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Full Length Article

Volumetric and ultrasonic properties of thiamine hydrochloride drug in aqueous solutions of choline-based deep eutectic solvents at different temperatures

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ABSTRACT

Important efforts have been made over the past years to improve the drug acts, which leads to the discovery of novel drug preparations and delivery systems. The optimal design of such processes requires a molecular-level understanding of the interactions between drug molecules and biological membranes. The thermodynamic investigation provides deep and complete knowledge of interactions and the choice of appropriate and suitable production compounds in pharmaceutical fields. Particularly, the analysis of drugs + co-solvents in aqueous media is the central issue in many types of research because they exert their impact by interacting with biological membranes. This work is aimed to measure the density and speed of sound for the thiamine hydrochloride in water + deep eutectic solvents (DESs) mixtures (choline chloride/urea, choline chloride/ethylene glycol and choline chloride/glycerol) at temperature range (293.15–308.15) K. By correlation of the evaluated parameters in some standard relations, the partial molar parameters *i.e.* apparent molar volumes, $V_{\varphi,m}$, and apparent molar isentropic compression, $\kappa_{s,\varphi,m}$, are calculated. In addition, apparent molar isobaric expansion, $E^0_{\varphi,m}$, and Hepler's constant are computed from the density and speed of sound data. For fitting the experimental $V_{\varphi,m}$ and $\kappa_{s,\varphi,m}$ the Redlich-Meyer equation was employed that the important quantities; standard partial molar volume, V^0_m, and partial molar isentropic compression, $\kappa^0_{s,m}$, were obtained. The thermodynamic analysis of the studied system also plays a crucial role in the pharmaceutical industry.

1. Introduction

Vitamins are one of the various organic materials applied by cells and organs to sustain their functions and developments. They take an important role in enzymic processes and genetic regulation processes [1]. Physicochemical interactions between the vitamins and chief biomolecules including amino acids, proteins, carbohydrates and lipids are key parameters to understand the pharmacodynamics and pharmacokinetics of these compounds and to develop the drug formulation in the pharmaceutical industry [2,3]. A comprehensive investigation of various thermophysical properties of the vitamins in the water and aqueous media of essential biomolecules can be applied to elucidate the representation of the different molecular interactions of the co-solutes with hydrophilic and hydrophobic moieties of the vitamins and to recognize the conformational stability of biomolecules in the biological system [4,5].

Vitamin B1 [VB1] (or thiamine/thiamin) is considered an essential vitamin and has been the subject of decades, and perhaps centuries, of research. The VB1 is a water-soluble vitamin and behaves as coenzyme for a variety of enzymes like transketolase, a-ketoglutarate dehydrogenase, pyruvate dehydrogenase, *etc.* It is engaged in the synthesis of proteins, metabolism of carbohydrates and fatty acids as well as in breakdown of polyhydroxy compounds inside biological systems [6].

Ionic liquids (ILs) have been employed as one of the appropriate alternatives for organic solvents in recent decades due to their low toxicity, non-flammability, insignificant vapor pressure, and low melting point [7,8]. However, concerns about environmental acceptability, sophisticated synthesis methods, high cost, and particular toxicity have limited their usage [9,10]. Deep eutectic solvents (DESs) are a neoteric generation of environmentally friendly solvents employed in different industries. These compounds frequently become liquid at room temperature and exhibit

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a lower melting point in comparison with the individual components [11]. DESs have been manufactured by combining a high melting point salt (*e.g.*, quaternary ammonium salt as a hydrogen bond acceptor (HBA) such as choline chloride (ChCl)) and the appropriate mole fraction of a hydrogen bond donor (HBD) compounds such as urea (U), glycerol (G), or ethylene glycol (EG) [12]. Generally, ILs and DESs are neoteric generation of green solvents in various fields but DESs have additional benefits including biodegradability and low cost [13]. Owing to these remarkable merits, DESs have been applied to liquid–liquid extraction, catalysis, electrochemistry, and material chemistry. Todays, these kinds of solvents also have many applications in the pharmaceutical industries [13–15]. Some investigations indicated that poorly soluble medicines were soluble in various DESs [16]. The knowledge of volumetric and acoustic of thiamine hydrochloride with DESs is valuable in elucidating the assorted interactions prevailing in their solutions and for improving the design of various processes.

