DIELECTRIC RELAXATION IN BROMOBENZENE-BENZENE AND BROMOBENZENE-HEXANE BINARY SYSTEMS: THERMODYNAMIC ANALYSIS AND SOLVENT INFLUENCE

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The influence of the nature of a nonpolar solvent on the molecular dynamics and thermodynamics of dielectric relaxation of bromobenzene was investigated in the binary bromobenzene–benzene and bromobenzene–hexane systems. The real and imaginary components of the complex dielectric permittivity were experimentally determined in the microwave range at a wavelength of λ = 12.8 mm, corresponding to a frequency of approximately 23.4 GHz, over the temperature range of 293-193 K. The study aimed to establish the relationship between molecular dynamics, thermodynamic parameters, and solvent nature during the dielectric relaxation process. Measurements were performed at different bromobenzene concentrations under continuous cooling; the experimental uncertainty did not exceed 1% for ε′ and 4% for ε″. Relaxation times were determined from the temperature dependences of the dielectric permittivity, and the thermodynamic activation parameters—Gibbs free energy ΔF, enthalpy ΔH, and entropy ΔS – were calculated within the framework of the absolute reaction-rate theory. The obtained ΔF values ranged from 2.38 to 2.76 kcal mol^{-1} for the bromobenzene–benzene system and from 2.08 to 2.76 kcal mol^{-1} for the bromobenzene–hexane system, whereas ΔH varied from 1.43 to 1.65 and from 1.06 to 1.65 kcal mol^{-1}, respectively. The activation entropy exhibited negative values, indicating an increase in molecular ordering during the relaxation transition. The results demonstrate that the solvent substantially affects the energy barrier and the degree of molecular ordering. The novelty of this study lies in establishing a correlation between the thermodynamic parameters of bromobenzene dielectric relaxation and the molecular properties of benzene and hexane, demonstrating the potential of dielectric relaxation as a sensitive tool for probing intermolecular interactions and the solvation environment.

Keywords: dielectric relaxation, bromobenzene, benzene, hexane, thermodynamics, microwave range, relaxation time, intermolecular interactions.

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1. INTRODUCTION

Dielectric relaxation in molecular liquids and binary solutions is a sensitive probe of molecular mobility, orientational polarization, and intermolecular interactions [1,2]. When a polar molecule is subjected to an alternating electric field, its dipole moment undergoes reorientation, and the characteristic relaxation time reflects the rate at which the molecular system responds to the applied field. Consequently, temperature-dependent dielectric relaxation provides information not only on molecular dynamics but also on the energetic and structural factors governing molecular reorientation [3,4]. In binary systems, this problem becomes particularly important because the relaxation behavior of a polar component is determined not only by its intrinsic molecular properties but also by the local environment created by the solvent. Changes in solvent structure, molecular packing, and intermolecular interactions can modify the energy barrier associated with dipole reorientation and, therefore, alter the observed relaxation dynamics.

Two complementary approaches are commonly used to describe dielectric relaxation. The Debye theory relates the relaxation time to molecular size and the viscosity of the surrounding medium, providing a useful description of the dynamic response of polar molecules in liquids [5]. A different interpretation follows from the theory of absolute reaction rates, in which dielectric relaxation is treated as a thermally activated kinetic process [6,7]. Within this approach, the temperature dependence of the relaxation time can be related to thermodynamic activation parameters, including the activation free energy, enthalpy, and entropy. This framework makes it possible to go beyond the determination of a characteristic relaxation time and to examine the energetic barrier and degree of molecular ordering associated with molecular reorientation.

Previous dielectric studies have demonstrated that the relaxation behavior of polar molecules is strongly affected by molecular structure and by the nature of the surrounding medium. Investigations of halogenated aromatic compounds and their solutions have shown that changes in the molecular structure of the polar component are reflected in the temperature dependence of dielectric relaxation. In particular, previous studies of chlorobenzene and related systems have demonstrated the sensitivity of relaxation parameters to the molecular environment and solvent composition. The present study builds on these observations by focusing on bromobenzene, whose molecular structure and polar character make it a suitable probe for examining solvent-dependent molecular reorientation [8-10].

