

Ultrasonic sound velocities, density, adiabatic compressibility, coefficient of thermal expansion of aqueous ethanol at various temperatures (atmospheric pressure)

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Abstract. *A two-layer system of miscible liquids is a common model situation in the study of both fundamental and applied problems in hydrodynamics related to stratified environments. Processes such as Rayleigh-Taylor instabilities or double diffusion can lead to the disruption of mechanical equilibrium. By examining the volumetric properties of solutions, it is possible to identify the mechanism of density profile instability development, the mechanism of molecule interaction in a static environment, and determine the properties of the emerging solvent-solute system in a dynamic process. In this work, a physical mechanism of the influence of key state parameters on intermolecular interaction at the molecular level in a complex system is proposed based on the study of environment parameters such as density, sound velocity, viscosity, as well as compressibility and expansion coefficients.*

Keywords. water-alcohol solution, properties, ultrasonic velocities, density, adiabatic compressibility, statistical mechanics

Mathematics Subject Classification (2010): 80A19

1 Introduction

Despite the vast number of works dedicated to water and aquatic systems, there is still no unified perspective on the interpretation of experimental data. This is due to the presence of hydrogen bonds alongside intermolecular interactions in water and aquatic systems, leading to collective thermal movements and associated structures. A water-alcohol solution serves as a standard representative of associated liquids. Sometimes they are referred to as mixtures since there is no dissociation of molecules in them. Alcohol molecules strongly influence the structure of water, leading to anomalies in many thermodynamic and transport properties of aqueous alcohol solutions. Alcohol molecules restructure the water's arrangement in the mixture. Studies of the mechanism and nature of intermolecular (structural) interactions, such as solvent-solute, solvent-solvent, and solute-solute, are essential to understanding the

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compressibility and expansion processes of solutions under the influence of pressure and temperature.

The application of static mechanics models to water-alcohol solutions becomes impractical due to the complex behaviour of the system. Therefore, empirical analysis and qualitative characteristics are used to describe the structure and properties.

Volumetric properties provide science and technology with crucial information about the physicochemical nature of anomalous properties in aqueous solutions. They offer insights into the structure and mechanism of intermolecular interactions between solvent and solute molecules, which come in various forms, sizes, and chemical natures. Key criteria for assessing liquid quality include density, viscosity-temperature properties, and chemical and physical stability.

Aqueous alcohol solutions serve as excellent solvents for electrolytes. The addition of electrolytes leads to strong electrostatic interactions (Columbic interactions), significantly altering the solution's structure and properties. Adding electrolytes to water-organic solutions affects all solution properties and the mechanism of molecule interaction during electrolyte dissolution in water. This process occurs in extractive distillation, liquid-liquid extraction, crystallization, equilibrium shifting in liquid-gas systems, and can be utilized to control salt precipitation and various biological processes. Additionally, it plays a role in energy and refrigeration systems. Knowledge of hydraulic laws is essential for solving practical problems in heat and gas supply, including the calculation of water supply systems, thermal networks, heat exchange equipment, pumps, and more.

Using the laws of statistical mechanics to predict changes in the properties and behaviour of solutions, as well as understanding the molecular-level mechanism, is challenging due to the complex nature of interactions between heterogeneous molecules. Accurate experimental data on volumetric properties are essential for a deep understanding at the microscopic level of the nature and physicochemical mechanism of anomalies in the properties of aqueous alcohol solutions. In this regard, partial molar volumes (\bar{V}_1, \bar{V}_2) are useful parameters for a profound understanding of the interaction between solvent and solute molecules.

The influence of adding alcohol to the solubility of salt in water is a very interesting scientific problem. For example, ethanol mixes with water in arbitrary ratios, and significant volume reduction of the mixture, up to several percent, is observed upon mixing compared to the initial combined volume of pure substances. For instance, when mixing 50 ml of ethanol with 50 ml of water, a 97 ml solution is formed. Mixing is also accompanied by some heating of the mixture. By manipulating the content of alcohol or electrolyte, it is possible to alter and control the properties of the ternary system and the crystallization process. Thermodynamics of heterogeneous media provides theoretical foundations necessary for understanding and calculating the movement of heterogeneous or multiphase mixtures in various situations [19]. Generalizing centuries of fluid mechanics science, it should be noted that fluid mechanics questions are relevant, and the thermophysical properties of liquids, depending on changes in state parameters, have a decisive influence on the structure and mechanics of a continuous medium. [1-3, 5, 8, 10, 12, 13, 15-18, 20, 21].