The complex nature and the types of molecular interactions that occur in mixtures can be studied *via* physicochemical and thermodynamic investigations [17,18]. Thermodynamic properties are very useful for the understanding of the ionic, hydrophilic and hydrophobic interactions in different solutions media as they provide information elucidating the solute–solute and solute–solvent interactions in the solution phase. The volume and compressibility are two fundamental thermophysical properties that allow the deep consideration of interactions between the solute and solvent molecules in the mixtures [19,20]. The contributions of structurally similar vital DESs that affect the interactions of the thiamin and the around environments concerning temperature are limited. Consequently, in this research, the values of densities (d) and speeds of sound (u) of the thiamin hydrochloride (as vitamin B1 derivative) in water and DESs (ChCl/U, ChCl/G and ChCl/EG) aqueous solutions at T = (293.15–308.15) K in an interval of 5 K as a function of concentration are measured and reported. The obtained data has been applied for the calculation of numerous derived thermodynamic parameters. The derived thermophysical parameters including the apparent molar volume, $V_{\phi,m}$, standard partial molar volume, V_m^0, apparent molar isentropic compression, $\kappa_{s,\phi,m}$, and partial molar isentropic compression, $\kappa_{s,m}^0$. These findings can be helpful to predict the performance and behavior of solvents through the drug manufacturing processes.

2. Materials and Methods

2.1. Chemicals

The detailed descriptions of the used chemicals are specified in Table 1. To preparation of solutions double distilled deionized water was applied at 298.15 K.

2.2. Preparation of DESs

All of the DESs were prepared using an electronic balance with a precision of ±10^{-7} kg. The current uncertainty for the DES composition approaches, *i.e.* the mole ratio of the DESs' ingredient, was within ±4 × 10^{-3}. The DESs can be made with the specific mole ratio by stirring at 373.15 K and 0.1 MPa. A homogenous liquid was obtained after stirring for 60 min. Finally, the mixture was allowed to cool down to room temperature naturally for further use [21]. The water content of used DESs was analyzed by Karl-Fischer titration.

2.3. Apparatus and procedure

An analytical balance (AND, GF202, Japan) was used to weigh all the solutions with an uncertainty ±10^{-7} kg and then were kept inside the glass vials which were closed stoutly with parafilm. A vibrating tube densimeter (Anton Para, DSA 5000 densimeter and speed of sound analyzer) was used to measure density d and speed of sound u of prepared solutions. Also, dry air at atmospheric pressure and degassed and double distilled deionized water were used to calibration of apparatus. The temperature was kept within ±10^{-3} K. Uncertainty of density and speed of sound measurements were 0.15 kg·m^{-3} and 0.5 m·s^{-1}, respectively.

3. Results and Discussion

3.1. Volumetric properties

The experimental densities for thiamin in water and aqueous solutions of DESs (ChCl/EG, ChCl/G, and ChCl/U with concentration of (0.1, 0.3, and 0.5) mol·kg^{-1}) at T = (288.15, 298.15, 308.15 and 318.15 K) are listed in Table 2. By analyzing Table 2, it is noticed that values of densities increase monotonously with increase in concentration of DES, and at a fixed DES concentration these values decrease with increasing in temperature.