However, the available results do not provide a sufficiently systematic picture of how the nature of a nonpolar solvent modifies the thermodynamic characteristics of bromobenzene relaxation. In particular, the relationship between the experimentally determined relaxation time and the activation

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parameters of the relaxation process requires further clarification when the same polar molecule is studied in solvents with substantially different molecular characteristics. Thus, measurements of dielectric permittivity alone are insufficient to fully characterize the physical origin of the observed differences; a thermodynamic analysis of the temperature dependence of the relaxation time is also required.

The bromobenzene - benzene and bromobenzene - hexane binary systems provide a well-defined model for addressing this problem. In both systems, bromobenzene is the same polar molecular component, whereas benzene and hexane represent different nonpolar molecular environments. This design makes it possible to separate, to a significant extent, effects associated with the molecular properties of the relaxing species from those associated with the surrounding solvent. The comparison is therefore particularly useful for determining whether changes in dielectric relaxation originate from the intrinsic properties of bromobenzene or from differences in solvent-mediated intermolecular interactions and molecular organization. Earlier measurements of the dielectric properties of these systems have provided experimental evidence of concentration- and temperature-dependent behavior, including pronounced changes near the liquid–solid transition.

An additional feature of these systems is their behavior upon cooling through the phase-transition region. The experimental data show a pronounced decrease in both the real and imaginary components of the dielectric response upon solidification. In the bromobenzene–hexane solutions, the transition is noticeably broadened compared with the corresponding behavior of the pure polar component, whereas the effect is less pronounced in the benzene solutions. At sufficiently low temperatures, the orientational contribution to the dielectric polarization becomes strongly suppressed, providing an additional indication that the temperature evolution of the dielectric response is directly related to molecular mobility and phase state.

Despite these observations, a clear scientific gap remains: the solvent-dependent modification of the thermodynamic parameters governing bromobenzene dielectric relaxation has not been sufficiently established through a direct comparison of benzene and hexane environments over a broad temperature interval. In particular, it remains necessary to determine how the solvent affects the energetic barrier separating alternative equilibrium orientations of the bromobenzene dipole and how this effect is reflected in the activation enthalpy, free energy, and entropy. Addressing this gap requires combining temperature-dependent dielectric measurements with a kinetic–thermodynamic analysis of the relaxation process rather than considering the dielectric response solely as a function of temperature and concentration.

The present work addresses this problem by experimentally investigating the dielectric response of bromobenzene–benzene and bromobenzene–hexane binary systems at a microwave wavelength of $\lambda = 12.8$ mm over the temperature range 293–193 K and at different component concentrations. The temperature dependences of the dielectric permittivity are used to determine the relaxation time, after which the activation free energy, enthalpy, and entropy are evaluated within the framework of the absolute reaction-rate theory. Particular attention is given to comparing the two solvent environments in order to identify the role of solvent molecular structure in determining the energetic and ordering characteristics of bromobenzene reorientation.

The aim of this study is therefore to establish the relationship between the molecular dynamics of bromobenzene, the thermodynamic parameters of its dielectric relaxation, and the nature of the surrounding nonpolar solvent. The principal scientific novelty consists in establishing a direct correlation between the thermodynamic characteristics of bromobenzene relaxation and the molecular properties of benzene and hexane as solvents. The comparison demonstrates how solvent-dependent intermolecular interactions modify the potential barrier for dipole reorientation and the degree of molecular ordering in the binary systems. Thus, the work extends the interpretation of dielectric relaxation from a purely dynamic parameter toward a thermodynamic description of solvent-controlled molecular mobility.

2. MATERIALS AND METHODS

The experimental methodology is described in [11]. Measurements of the dielectric permittivity (ε') and dielectric loss tangent (ε") of the solutions under study were performed using a R2-66 panoramic standing wave ratio meter and a indicator device Ya2R – 67 indicator device in the microwave range at a wavelength $\lambda = 12.8$ mm. The temperature of the samples was controlled with an accuracy of ±0.1 K. The objects of study were placed in a specially designed short-circuited measuring waveguide cell at the end. The measuring cell was placed in a thermostat and contained a device for smooth adjustment of the solution layer thickness. The research results were obtained during continuous cooling at a rate of 0.1–0.2 degrees/min. The spread of measurement results for ε' is less than 1%, and for ε" less than 4%.

Bromobenzene, benzene, and hexane were thoroughly purified by rectification to achieve a high degree of purity – chemically pure. Double systems were prepared by mixing the components in specific molar ratios.

