 Hz to 1 MHz upon heating from 273 K to 493 K. This study discusses the characteristic features of the frequency and temperature dependences of the electrophysical properties of the Rochelle salt / zeolite A nanocomposite.

Keywords: ferroelectrics, Rochelle salt, permittivity, dielectric losses, electrical conductivity, zeolite, nanocomposite

Introduction

The matrix method for creating nanocomposite materials, proposed in the late 20th century (Bogomolov 1978; Stucky, Mac Dougall 1990), offers numerous advantages, including the ability to study ensembles of nanoparticles at high concentrations and with ultra-small sizes (up to 1 nm).

This paper employs this method to study size dependence of the upper ferroelectric Curie point T_C in the well-known crystalline substance — Rochelle salt (double potassium sodium tartrate $\text{KNaC}_4\text{H}_4\text{O}_6 \times 4\text{H}_2\text{O}$; RS in what follows) embedded into a NaA-type zeolite matrix.

Previous investigations have examined the ferroelectric properties of Rochelle salt dispersed in various porous media. In the case of RS embedded into a microporous glass matrix with particle diameters of ~ 7 nm ferroelectric features appeared only on the derivative ($d\epsilon/dT$) curve (Colla et al. 1996). RS inclusions in nanoporous alumina with pore diameters from 10 to 40 nm retain a ferroelectric state at temperatures exceeding the Curie point of bulk crystals by several tens of degrees (Rogazinskaya et al. 2009). In contrast, a low-temperature shift of the Curie point T_C of the RS guest substance’s upper ferroelectric transition due to confined geometry has been observed in mesoporous sieves MCM-41 with 2.6 nm pores (Tien et al. 2008), as well as in NaA zeolite (Matveeva, Solovyev 2022; Puchkov et al. 2025). NaA zeolite is a porous dielectric matrix containing an ordered system of interconnected voids and

three-dimensional channels with diameters of ~ 1 nm (Breck 1974). The lattice parameters of Rochelle salt ($a \approx 1.2$ nm, $b \approx 1.4$ nm, and $c \approx 0.6$ nm) (Solans et al. 1997) are rather large; thus, we assume that RS molecules can only reside within the α -cavities of the NaA crystal, where interactions between neighbouring molecules are possible.

In a previous work (Matveeva et al. 2024) we studied electrophysical properties of RS ferroelectric / zeolite NaA near the ferroelectric phase transition temperature over the frequency range from 100 Hz to 100 kHz. It was found that this nanocomposite shows a shift of the RS upper ferroelectric transition Curie point by ~ 5 K towards lower temperatures, relative to the phase transition temperature in a bulk ferroelectric ($T_C = 297$ K).

The present study aims to experimentally determine the electrophysical characteristics of this nanocomposite material over a wide range of frequencies (0.1 Hz–1 MHz) and temperatures (273–493 K).

Experiment

We used granules (~ 2 mm in diameter) of NaA zeolite as a porous dielectric host matrix. A saturated aqueous solution at room temperature introduced the Rochelle salt guest substance into the zeolite voids; thereafter, the ferroelectric / zeolite (RS / NaA) nanocomposite sample was washed with distilled water and dried at $T = 300$ K. To remove Rochelle salt from the sample surface, it was subjected to mechanical processing (surface grinding), and then shaped into a tablet with a diameter of 2 mm and a thickness of approximately 0.5 mm. After that, the sample was clamped between the metal electrodes of the measuring cell.

The complex dielectric permittivity was measured using a Novocontrol Concept 41 spectrometer at the Interdisciplinary Resource Center for Collective Use of Herzen State Pedagogical University of Russia (Modern Physical and Chemical Methods for the Research and Development of Materials for Industry, Science, and Education). The frequency range spanned from 0.1 Hz to 1 MHz, and measurements were conducted at controlled temperatures between 273 and 493 K.

Results and discussion

Figure 1 shows frequency dependences of the electrical conductivity for the RS ferroelectric / zeolite A nanocomposite in the ferroelectric and paraelectric phases. One can observe a power-law frequency dependence of conductivity, $\sigma \sim f^s$, which is often found in many materials (Jonscher 1972; Mott, Davis 1979) and may arise from two distinct mechanisms: (1) a rotational motion of molecular dipoles with frictional losses (when $s \approx 2$), and (2) a translational motion of free electric charges ($0 < s < 1$). For the experimental results shown in Fig. 1, $s \approx 1.3$ ($10^4 \text{ Hz} \leq f \leq 10^6 \text{ Hz}$). This exponent value can be explained by the simultaneous existence of both mechanisms; a similar situation has already been observed in another ferroelectric material (Mostafa et al. 2005).