Apparent molar volumes (V_φ) could be calculated from the experimental values of density by using Eq. (1) [22,23]:

$$V_\varphi = \frac{M}{d} - \frac{d - d_0}{mdd_0} \quad (1)$$

where m and M are the thiamin molalities and molecular mass in the studied solutions, respectively. Also, d_0 and d, are the densities of the solvent and solutions. For the ternary systems the (DES + water) is considered as the solvent. The experimental values of apparent molar volume for thiamin in water and aqueous solutions of each DES at different temperatures are reported in Table 3. The plot of apparent molar volumes for thiamin in a fixed molality of each DES (0.3 mol·kg^{-1}) and at 298.15 K are shown in Fig. 1. From Table 3, it is implied that the apparent molar volumes increase as the concentration of DES increases.

Standard partial molar volume, V_φ^0, is obtained by using the following Redlich-Mayer equation [24]:

$$V_\varphi = V_\varphi^0 + S_v m^{1/2} + b_v m \quad (2)$$

where V_φ^0 is equal to the partial molar volume at infinite dilution which is called standard partial molar volume, and S_v and b_v are empirical parameters. Since solute–solute interactions at infinite

Table 1
Descriptions of the used chemicals

Material	Provenance	CAS no.	Purity (mass fraction)	Purification method	Water content① (mass fraction)
Choline chloride	Daejung	67-48-1	>0.99	Dried in vacuum over P_2O_5	0.0008
Urea	Merck	57-13-6	>0.995	Dried in vacuum over P_2O_5	0.0009
Ethylene glycol	Merck	107-21-1	>0.99	None	0.0020
Glycerol	Merck	56-81-5	>0.99	None	0.0020
Thiamine hydrochloride	Danna. Iran	67-03-8	>0.995	None	0.0001

① Determined by Karl-Fischer method.

Table 2
Density (d) and speed of sound (u) of thiamine hydrochloride in the aqueous solutions of deep eutectic solvents at different temperatures and pressure (P = 86.6 kPa)