The relaxation time was determined using the formula [12,13]:

$$\tau = \frac{1}{\omega} \frac{\varepsilon''}{\varepsilon' - \varepsilon_\infty}$$

3. RESULTS AND DISCUSSION

Tables 1 and 2 present the results of measurements of dielectric permittivity (ε') and dielectric loss tangent (ε") for bromobenzene-benzene

and bromobenzene-hexane systems as a function of temperature and concentration. These results were obtained during continuous cooling at a rate of approximately 0.1–0.2 degrees/min. In most cases, a sharp decrease in ε′ and ε″ is observed at the solidification point. At the same time, it was noted that at the onset of solidification, the wave number q = Emin/Emax (E – electric field strength) passes through a sharply pronounced maximum. The wave number is not reflected in the values of ε", since at the same time there is a very rapid and significant shift of the standing wave node. It is possible that in this case, in order to obtain more complete information, it is necessary to take a larger number of experimental points in a very narrow temperature range near the solidification point. After the jump, there is a further decrease in ε' and a drop in ε" to zero. This behavior has already been noted for pure polar components and attributed to the existence of individual "free" molecules in liquids along with clusters [14-17]. These, in turn, contribute to the orientation polarization in a small temperature range below the solidification point. This point of view is confirmed to a certain extent in the case of hexane solutions, where such a transition is noticeably prolonged compared to a pure polar component (Fig. 1, 2). In benzene solutions, this is less pronounced.

Table 1

Temperature dependence of the dielectric constant ε' and dielectric loss ε" for the bromobenzene–benzene system, with bromobenzene concentrations in mole fractions: 1 – 0.459; 2 – 0.560; 3 – 0.718; 4 – 1.000

Bromobenzene – benzene								
t,^{0}C	x_2 = 0,459		x_2 = 0,560		x_2 = 0,718		x_2 = 1,000	
	ε'	ε''	ε'	ε''	ε'	ε''	ε'	ε''
Liquid state								
20	3,98	0,33	4,27	0,40	4,66	0,52	5,25	0,73
10	4,02	0,37	4,32	0,45	4,74	0,58	5,35	0,83
0	4,06	0,43	4,39	0,52	4,82	0,69	5,39	0,97
– 10	4,10	0,51	4,45	0,63	4,90	0,82	5,43	1,12
– 20	4,13	0,60	4,50	0,75	4,94	0,98	5,38	1,31
– 30	4,15	0,70	4,52	0,88	4,93	1,16	5,29	1,49
– 33	4,16	0,74	4,53	0,94	4,92	1,21	–	–
– 40	–	–	4,54	1,04	4,90	1,32	–	–
– 42	–	–	4,54	1,07	4,88	1,35	–	–
– 47	–	–	–	–	4,84	1,43	–	–
Solid state								
–31	–	–	–	–	–	–	3,12	0,31
–34	3,38	0,52	–	–	–	–	2,98	0,16
–38	3,00	0,22	–	–	–	–	2,87	0,08
–43	2,63	0,07	3,25	0,40	–	–	2,84	0,02
–48	2,57	0,02	2,81	0,12	3,17	0,34	2,83	–
–50	2,56	–	2,69	0,06	2,90	0,19	2,83	–
–52	2,55	–	2,64	0,03	2,79	0,15	2,83	–
–58	2,55	–	2,63	0,01	2,74	0,04	2,83	–
–60	2,55	–	2,62	–	2,72	0,02	2,83	–
–70	2,55	–	2,62	–	2,71	–	2,83	–
–80	2,55	–	2,62	–	2,71	–	2,83	–

Table 2

Temperature dependence of the dielectric constant ε' and dielectric loss ε" for the bromobenzene – n-hexane system, with bromobenzene concentrations in mole fractions: 1 – 0.300; 2 – 0.562; 3 – 0.794; 4 – 1.000

Bromobenzene – n-hexane								
t,^{0}C	x_2 = 0,300		x_2 = 0,562		x_2 = 0,794		x_2 = 1,000	
	ε'	ε''	ε'	ε''	ε'	ε''	ε'	ε''
Liquid state								
20	2,80	0,10	3,60	0,23	4,34	0,40	5,25	0,73
10	2,84	0,11	3,65	0,25	4,42	0,45	5,35	0,83
0	2,88	0,13	3,69	0,29	4,49	0,53	5,39	0,97
– 10	2,92	0,15	3,72	0,34	4,53	0,62	5,43	1,12
– 20	2,95	0,17	3,75	0,39	4,52	0,71	5,38	1,31

