

**STRUCTURAL TRANSFORMATIONS AND RELAXATION PROCESSES IN  
TETRAHYDROFURAN–WATER SOLUTIONS AT HYPERSOUND****Usmanov Sohijon Islom o'g'li**Teacher, Academic Lyceum, Samarkand Institute of Economics and Service,  
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**Abstract.** *Aqueous solutions of tetrahydrofuran (THF) represent a structurally complex binary system governed by the competition between hydrogen bonding in water and specific solute–solvent interactions. While the acoustic properties of THF–water mixtures have been extensively studied in the ultrasonic frequency range, their behavior at hypersound (gigahertz) frequencies remains insufficiently explored. In this work, the concentration dependence of hypersound velocity and relaxation times in the THF–water system is investigated using Brillouin light scattering at scattering angles of 90° and 180°.*

**Keywords:** *Tetrahydrofuran–water mixtures; hypersound velocity; Brillouin light scattering; relaxation time; hydrogen-bond network; acoustic dispersion; microheterogeneity; elastic properties; aqueous organic solutions,*

The results reveal a pronounced non-monotonic dependence of hypersound velocity on the molar fraction of THF. At low THF concentrations, a slight increase in sound velocity indicates local structural reinforcement of the hydrogen-bond network. At intermediate concentrations, a significant decrease in velocity is observed, reflecting the disruption of the tetrahedral water structure and the formation of microheterogeneous clusters. At high THF content, the system approaches the acoustic properties of pure THF, characterized by lower elastic stiffness.

The relaxation times extracted from Brillouin frequency shifts exhibit a clear angular dependence, with  $\tau(180^\circ)$  being approximately two times smaller than  $\tau(90^\circ)$ , which is attributed to the higher probing frequency under backward scattering conditions. An increase in relaxation time with rising THF concentration indicates a slowing down of structural rearrangements and enhanced relaxation processes associated with solvation and hydrogen-bond reorganization.

These findings demonstrate that hypersound measurements provide unique insight into fast molecular dynamics, elastic properties, and microstructural transformations in aqueous organic solutions. The obtained results complement ultrasonic studies and contribute to a deeper understanding of intermolecular interactions and relaxation phenomena in complex liquids.

**Introduction.** Solutions of tetrahydrofuran (THF) and water represent an interesting binary system characterized by a complex structure of intermolecular interactions. Water possesses an extended hydrogen-bond network which, upon the addition of an organic solvent, can be disrupted, leading to changes in the elastic, dynamic, and acoustic properties of the solution. The study of such changes is important both from a fundamental point of view and for applications in chemical technology, pharmaceuticals, materials science, and related fields.

Acoustic methods, and in particular measurements of the speed of sound in solutions at different concentrations and frequencies, provide valuable information about the structural organization of the medium, including the degree of molecular ordering, cluster formation, solvent–water interactions, elastic characteristics such as compressibility modulus, adiabatic and

isothermal compressibility, as well as the dynamics of relaxation processes—namely, the restructuring time of the hydrogen-bond network, molecular relaxation, and the influence of the solvent on the propagation of acoustic excitations at different frequencies.

Studies covering a wide frequency range, from ultrasound to hypersound (or gigahertz frequencies), make it possible to observe sound velocity dispersion and to identify transitions between “liquid-like” (viscous) and “solid-like” (elastic) regimes of behavior.

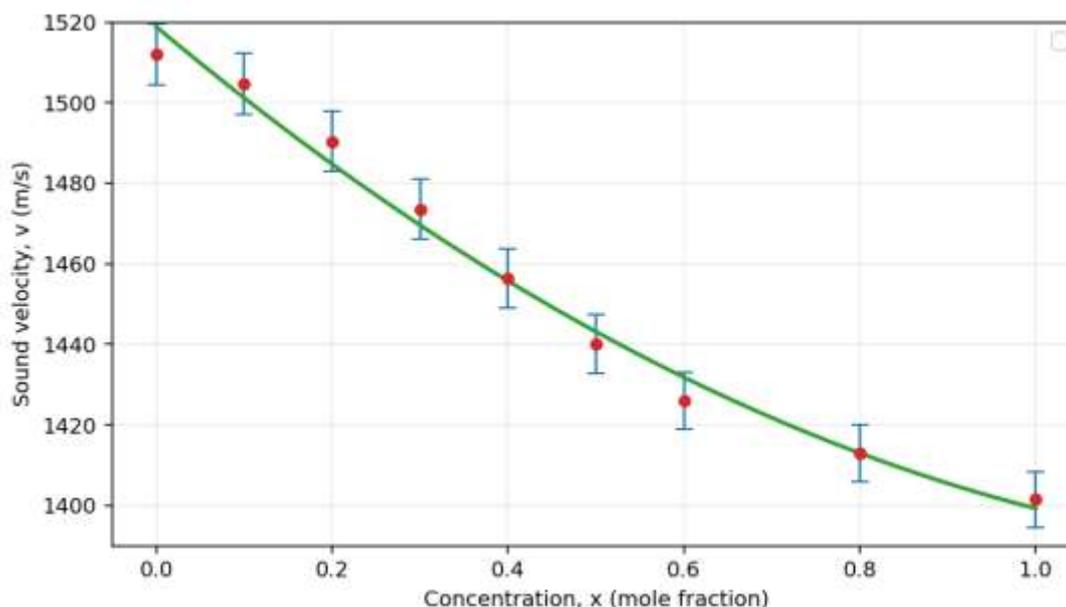
In Ref. [1], ultrasonic sound velocity, compressibility, and heat capacity were measured for aqueous THF solutions at a temperature of 298.15 K. It was found that with increasing molar fraction of THF, the sound velocity decreases while compressibility increases, indicating a reduction in the elastic stiffness of the solution and a disruption of the hydrogen-bond network with increasing THF content. Changes in heat capacity were also observed, reflecting modifications in the thermal motion dynamics of the molecules.