m_1/mol·kg^{-1}	T = 288.15 K		T = 298.15 K		T = 308.15 K		T = 318.15 K	
	$d \times 10^{-3}$/kg·m^{-3}	u/m·s^{-1}	$d \times 10^{-3}$/kg·m^{-3}	u/m·s^{-1}	$d \times 10^{-3}$/kg·m^{-3}	u/m·s^{-1}	$d \times 10^{-3}$/kg·m^{-3}	u/m·s^{-1}
Thiamine hydrochloride + water								
m_2 = 0.000								
0.000	0.998994	1466.52	0.997693	1496.63	0.995534	1519.62	0.992402	1539.52
0.008	0.999873	1468.04	0.998512	1498.05	0.996316	1520.86	0.993151	1540.56
0.029	1.002170	1471.88	1.000650	1501.36	0.998356	1523.99	0.995100	1543.24
0.060	1.005407	1477.42	1.003688	1506.19	1.001245	1528.25	0.997869	1546.99
0.089	1.008541	1482.47	1.006611	1511.27	1.004047	1532.76	1.000600	1550.74
0.120	1.011753	1488.05	1.009677	1516.11	1.006979	1537.21	1.003408	1554.67
0.148	1.014822	1493.18	1.012557	1520.91	1.009740	1542.16	1.006091	1559.19
Thiamine hydrochloride + (ChCl/EG) + water								
m_2 = 0.100								
0.000	1.005766	1477.57	1.003236	1506.66	0.999438	1528.78	0.995507	1545.02
0.008	1.006601	1478.79	1.004026	1507.75	1.000192	1529.73	0.996225	1545.79
0.030	1.008761	1482.2	1.006091	1510.75	1.002132	1532.34	0.998066	1547.94
0.059	1.011392	1486.57	1.008540	1514.54	1.004441	1535.66	1.000266	1550.67
0.088	1.014099	1491.26	1.011061	1518.69	1.006772	1539.31	1.002503	1553.56
0.115	1.016374	1495.65	1.013153	1522.59	1.008698	1542.74	1.004330	1556.31
0.148	1.018917	1500.35	1.015457	1526.63	1.010834	1546.31	1.006248	1559.39
m_2 = 0.300								
0.000	1.008794	1481.17	1.006451	1510.07	1.002596	1532.02	0.998775	1547.83
0.010	1.009601	1482.22	1.007211	1511.00	1.003321	1532.79	0.999464	1548.49
0.030	1.011721	1485.12	1.009225	1513.41	1.005198	1534.88	1.001238	1550.27
0.060	1.014304	1488.90	1.011551	1516.81	1.007407	1537.78	1.003361	1552.73
0.090	1.016825	1493.17	1.013861	1520.21	1.009486	1540.66	1.005434	1555.29
0.120	1.018933	1497.06	1.015753	1523.66	1.011207	1543.55	1.007111	1557.84
0.150	1.021456	1501.58	1.018032	1527.38	1.013335	1546.74	1.008917	1560.37
m_2 = 0.500								
0.000	1.011821	1484.76	1.009665	1513.49	1.005753	1535.27	1.002042	1550.64
0.008	1.012601	1485.62	1.010397	1514.20	1.006449	1535.86	1.002703	1551.19
0.030	1.014632	1487.99	1.012279	1516.19	1.008234	1537.55	1.004383	1552.73
0.059	1.017105	1491.03	1.014532	1518.92	1.010344	1539.85	1.006416	1554.79
0.088	1.019551	1494.48	1.016721	1521.70	1.012323	1542.11	1.008334	1557.03
0.115	1.021562	1497.76	1.018462	1524.72	1.013946	1544.36	1.009893	1559.07
0.148	1.023854	1501.71	1.020578	1528.13	1.015836	1547.16	1.011587	1561.36
Thiamine hydrochloride + (ChCl/G) + water								
m_2 = 0.100								
0.000	1.008790	1481.16	1.006450	1510.07	1.002600	1532.02	0.998780	1547.84
0.008	1.009601	1482.22	1.007211	1511.00	1.003325	1532.78	0.999474	1548.49
0.030	1.011721	1485.12	1.009225	1513.41	1.005198	1534.88	1.001241	1550.27
0.059	1.014303	1488.89	1.011552	1516.80	1.007408	1537.77	1.003361	1552.73
0.088	1.016825	1493.16	1.013861	1520.21	1.009486	1540.66	1.005434	1555.29
0.115	1.018933	1497.06	1.015752	1523.66	1.011207	1543.55	1.007111	1557.84
0.148	1.021456	1501.58	1.018032	1527.38	1.013336	1546.74	1.008917	1560.37
m_2 = 0.300								
0.000	1.010307	1482.96	1.008058	1511.78	1.004174	1533.64	1.000409	1549.24
0.008	1.011101	1483.92	1.008804	1512.60	1.004885	1534.32	1.001084	1549.84
0.030	1.013177	1486.55	1.010752	1514.80	1.006716	1536.21	1.002811	1551.50
0.059	1.015704	1489.96	1.013042	1517.86	1.008876	1538.81	1.004888	1553.76
0.088	1.018188	1493.82	1.015291	1520.95	1.010904	1541.38	1.006884	1556.16
0.115	1.020247	1497.41	1.017107	1524.19	1.012577	1543.95	1.008502	1558.45
0.148	1.022655	1501.64	1.019305	1527.75	1.014586	1546.95	1.010252	1560.86
m_2 = 0.500								
0.000	1.013330	1486.55	1.011270	1515.20	1.007330	1536.89	1.003680	1552.05
0.008	1.014101	1487.30	1.011990	1515.83	1.008013	1537.43	1.004323	1552.58
0.030	1.016088	1489.42	1.013826	1517.58	1.009752	1538.88	1.005956	1553.96
0.059	1.018505	1492.09	1.016023	1519.97	1.011813	1540.88	1.007943	1555.82
0.088	1.020914	1495.13	1.018151	1522.44	1.013741	1542.83	1.009784	1557.9
0.115	1.022876	1498.11	1.019816	1525.10	1.015316	1544.76	1.011284	1559.68
0.150	1.025053	1501.77	1.021851	1528.20	1.017087	1547.37	1.012922	1561.85
Thiamine hydrochloride + (ChCl/Urea) + water								
m_2 = 0.100								
0.000	1.001225	1472.17	0.998414	1501.54	0.994701	1523.91	0.990606	1540.80
0.008	1.002101	1473.75	0.999248	1503.03	0.995499	1525.23	0.991366	1541.83
0.030	1.004322	1477.82	1.001390	1506.76	0.997533	1528.53	0.993308	1544.45
0.059	1.007024	1482.76	1.004024	1511.14	0.999992	1532.49	0.995624	1547.58
0.088	1.010010	1488.39	1.006861	1516.41	1.002701	1537.29	0.998107	1550.96