– 30	2,99	0,20	3,75	0,44	4,49	0,81	5,29	1,49
– 34	3,00	0,21	3,75	0,48	4,46	0,86	–	–
– 40	3,02	0,24	3,75	0,51	–	–	–	–
– 50	3,04	0,29	–	–	–	–	–	–
– 55	3,04	0,32	–	–	–	–	–	–
Solid state								
–31	–	–	–	–	–	–	3,12	0,31
–35	–	–	–	–	3,52	0,57	2,96	0,15
–38	–	–	–	–	3,19	0,32	2,87	0,08
–41	–	–	3,58	0,42	2,96	0,20	2,85	0,06
–45	–	–	3,02	0,21	2,75	0,11	2,84	0,01
–51	–	–	2,79	0,09	2,70	0,04	2,83	–
–53	–	–	2,66	0,05	2,67	0,01	2,83	–
–56	2,92	0,24	2,55	0,03	2,65	–	2,83	–
–58	2,77	0,14	2,52	0,01	2,65	–	2,83	–
–60	2,62	0,11	2,50	–	2,65	–	2,83	–
–65	2,45	0,04	2,50	–	2,65	–	2,83	–
–70	2,33	0,02	2,50	–	2,65	–	2,83	–
–80	2,32	–	2,50	–	2,65	–	2,83	–

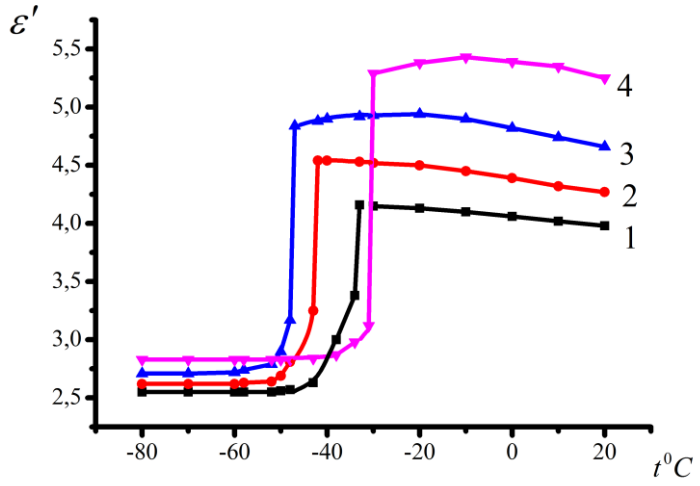


Fig. 1. Temperature dependence of the dielectric constant ε' for the bromobenzene – benzene system, at a bromobenzene concentration in mole fractions: 1 – 0.459; 2 – 0.560; 3 – 0.718; 4 – 1.000

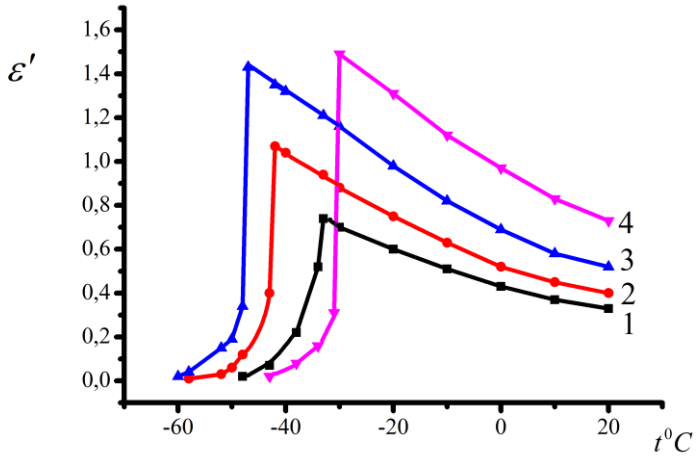


Fig. 2. Temperature dependence of dielectric losses ε" for the bromobenzene – benzene system, at bromobenzene concentration in mole fractions: 1 – 0.459; 2 – 0.560; 3 – 0.718; 4 – 1.000