2 Experiment

The dependence of sound velocity on the change in liquid density induced by pressure forms the basis for reliable calculations of the adiabatic compressibility of the liquid, isothermal compressibility coefficient, thermal expansion coefficient, thermal pressure coefficient, enthalpy, isochoric heat capacity, isobaric heat capacity, partial derivative of enthalpy with respect to pressure, and partial derivative of internal energy.

In this study, experimental data on density and sound velocity for aqueous ethanol solutions at concentrations of 4 mass% and 21 mass%, at temperatures ranging from 278 to 333

K, and at atmospheric pressure were obtained using the DSA 5000 M automatic instrument by Anton Paar [1, 5].

The new generation density meters DMA 5000 provide high precision in measuring density and sound velocity and are user-friendly. Thanks to a density measurement accuracy of 0.000005 g/cm^3 , temperature control precision, and a minimal sample requirement of only 1 ml for measurement, DMA series density meters are highly accurate instruments with minimal sample consumption, suitable for metrology services and independent laboratories.

Experimental data and their analytical processing for aqueous ethanol solutions over a wide range of state parameter variations are presented in our work [1, 5]. Increasing pressure and temperature induce changes in various characteristics and mechanical properties of solutions, such as viscosity, density, refractive indices, etc. The influence of pressure on the rate constants and equilibrium of chemical reactions, as well as the calculation of activation volumes, can be performed, for example, using the Tate equation. Another objective of the present study was to calculate other key derived thermodynamic properties such as adiabatic coefficient of bulk compressibility, coefficient of thermal expansion, thermal pressure coefficient, isothermal coefficient of bulk compressibility, isochoric and isobaric heat capacities, enthalpy and enthalpy difference, partial pressure derivative of enthalpy, entropy, and partial derivatives of internal energy (internal pressure) based on the measured density and speed sound and the reported thermal pressure coefficient data. The measured density and speed of sound data as a function of temperature together with reported data at atmospheric pressure were used to develop wide - ranged reference correlation equations. The reference correlation equation for density $\rho_0(T)$ can be used to develop Tait type equation of state to predict high -pressure *PVT* data as

$$\frac{\rho - \rho_0}{\rho_0} = c \ln \left(\frac{B + P}{B + P_0} \right), \quad (2.1)$$

where $\rho_0(T)$ is the temperature dependence of the density as atmospheric pressure

$P_0=101.15 \text{ kPa}$. Also, other derived caloric properties (C_V, C_P) can be used as a reference data for calculation of other thermodynamic properties from *PVT* equation of state at high pressures, since the calculations are based on the well-known thermodynamic relations by integrating.

3 Results and Discussion

Mathematical models employed for solving diverse problems in physics, chemistry, and technology within continuous media often incorporate simplifying hypotheses and empirical parameters. Utilizing the general conservation equations of heterogeneous multi-component mixtures, a mathematical model for an equilibrium two-phase mixture can be developed. This model exhibits the hyperbolicity characteristic of equilibrium two-phase mixtures. The hyperbolic nature of this mathematical model dictates that the speed of disturbance propagation (sound velocity) in the mixture is a finite quantity. This aspect is critical for analyzing processes involving the initiation of shock waves in the detonation regime within dynamic mechanics. The assumption of mixture equilibrium significantly streamlines the overall mathematical model of heterogeneous multicomponent mixtures for detonation initiation calculations. The system of conservation laws in the equilibrium mathematical model of the two-phase mixture can be simplified to a system of conservation laws for the mixture, where the closing equations consist of equations of state for specific internal energy and pressure of phases, as well as typical relationships for heterogeneous mixtures.

The practical application of determining the sound velocity of solutions is noteworthy. For instance, knowledge of the sound velocity in a medium allows the determination of the concentration and density of the solution in the feedwater of steam boilers.

In this study, thermodynamic properties of two-component solutions were explored experimentally and analytically, providing insights into the mechanical properties of the medium. Liquid compressibility reflects the balance between intermolecular attraction and repulsion energies. This property significantly influences the changes in partial molar volume of compounds in different environments, alterations in viscosity under increased pressure, and aids in refining the reasons for changes in activation volume, reaction volume, and the pressure-accelerated reaction effect [4, 7]. To assess the compressibility of substances over a wide range of pressures, equations of state expressing the relationship between pressure (P), volume (V), and temperature (T) were employed. The compressibility coefficient can be directly determined from volume changes under pressure, acoustic measurements of elastic wave propagation speed in the substance, and shock compression experiments, establishing a relationship between density (ρ) and pressure at maximum experimentally obtained pressures. Compressibility is also determined from measurements of crystal lattice parameters under pressure using X -ray structural analysis.