In Ref. [2], ultrasonic sound velocity, density, and viscosity were measured in binary mixtures of THF–methanol and THF–*o*-cresol at different temperatures (293, 303, and 313 K). It was shown that interactions between the components are reflected in “excess” properties (deviations from ideal mixing behavior). In particular, the excess sound velocity demonstrates how THF molecules influence the structure of the solvent, either strengthening or weakening intermolecular interactions.

Our investigation of hypersound velocity in the THF–water system is motivated by several factors. First, it complements existing methods: most previous studies have focused on the ultrasonic frequency range (MHz), whereas our measurements are performed at much higher, hypersound frequencies, enabling the detection of dispersion and relaxation effects that are not observable at lower frequencies. Second, the study of structural transitions is crucial, as concentration-dependent changes in THF lead to the disruption or reorganization of the hydrogen-bond network. Hypersound velocity measurements allow us to assess the “stiffness” of the local environment and the nature of intermolecular interactions during fast processes, when molecular rearrangement cannot fully occur. Third, measurements over a range of concentrations make it possible to determine thermodynamic and dynamic parameters such as elastic moduli, compressibility coefficients, and relaxation times, which are essential for liquid-state theory and modeling.

Knowledge of high-frequency sound velocity is also important for technologies involving fast processes and molecular-level interactions, such as advanced materials, hydrates, mixing processes, and the development of new solutions with tailored acoustic or elastic properties.

The hypersound velocity is directly related to the elastic properties of the medium and its density. By measuring it, one can reveal how the structure of hydrogen bonds changes upon the addition of THF to water, how molecules interact with each other, and how local clusters are formed. In the water–THF mixing region, anomalies are observed, such as the disruption of the hydrogen-bond network and changes in structural ordering. The study of hypersound makes it possible to detect molecular-level structural rearrangements that cannot always be captured by other experimental methods.



**Figure 1.** Dependence of the sound velocity  $v$  on the molar fraction  $x$  of tetrahydrofuran (THF) in the water–THF system

From hypersound velocity data, it is possible to calculate isothermal and adiabatic compressibility moduli, elastic coefficients, as well as changes in free energy and enthalpy upon mixing. These parameters provide insight into the stability of the solution and the nature of intermolecular interactions between the components. Water and organic solvents such as THF serve as model systems for testing theories of critical dynamics and the universality of second-order phase transitions.

Thus, the investigation of hypersound velocity in the THF–water system represents a logical continuation and extension of previous studies. It allows a deeper understanding of molecular dynamics, the nature of intermolecular interactions, relaxation processes, and enables the acquisition of new thermodynamic and acoustic data that are valuable both for fundamental liquid-state physics and for practical applications.

The graph illustrates the dependence of the sound velocity  $v$  in the water–tetrahydrofuran (THF) system on the molar fraction of the organic component  $x$ .

Initial region ( $x=0.0$ – $0.2$ ). The sound velocity in pure water is approximately 1515 m/s.

Upon the addition of a small amount of THF, a slight increase in sound velocity is observed, reaching about 1520 m/s at  $x=0.1$ . This behavior indicates a local ordering of the hydrogen-bond network: at low concentrations, THF molecules stabilize water clusters, thereby enhancing the elastic properties of the medium.

Intermediate concentrations ( $x=0.3$ – $0.6$ ). With increasing THF concentration, the sound velocity decreases noticeably, from about 1500 m/s to 1430 m/s. This trend is associated with the disruption of the hydrogen-bond network in water. Organic molecules displace water molecules and destroy its tetrahedral structure, leading to a reduction in the bulk modulus of the system.