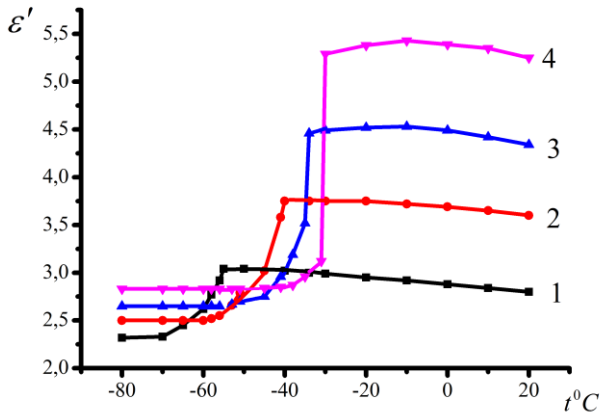


Fig. 3. Temperature dependence of the dielectric constant ε' for the bromobenzene – n-hexane system, at a bromobenzene concentration in mole fractions: 1 – 0.300; 2 – 0.562; 3 – 0.794; 4 – 1.000

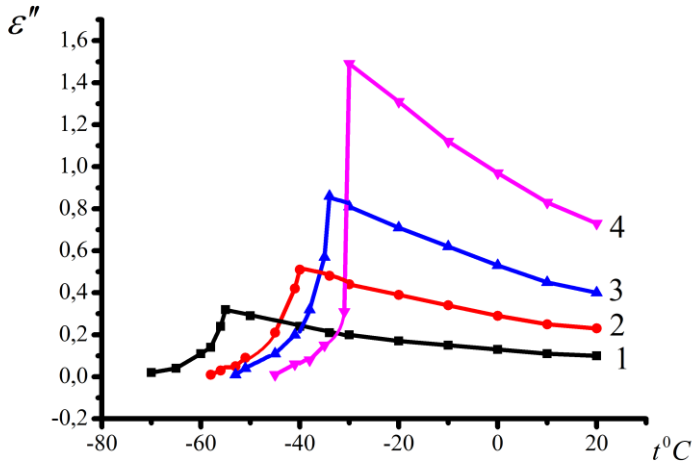


Fig. 4. Temperature dependence of dielectric losses ε" for the bromobenzene – n-hexane system, at bromobenzene concentration in mole fractions: 1 – 0.300; 2 – 0.562; 3 – 0.794; 4 – 1.000

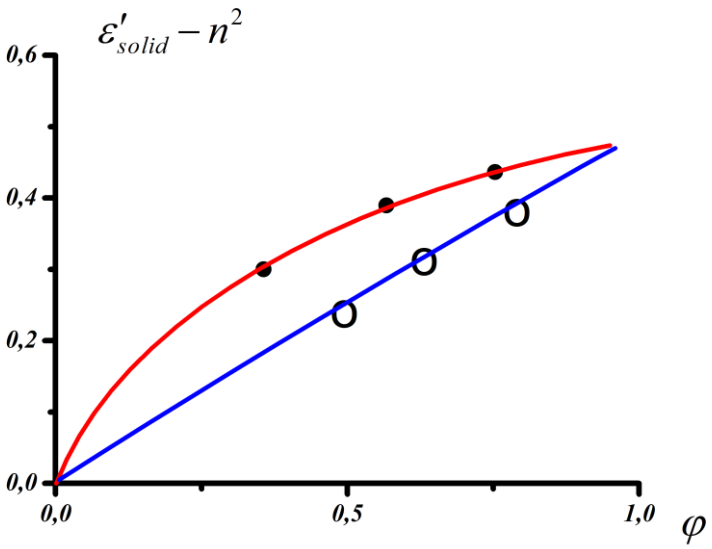


Fig. 5. Dependence of $\varepsilon_{solid}' - n^2$ for the bromobenzene – benzene (1) and bromobenzene – hexane (2) systems on the concentration expressed in mole fractions

Figures 1 – 4 show the temperature dependence of the dielectric constant for the bromobenzene–benzene and bromobenzene–hexane systems.

Thus, at these concentrations, the solid state is characterized by the presence of two phases – an intermediate metastable phase, which transitions to a stable low-temperature phase with further cooling.

When the temperature reaches approximately –60°C, the dielectric permittivity ceases to change in almost all cases, and the contribution of orientation polarization can be considered negligible. However, as can be seen in Fig. 5, the dielectric permittivity ε_{solid}' in the solid state greatly exceeds the corresponding value of the square of the refractive index n^2 at 20°C. Since the change in density with temperature is insignificant, the difference $\varepsilon_{solid}' - n^2$ should be explained by the contribution of infrared polarization.