Thermal expansion coefficients, isothermal compressibility coefficients, and the thermal pressure coefficient were calculated based on experimental data on PVT properties for aqueous ethanol solutions using the least squares method. It is known that if a deformation is created in a certain portion of an elastic medium (e.g., compressing or stretching the medium), then due to elasticity and inertia, the disturbance of the medium will be transmitted to neighbouring sections and propagate at a certain speed. Such propagating disturbances are called acoustic waves.

This task is dedicated to measuring the speed of acoustic waves in a water-alcohol mixture while varying temperature and concentrations to determine important qualitative characteristics of the medium. The propagation of a sound wave induces compression and rarefaction in the medium, accompanied by local changes in temperature. This process can be considered reversible and described by the adiabatic equation of state and the Tate equation, where equilibrium values of density, pressure, and internal pressure are used. The values of the adiabatic compressibility coefficient and the thermal expansion coefficient were determined using known relationships (3.1) and (3.3).

Measured values of density and sound velocity for natural geothermal fluids can be used to calculate thermodynamic parameters, such as β_T , α_P , γ_V , ΔH , C_P , C_V , $\left(\frac{\partial H}{\partial P}\right)_T$.

Adiabatic compressibility coefficient:

$$\beta_S = \frac{1}{W^2 \rho}, \quad (3.1)$$

or

$$\beta_S = -\frac{1}{V} \left(\frac{\partial V}{\partial P} \right)_S$$

was calculated using measured densities (ρ) and sound velocity data (W) from Tables 1 and 2 for various concentrations.

Thermal (heat) expansion coefficient:

$$\alpha_P = -\frac{1}{\rho} \left(\frac{\partial \rho}{\partial P} \right)_P, \quad (3.2)$$

calculated using measured densities (ρ) as a function of temperature from Tables 1 and 2 for various concentrations.

As is known, liquids are elastic bodies and, under certain assumptions, follow Hooke's law. The compressibility of a liquid is characterized by the coefficient of relative volume compression β_S , reflecting the balance of energies of intermolecular attraction and repulsion. There is a need for experimental determination of this characteristic through other quantities. This property of the liquid significantly determines the change in partial molar

volume of compounds in different environments, the magnitude of electrostriction in various electrolyte solutions, changes in dielectric permittivity under increased pressure, refractive index, viscosity, and also helps refine the reasons for volume changes under pressure. The quantity inverse to β_S is called the bulk modulus of elasticity of the liquid under isotropic compression E .

Reliable data on the compressibility of liquids over a wide range of state parameters are insufficient. The dependence of the speed of sound on changes in the density of a liquid induced by pressure allows providing reliable data for calculating the adiabatic compressibility of the liquid. However, using these data to calculate the isothermal compressibility at given values of pressure and temperature requires knowledge of the heat capacities under these conditions. Small isothermal expansion does not disrupt all intermolecular interactions but affects only the weakest van der Waals interactions. In these studies, the values of the volumetric deformation of the working vessels are important.

Tables 1 and 2 present experimental data on density, sound velocity, as well as calculated values of the thermal expansion coefficient (α_p, K^{-1}) and adiabatic compressibility coefficient ($\beta_S \cdot 10^{-10}, Pa^{-1}$) for the $C_2H_5OH + H_2O$ mixture at concentrations $x = 4$ mass.% and $x = 21$ mass.% (ethanol) at atmospheric pressure.

Based on the data obtained from the conducted experiment, a Lagrange polynomial function $L(x)$ - has been constructed to represent the dependence of the mixture density on changes in temperature at constant atmospheric pressure. A Lagrange polynomial is a polynomial that passes through all points of a given data set. Its form depends on the number and values of these points. To construct a Lagrange polynomial, knowledge of the values of the function (or data) at specified points is required. In such problems, it is typically necessary to interpolate the value of an unknown function corresponding to a certain x using the Lagrange interpolation polynomial formula derived from a known set of points with values of the unknown function ($x, f(x)$). For computational convenience, we used the computer program <https://planetcalc.ru/8692/> - Lagrange interpolation polynomial.