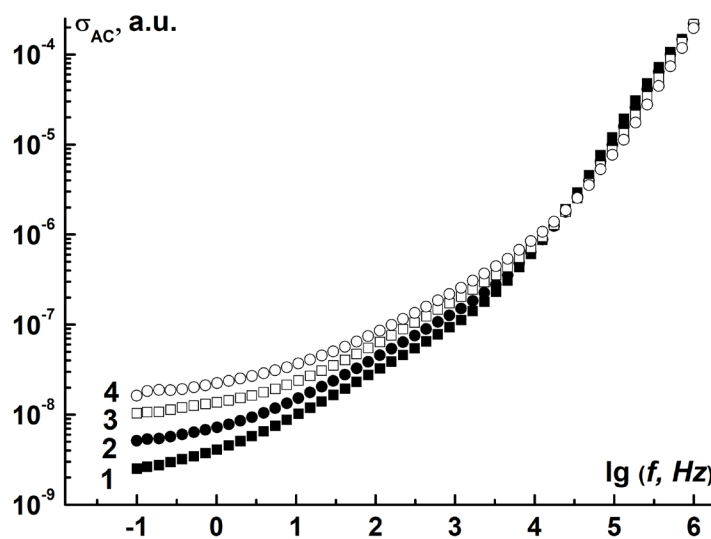


Fig. 1. Frequency dependences of the electrical conductivity for the RS / NaA composite at 283 K (curve 1), 293 K (curve 2), 303 K (curve 3), 313 K (curve 4)

The frequency dependences of the dielectric losses for the RS / NaA composite (Fig. 2) display a decrease at relatively low frequencies — up to approximately 10 kHz — after which $\text{tg}\delta$ begins to increase. A similar pattern is observed in the frequency dependences of the imaginary part of the permittivity (Fig. 3). This high-frequency increase can be explained by proton conductivity (Colomban, Novak 1988; Ovchinnikova et al. 1997). Figures 2 and 3 again show an increase in dielectric losses at low frequencies, as well as a shift of the graph minima toward higher frequencies at the phase transition of Rochelle salt to the paraelectric phase.

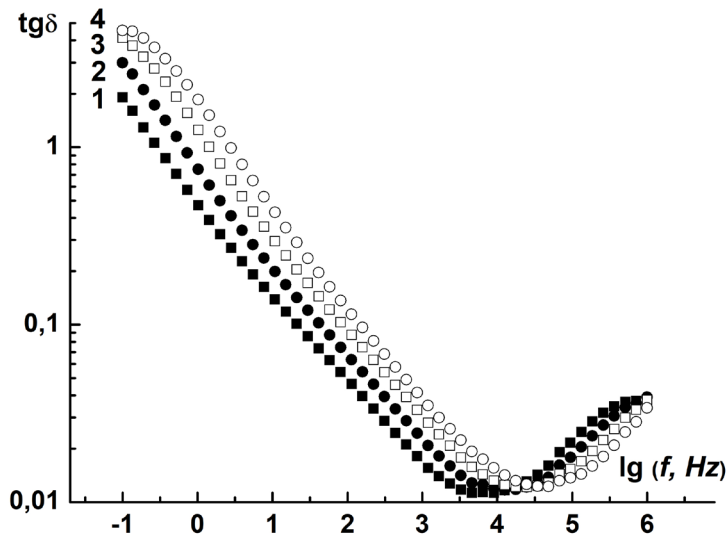


Fig. 2. Frequency dependences of the dielectric losses $\text{tg}\delta$ for the RS / NaA composite at 283 K (curve 1), 293 K (curve 2), 303 K (curve 3), 313 K (curve 4)

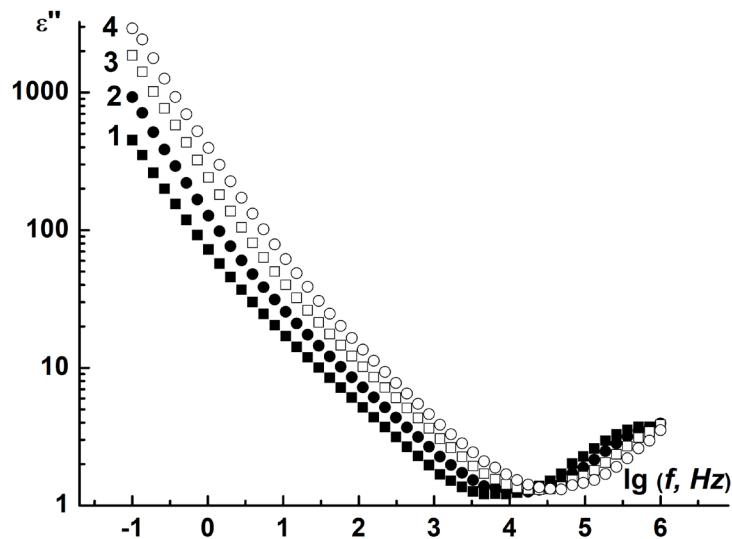


Fig. 3. Frequency dependences of the imaginary part of dielectric permittivity ϵ'' for the RS / NaA composite at 283 K (curve 1), 293 K (curve 2), 303 K (curve 3), 313 K (curve 4)