It is interesting to note that the high-frequency dielectric permittivity ε_∞ at 20°C, obtained from the circular diagram, also exceeds the value n^2 at all concentrations [18-20].

On the other hand, ε_{solid}' is slightly greater than ε_∞. This confirms the assumption made in [21-23] that the difference between ε_∞ and n^2 is mainly due to additional resonance-type dispersion.

Interpreting dielectric relaxation as a kinetic process makes it possible to reveal new details of the mechanism of this phenomenon. The relaxation time is expressed by the Arrhenius equation:

$$\tau = \frac{h}{kT}\, exp\left(\Delta F/_{RT}\right) \qquad (1)$$

where ΔF is the free activation energy, defined as:

$$\Delta F = \Delta H - T\Delta S \qquad (2)$$

where ΔH is the activation enthalpy, ΔS is the activation entropy, h is Planck's constant, k is Boltzmann's constant, R is the universal gas constant, and T is the absolute temperature. The value of ΔH was calculated from the slope of the dependence of ln(Tτ) on 1/T, and ΔS was calculated using equation (2).

Table 3 shows the results of the corresponding calculations. It was assumed that ε_∞ in solution changes additively.

Table 3

The values of ΔF (free activation energy), ΔH (activation enthalpy), and TΔS (bound energy) for the bromobenzene–benzene system, at bromobenzene concentrations in mole fractions: 1 – 0.459; 2 – 0.560; 3 – 0.718; 4 – 1.000; and for the bromobenzene–hexane system, with bromobenzene concentrations in mole fractions: 1 – 0.294; 2 – 0.556; 3 – 0.745; 4 – 1.000

x_2	ΔF				$T\Delta S$				ΔH
	20^0	0^0	-20^0	-40^0	20^0	0^0	-20^0	-40^0	
Bromobenzene – benzene									
0,459	2,61	2,52	2,44	–	1,18	1,09	1,01	–	1,43
0,560	2,64	2,53	2,47	2,38	1,15	1,04	0,98	0,89	1,49
0,718	2,70	2,60	2,52	2,44	1,14	1,04	0,96	0,88	1,56
1,000	2,76	2,70	2,61	–	1,11	1,05	0,96	–	1,65
Bromobenzene - hexane									
0,294	2,36	2,26	2,16	2,08	1,30	1,20	1,10	1,02	1,06
0,556	2,49	2,38	2,31	2,23	1,24	1,13	1,06	0,98	1,25
0,745	2,60	2,50	2,43	–	1,18	1,08	1,01	–	1,42
1,000	2,76	2,70	2,61	–	1,11	1,05	0,96	–	1,65

4. CONCLUSIONS

The present study demonstrates that the dielectric relaxation of bromobenzene in binary systems is strongly governed by the nature of its molecular environment. A comparative analysis of the bromobenzene–benzene and bromobenzene–hexane systems, containing the same polar component, made it possible to distinguish the effect of solvent nature on the dynamics and thermodynamics of the relaxation process.

1. The temperature dependence of the relaxation time, considered within the framework of the absolute rate theory, provides a quantitative relationship between molecular dynamics and the thermodynamic activation parameters ΔF, ΔH, and ΔS. The negative values of ΔS indicate an increase in the degree of molecular ordering upon formation of the activated state.
2. The comparative analysis shows that the solvent environment modifies the energetic conditions for dipole reorientation of bromobenzene. In the investigated composition range, the bromobenzene–hexane system exhibits lower values of ΔF and ΔH upon dilution of bromobenzene compared with the bromobenzene–benzene system, indicating a difference in the relaxation energy barrier between the two media.
3. The observed differences establish a relationship between the relaxation characteristics and the

molecular properties of the solvent, as well as its influence on the local organization of the system. Therefore, the nature of the nonpolar solvent represents an independent factor governing both the energetic and structural components of dielectric relaxation.

4. The scientific novelty of this work lies in the direct comparison of the thermodynamics of relaxation of the same polar component in two different nonpolar media. The results demonstrate the potential of dielectric relaxation as a sensitive method for analyzing intermolecular interactions and the solvation environment in binary molecular systems.

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