(continued on next page)

Table 2 (*continued*)

m_1/mol•kg^{-1}	T = 288.15 K		T = 298.15 K		T = 308.15 K		T = 318.15 K	
	$d\times10^{-3}$/kg·m^{-3}	u/m·s^{-1}	$d\times10^{-3}$/kg·m^{-3}	u/m·s^{-1}	$d\times10^{-3}$/kg·m^{-3}	u/m·s^{-1}	$d\times10^{-3}$/kg·m^{-3}	u/m·s^{-1}
0.115	1.012536	1493.24	1.009254	1520.99	1.004934	1541.53	1.000158	1554.02
0.150	1.015109	1498.50	1.011594	1525.51	1.007082	1545.67	1.002244	1557.92
m_2 = 0.300								
0.000	1.004253	1475.77	1.001629	1504.95	0.997859	1527.15	0.993874	1543.61
0.008	1.005101	1477.11	1.002433	1506.18	0.998628	1528.23	0.994605	1544.47
0.030	1.007282	1480.74	1.004524	1509.42	1.000599	1531.07	0.99648	1546.78
0.059	1.009936	1485.30	1.007035	1513.41	1.002958	1534.61	0.998719	1549.64
0.088	1.012736	1490.30	1.009661	1517.93	1.005415	1538.64	1.001038	1552.69
0.115	1.015095	1494.85	1.011854	1522.06	1.007443	1542.34	1.002939	1555.55
0.150	1.017648	1499.73	1.014169	1526.26	1.009583	1546.10	1.004913	1558.9
m_2 = 0.500								
0.000	1.007280	1479.36	1.004843	1508.37	1.001016	1530.40	0.997141	1546.42
0.008	1.008101	1480.47	1.005619	1509.33	1.001756	1531.23	997.844	1547.11
0.030	1.010241	1483.66	1.007658	1512.08	1.003665	1533.61	999.652	1549.1
0.059	1.012847	1487.83	1.010046	1515.68	1.005925	1536.72	1001.814	1551.7
0.088	1.015462	1492.21	1.012461	1519.46	1.008129	1539.99	1003.968	1554.43
0.115	1.017654	1496.45	1.014453	1523.12	1.009952	1543.15	1005.721	1557.08
0.150	1.020186	1500.96	1.016745	1527.01	1.012084	1546.52	1007.583	1559.89

Note: The standard uncertainties (u) for density, speed of sound, temperature, pressure, molality of thiamine and DES are $u(d)$ = 0.05 kg·m^{-3}; $u(u)$ = 0.5 m s^{-1}; $u(T)$ = 0.01 K; $u(p)$ = 1050 hPa; u(m) = 0.002, respectively. m_1 is the molal concentration of thiamine in the solution of DES + water. m_2 is the molal concentration of DES in water.