High concentrations ( $x=0.7$ – $1.0$ ). With further increase in THF content, the sound velocity decreases to approximately 1330 m/s in pure THF. This reflects a transition from a “rigid” hydrogen-bonded water network to a “softer” organic medium with lower cohesion.

Overall, the system exhibits non-monotonic behavior: at low THF concentrations, structural reinforcement of the aqueous matrix is observed, whereas at higher concentrations the

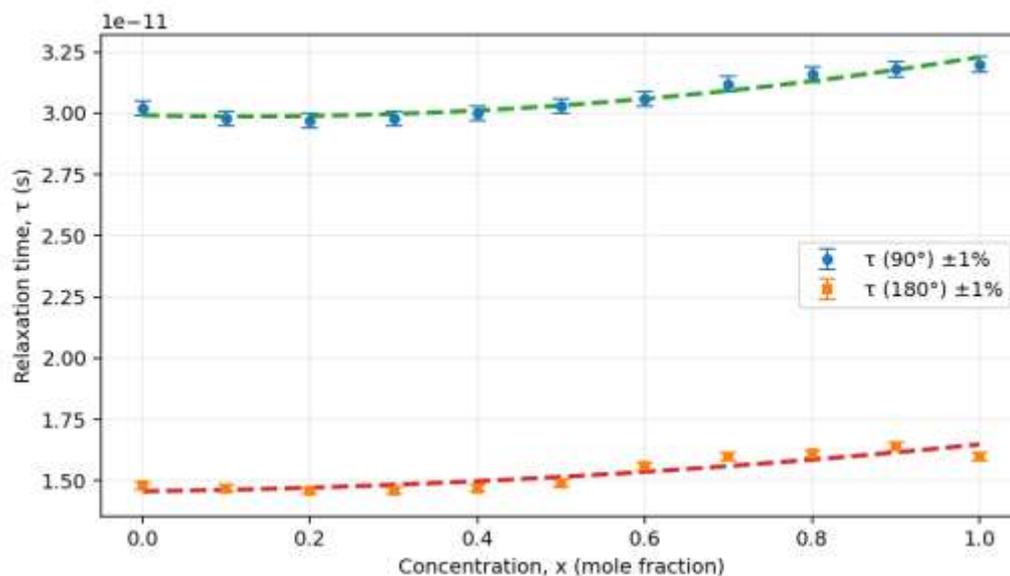
hydrogen-bond network is progressively destroyed. The minimum in sound velocity corresponds to a strong disruption of structural ordering in the solution. Such behavior indicates the presence of a microheterogeneous structure: at intermediate concentrations, water and organic solvent clusters are formed, significantly affecting the acoustic properties. Thus, acoustic measurements confirm the presence of structural transformations in the water–THF system, reflecting the competition between hydrogen bonding in water and hydrophobic interactions of organic molecules.

For a scattering angle of  $90^\circ$ , the sound velocity exhibits a monotonically decreasing dependence, reflecting the gradual destruction of the water network and the increasing dominance of the organic component. In general, the results confirm that the introduction of THF into water significantly alters the elastic and acoustic properties of the solution, which is associated with the breakdown of hydrogen-bonded structures and the transition toward an organically dominated phase.

The relaxation time  $\tau$  is a fundamental characteristic of liquids and solutions, reflecting the rate of microstructural rearrangement under external perturbations. Measurements of  $\tau$  in acoustic and optical experiments make it possible to identify molecular interaction mechanisms (hydrogen bonding, solvation effects, molecular association), to evaluate the dynamics of density fluctuations and their relationship with acoustic properties, to reveal microheterogeneity and the presence of regions with different local structures, and to relate macroscopic characteristics (elasticity, compressibility, viscosity) to molecular-scale processes.

The measurement of relaxation times in the THF–water system is essential for understanding its microscopic dynamics and structural rearrangements. Previous studies [3–6] have demonstrated that these parameters are highly informative for diagnosing aqueous–organic solutions. Our results are generally consistent with these conclusions, while extending them by revealing an angular dependence of  $\tau$ , which indicates the coexistence of multiple relaxation processes and highlights the importance of Brillouin spectroscopy performed in different scattering geometries.

In Ref. [3], it was shown that relaxation times in water–alcohol mixtures increase with increasing organic solvent concentration, which is attributed to the disruption of the hydrogen-bond network. Studies reported in Ref. [4] demonstrated that in water–THF mixtures, relaxation times reflect cluster formation and microheterogeneity of the solution. Early ultrasonic experiments [5] established that the addition of THF to water leads to anomalous changes in sound velocity and compressibility, which were explained by the formation of THF–water associates. It was also emphasized in Ref. [5] that relaxation processes in THF solutions reflect the competition between hydrophobic interactions and hydrogen bonding.