Figure 4 shows temperature dependences of the electrical conductivity for the RS / NaA composite at low frequencies. One can see two maxima at temperatures of approximately 323–333 and 363 K. It should be noted that at $T \approx 329$ K, Rochelle salt decomposes into a mixture of sodium and potassium tartrates (Jona, Shirane 1962); this process is often referred to as the ‘melting’ of Rochelle salt. According to our assumption, the second maximum likely corresponds to the subsequent stage of decomposition of these components. The temperature dependences of the dielectric losses (Fig. 5) exhibit similar peculiarities.

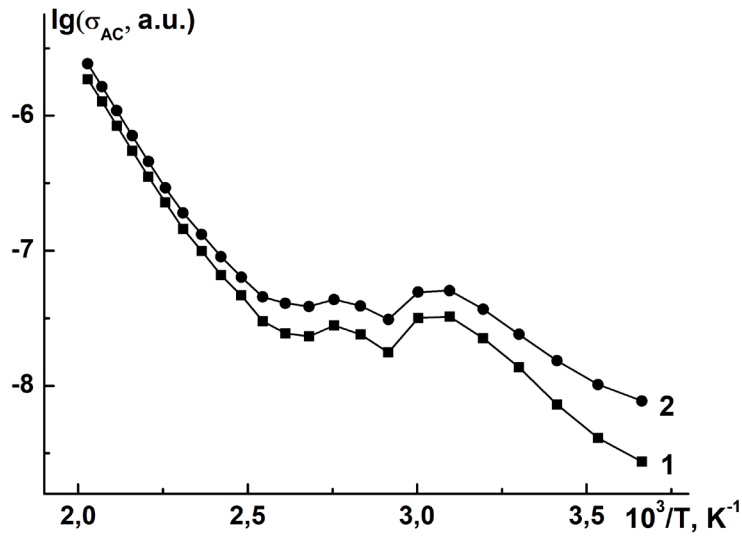


Fig. 4. Temperature dependences of the electrical conductivity for the RS / NaA composite at 1 Hz (curve 1) and 10.8 Hz (curve 2)

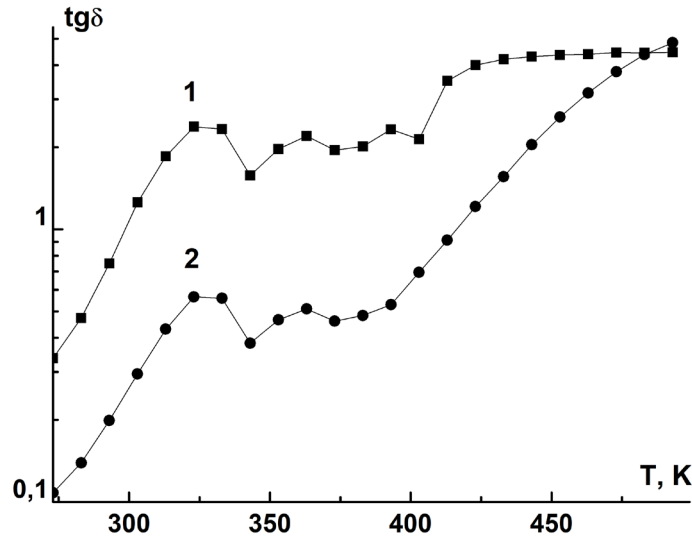


Fig. 5. Temperature dependences of the dielectric losses $\text{tg}\delta$ for the RS / NaA composite at 1 Hz (curve 1) and 10.8 Hz (curve 2)

Conclusions

The ferroelectric / zeolite nanocomposite material obtained by infiltrating the NaA zeolite host matrix with the Rochelle salt guest substance exhibits a power-law frequency dependence of electrical conductivity which can be explained by the simultaneous existence of two mechanisms: (1) rotational motion of dipoles with frictional losses, and (2) translational motion of free electric charges.

Upon heating from room temperature, the study observes a significant increase in dielectric losses at frequencies below 10 kHz, and this increase continues into the paraelectric phase.

Conflict of Interest

The authors declare that there is no conflict of interest, either existing or potential.

Author Contributions

All the authors discussed the final work and took part in writing the article.

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