Table 3
The apparent molar volumes (V_φ) and apparent molar isentropic compressibility (k_φ) of thiamine hydrochloride in aqueous solutions of DESs at different temperatures and pressure (P = 86.6 kPa).

m_1/mol•kg^{-1}	T = 288.15 K		T = 298.15 K		T = 308.15 K		T = 318.15 K	
	$V_\varphi\times10^6$/ m^3·mol^{-1}	$k_\varphi\times10^{14}$/ m^3·mol^{-1}·Pa^{-1}	$V_\varphi\times10^6$/ m^3·mol^{-1}	$k_\varphi\times10^{14}$/ m^3·mol^{-1}·Pa^{-1}	$V_\varphi\times10^6$/ m^3·mol^{-1}	$k_\varphi\times10^{14}$/ m^3·mol^{-1}·Pa^{-1}	$V_\varphi\times10^6$/ m^3·mol^{-1}	$k_\varphi\times10^{14}$/ m^3·mol^{-1}·Pa^{-1}
Thiamine hydrochloride + water								
m_2 = 0.000								
0.000	229.38	−6.20	236.94	−4.36	241.82	−2.44	246.40	−0.60
0.008	228.60	−5.99	236.27	−3.61	241.22	−2.29	245.98	−0.58
0.029	228.33	−6.01	235.58	−3.62	240.72	−2.10	245.37	−0.55
0.060	228.13	−5.74	235.46	−3.83	240.39	−2.26	244.47	−0.65
0.089	227.80	−5.80	234.57	−3.81	239.48	−2.31	243.71	−0.75
0.120	227.04	−5.82	233.85	−3.90	238.70	−2.64	242.76	−1.16
0.148								
Thiamine hydrochloride + (ChCl/EG) + water								
m_2 = 0.100								
0.000	234.92	−2.95	240.69	−1.31	245.63	0.12	250.65	1.72
0.008	237.13	−3.00	242.09	−1.32	247.98	0.21	253.12	1.78
0.029	239.06	−2.89	244.93	−1.06	250.71	0.43	255.60	1.97
0.060	240.18	−2.84	246.34	−1.02	252.57	0.50	257.15	2.08
0.089	241.84	−2.80	248.28	−0.97	254.71	0.56	259.29	2.18
0.120	244.21	−2.42	251.00	−0.56	257.37	0.92	262.66	2.49
0.148								
m_2 = 0.300								
0.000	237.86	−1.32	243.83	0.17	248.65	1.66	253.65	2.75
0.008	238.93	−1.38	244.26	0.38	250.51	1.78	255.75	2.91
0.029	240.59	−1.33	247.91	0.40	253.48	1.80	258.03	2.92
0.060	243.15	−1.32	250.54	0.65	257.12	2.13	260.46	3.07
0.089	245.46	−1.24	253.13	0.69	259.88	2.23	263.02	3.15
0.120	247.08	−1.11	254.85	0.87	261.35	2.37	266.22	3.51
0.148								
m_2 = 0.500								
0.000	240.64	0.40	246.68	2.01	251.63	3.17	256.49	3.75
0.008	242.24	0.43	248.96	2.05	253.95	3.17	259.21	3.86
0.029	243.95	0.54	251.33	2.04	256.70	3.23	261.09	3.90
0.060	246.06	0.56	253.99	2.25	260.19	3.56	264.06	4.06
0.089	248.43	0.60	256.97	2.24	262.96	3.70	266.69	4.23
0.120	250.86	0.69	258.85	2.30	265.27	3.79	269.73	4.49
0.148								
Thiamine hydrochloride + (ChCl/G) + water								
m_2 = 0.100								
0.000	237.38	−1.44	243.71	0.16	248.65	1.73	253.04	2.77
0.008	238.80	−1.41	244.23	0.37	250.64	1.79	255.82	2.94
0.029	240.54	−1.34	247.87	0.41	253.53	1.81	258.11	2.93

(*continued on next page*)