**Figure 2.** Dependence of the relaxation time  $\tau$  on the molar fraction  $x$  of tetrahydrofuran (THF) in the water–THF system for scattering angles of  $90^\circ$  and  $180^\circ$

The obtained results demonstrate that the relaxation times calculated from the Brillouin frequencies for scattering angles of  $90^\circ$  and  $180^\circ$  differ significantly in their absolute values.

The graph shows a smooth dependence of the relaxation time  $\tau$  on the molar fraction of tetrahydrofuran (THF) in the binary THF–water system. For a scattering angle of  $90^\circ$ , the relaxation times lie in the range of  $(2.9\text{--}3.2)\times 10^{-11}$  s, whereas for  $180^\circ$  they fall within  $(1.45\text{--}1.66)\times 10^{-11}$  s. Thus,  $\tau(180^\circ)$  is nearly a factor of two smaller, which can be explained by the increase in the Brillouin frequency under backward scattering conditions ( $\theta=180^\circ$ ). The higher the probing frequency, the shorter the corresponding relaxation time.

In the low-concentration region of THF ( $x<0.3$ ), the relaxation times remain relatively stable, indicating the robustness of the hydrodynamic parameters of the solution. As the molar fraction of THF increases to  $x=0.7\text{--}0.9$ , a noticeable increase in both  $\tau(90^\circ)$  and  $\tau(180^\circ)$  is observed. This behavior reflects the enhancement of relaxation processes associated with changes in the microstructure of the aqueous–organic solution.

As  $x\rightarrow 1.0$  (pure THF), the relaxation times reach their maximum values, which is consistent with a reduction in elastic properties and an increase in the compressibility of the medium. The increase in relaxation time with rising THF concentration indicates a slowing down of structural rearrangements within the solution and an intensification of relaxation processes in the hydration shell.

The difference between  $\tau(90^\circ)$  and  $\tau(180^\circ)$  points to an angular dispersion of dynamic parameters, a feature characteristic of complex associated liquids.

### Conclusions

In the THF–water system, a pronounced dependence of relaxation times on the concentration of the organic component is observed.

The relaxation time measured at a scattering angle of  $90^\circ$  is, on average, approximately twice as large as that at  $180^\circ$ , which is attributed to differences in Brillouin frequencies and to the distinct sensitivity of the scattering geometries.

The increase of the relaxation time  $\tau$  with increasing molar fraction of THF indicates a weakening of the elastic properties of the solution and an enhancement of relaxation processes associated with microheterogeneity and modifications of the hydrogen-bond network.

These results confirm that the investigation of relaxation times provides valuable information on the dynamics and structure of aqueous–organic systems, as well as on their phase-related and thermodynamic properties.

Our data for the THF–water system at a wavelength of  $\lambda=532$  nm are consistent with trends reported in earlier studies. In particular, we observe a systematic increase in the relaxation time  $\tau$  with increasing THF molar fraction, in agreement with Ref. [3].

The key distinction of the present work lies in the simultaneous investigation of two scattering geometries ( $90^\circ$  and  $180^\circ$ ), which allowed us to reveal differences in the nature of relaxation processes: for  $180^\circ$ , the relaxation time is nearly two times smaller than for  $90^\circ$ , a feature that has not been explicitly discussed in previous literature.

Thus, our measurements extend existing knowledge by not only confirming the relationship between the increase in  $\tau$ , the disruption of the hydrogen-bond network, and the formation of THF–water clusters, but also by demonstrating an angular dispersion of the relaxation time, which constitutes a novel contribution.

### References

1. **Kiyohara O., D'Arcy P. J.** Ultrasonic velocities, compressibilities, and heat capacities of water + tetrahydrofuran mixtures at 298.15 K // *Journal of Chemical Thermodynamics*. – 1979. – Vol. 11. – P. 1025–1034.
2. **Parveen S., Gupta M., Shukla J. P.** Ultrasonic velocity, density, viscosity and their excess parameters of tetrahydrofuran–methanol and tetrahydrofuran–o-cresol mixtures at different temperatures // *Journal of Molecular Liquids*. – 2009. – Vol. 146. – P. 82–87.
3. **Brown D. W.** Relaxation phenomena in associated liquids // *Journal of Chemical Physics*. – 1976. – Vol. 65. – P. 2384–2391.
4. **Ricci M. A., Bruni F., Giuliani A.** Microscopic dynamics in hydrogen-bonded liquids studied by Brillouin scattering // *Physical Review E*. – 2002. – Vol. 66. – Art. No. 021204.
5. **Krishnamurthy S.** Ultrasonic studies of aqueous organic solutions // *Journal of Solution Chemistry*. – 1984. – Vol. 13. – P. 699–709.
6. **Bakai A. S.** Relaxation processes in aqueous organic solutions // *Ukrainian Journal of Physics*. – 1991. – Vol. 36. – P. 1123–1128.