The obtained function allows analysing the dynamics of processes occurring in the thermodynamic system. For the mixture water + ethanol ($x=21$ mass. % ethanol), this function is the Lagrange polynomial:

$$\begin{aligned} \rho(T) = & -\frac{36384117436897}{1610905288244401800000} T^6 + \frac{145612105185512803}{3579789529432004000000} T^5 - \\ & -\frac{1964519224797623854861}{64436211529776072000000} T^4 + \frac{5229368774378795851099109}{429574743531840480000000} T^3 - \\ & -\frac{28155745108298371226013039037}{10309793844764171520000000} T^2 \\ & +\frac{560817298072452003505485171218677}{1718298974127361920000000000} T - \\ & \frac{80989212448835206073595437379289}{4989253699556800000000000} \end{aligned} \quad (3.3)$$

For a mixture of water + ethanol ($x = 4$ mass. % ethanol), this function is a Lagrange polynomial:

$$\begin{aligned} \rho(T) = & \frac{899170231915278203433055777287901}{5294775462967169960094064747071436977000} T^6 - \\ & -\frac{166332558018129652040888014143126913}{5402832105068540775606188517419833650000} T^5 + \\ & +\frac{615209291628250832245230987137530451927969}{26473877314835849800470323735357184885000000} T^4 - \end{aligned}$$

$$\begin{aligned}
& - \frac{2474997811366706759107664442023893842337621969}{26473877314835849800470323735357184885000000} T^3 + \\
& + \frac{62799552790732810479252724140505581307175932289}{29704210170923814342883953700260516000000000} T^2 - \\
& - \frac{53967883436036106821642124699291320260704715660089797}{21179101851868679840376258988285747908000000000} T + \\
& + \frac{304253130836101228338398867161209088331969974326293}{2378941972007346035083160800864200000000000} \quad (3.4)
\end{aligned}$$

The function used to calculate the coefficient of thermal expansion of the mixture includes the derivative of the density function with respect to temperature (T), as given by equation (3.5):

$$\begin{aligned}
\frac{\partial \rho}{\partial T} = & -1.35516785 \cdot 10^{-7} \cdot T^5 + 2.03380819 \cdot 10^{-4} \cdot T^4 - 0.1219512556 \cdot T^3 + \\
& + 36.52008541 \cdot T^2 - 5461.941438 \cdot T + 3263798.3476 \quad (3.5)
\end{aligned}$$

Table 1. Experimental data and calculated values of the coefficient of thermal expansion (α_p, K^{-1}) and the adiabatic compressibility coefficient ($\beta_s \cdot 10^{-10}, Pa^{-1}$) for the $C_2H_5OH+H_2O$ mixture, $C_2H_5OH + H_2O, x = 21$ mass %, (ethanol) at atmospheric pressure

T, K	$\rho, kg \cdot m^{-3}$	W, mc^{-1}	α_p, K^{-1}	$\beta_s \cdot 10^{-10}, Pa^{-1}$
278.8	981	1572.8	0.712116287	4.12
283.15	979	1577.18	0.772809303	4.11
293.15	976	1584.08	0.927016321	4.08
303.15	972	1588.02	1.106702485	4.08
313.15	966	1588.20	1.315997707	4.10
323.15	962	1584.78	1.552885386	4.14
333.15	956	1577.95	1.828830193	4.21

Table 2. Experimental data and calculated values of the coefficient of thermal expansion (α_p, K^{-1}) and the coefficient of adiabatic compressibility ($\beta_s \cdot 10^{-10}, Pa^{-1}$) for a mixture of $C_2H_5OH + H_2O, x = 4$ mass % (ethanol) at atmospheric pressure

T, K	$\rho, kg \cdot m^3$	W, mc^{-1}	α_p, K^{-1}	$\beta_s \cdot 10^{-10}, Pa^{-1}$
278.15	996.10	1454.42	0.058	4.75×10^{-10}
283.15	995.35	1472.9	0.060	4.63×10^{-10}
293.15	993.87	1503.62	0.066	4.45×10^{-10}
303.15	991.305	1526.06	0.072	4.33×10^{-10}
313.15	987.82	1382.87	0.078	5.29×10^{-10}
328.06	978.5	1399	0.0879	5.22×10^{-10}
333.08	977.27	1307.4	0.090	5.99×10^{-10}

As seen from the research results for two concentrations at atmospheric pressure, and previously at high state parameters [19], it can be concluded that with an increase in ethanol concentration to 25% by mass, a sufficient number of alcohol molecules disrupt the spatial network of hydrogen bonds characteristic of pure water. When alcohol molecules are introduced into the mixture, a restructuring of the hydrogen bond network occurs, i.e., alcohol molecules can no longer fit into the cavities of the spatial hydrogen network typical of

hydrogen bonds. Polar OH groups of ethanol replace water molecules, and the more extended hydrophobic groups of ethanol enter the cavities formed by water molecules. New compounds are formed – hydrates of the first type. At concentrations above 25% by mass, with an increase in temperature, hydrates of the second type are formed - water molecules form two or three hydrogen bonds. In hydrates of the second type, hydrogen bonds weaken compared to pure water. An increase in temperature also leads to a weakening of hydrogen bonds. This is valid for all investigated concentrations of water-alcohol solutions, where weak or broken hydrogen bonds are observed, and consequently, a decrease in the average hydrogen bond energy. With a decrease in temperature, the proportion of OH groups with strong hydrogen bonds increases [14].

As shown in the table, the coefficient of adiabatic expansion and thermal expansion for the aqueous ethanol solution is a positive quantity that increases with temperature. Based on the graph-analytical processing of the obtained data, it can be concluded that the rule of non-intersection of compression curves is satisfied. This corresponds to the fact that the initial compressibility determines the subsequent compressibility at higher pressures [11]. Since the compressibility of a liquid reflects the balance of attraction and repulsion energies, it is useful to compare the values calculated by different approaches. Knowing the density, viscosity, speed of sound in the system, as well as calculating the coefficient of adiabatic compressibility, thermal expansion, thermal pressure coefficient, one can determine the mechanism of intermolecular bonds in the studied system, find the energetic properties of the medium – activation enthalpy, isochoric heat capacity, isobaric heat capacity, enthalpy difference, partial enthalpy derivative with respect to pressure, and partial derivatives of internal energy with respect to volume [6, 9].

Fig. 1 and 2 graphically show the change in the coefficient of thermal expansion with temperature (300-440 K) and pressure (at 10 MPa and 40 MPa) based on our data at various concentrations of ethanol [1].

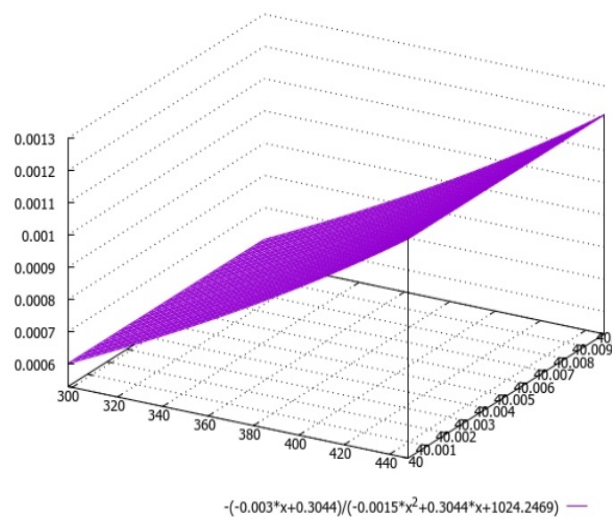


Fig. 1. Variation of the coefficient of thermal expansion with temperature (300-440 K) and pressure (40 MPa) based on our data at various ethanol concentrations

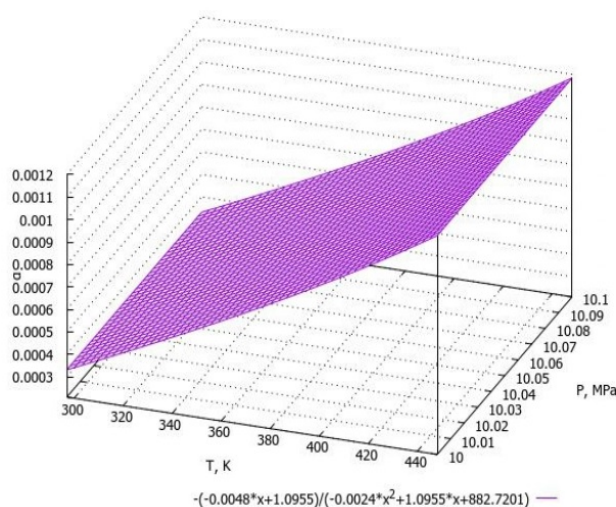


Fig. 2. Change in the coefficient of thermal expansion as a function of temperature (300-440K) and pressure of 10 MPa) according to our data at different ethanol concentrations

4 Conclusion

In this study, an attempt has been made to establish a connection between the values of density, viscosity, speed of sound in a medium, and a set of other physical characteristics of the liquid: the adiabatic compressibility coefficient, thermal expansion, knowing which one can predict the mechanism of intermolecular interaction in solutions, dynamics in the system under different environmental parameters.

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