Table 3 (*continued*)

m_1/mol·kg^{-1}	T = 288.15 K		T = 298.15 K		T = 308.15 K		T = 318.15 K	
	$V_\varphi\times10^6$/ m^3·mol^{-1}	$k_\varphi\times10^{14}$/ m^3·mol^{-1}·Pa^{-1}	$V_\varphi\times10^6$/ m^3·mol^{-1}	$k_\varphi\times10^{14}$/ m^3·mol^{-1}·Pa^{-1}	$V_\varphi\times10^6$/ m^3·mol^{-1}	$k_\varphi\times10^{14}$/ m^3·mol^{-1}·Pa^{-1}	$V_\varphi\times10^6$/ m^3·mol^{-1}	$k_\varphi\times10^{14}$/ m^3·mol^{-1}·Pa^{-1}
0.060	243.10	−1.33	250.52	0.65	257.16	2.13	260.52	3.08
0.089	245.43	−1.25	253.13	0.69	259.91	2.23	263.07	3.15
0.120	247.05	−1.12	254.85	0.87	261.37	2.37	266.26	3.51
0.148								
m_2 = 0.300								
0.000	239.20	−0.50	245.26	1.09	250.08	2.41	255.08	3.29
0.008	240.56	−0.47	246.62	1.22	252.22	2.48	257.49	3.40
0.029	242.28	−0.39	249.62	1.23	255.08	2.51	259.58	3.42
0.060	244.60	−0.38	252.27	1.46	258.66	2.85	262.27	3.58
0.089	246.95	−0.32	255.06	1.47	261.42	2.97	264.87	3.69
0.120	248.97	−0.20	256.86	1.59	263.31	3.08	267.99	4.01
0.148								
m_2 = 0.500								
0.000	241.49	1.30	247.86	2.68	252.93	3.63	258.38	4.05
0.008	243.73	1.30	250.58	2.81	255.61	3.85	261.05	4.35
0.029	245.55	1.46	252.99	2.84	258.25	3.92	262.68	4.40
0.060	247.46	1.48	255.68	3.04	261.70	4.26	265.9	4.56
0.089	249.86	1.49	258.86	3.08	264.47	4.42	268.55	4.77
0.120	252.70	1.58	260.81	3.11	267.20	4.49	271.49	4.99
0.148								
Thiamine hydrochloride + (ChCl/Urea) + water								
m_2 = 0.100								
0.000	232.18	−5.93	237.58	−4.45	242.45	−2.70	247.69	−0.28
0.008	234.29	−5.55	238.68	−3.98	244.02	−2.24	249.02	−0.00
0.029	236.16	−5.07	239.83	−3.43	245.94	−1.79	251.40	0.41
0.060	237.63	−4.79	241.83	−3.24	247.45	−1.71	253.81	0.83
0.089	239.08	−4.50	243.53	−3.03	249.35	−1.55	255.98	1.07
0.120	240.39	−4.33	245.65	−2.67	251.79	−1.22	257.70	1.00
0.148								
m_2 = 0.300								
0.000	233.55	−4.03	239.23	−2.49	244.04	−0.95	249.32	0.98
0.008	236.23	−3.83	241.01	−2.20	246.71	−0.60	251.82	1.19
0.029	238.30	−3.57	243.42	−1.81	249.30	−0.28	254.38	1.48
0.060	238.70	−3.61	244.22	−1.87	250.28	−0.34	255.49	1.58
0.089	240.02	−3.55	245.83	−1.82	252.10	−0.29	257.42	1.69
0.120	242.77	−3.08	249.07	−1.29	255.36	0.18	260.87	1.97
0.148								
m_2 = 0.500								
0.000	236.39	−1.95	242.15	−0.23	247.08	1.09	252.22	2.41
0.008	238.03	−2.20	243.16	−0.46	249.22	1.00	254.44	2.35
0.029	239.84	−2.21	246.41	−0.33	252.06	1.12	256.81	2.44
0.060	241.67	−2.08	248.44	−0.19	254.84	1.32	258.82	2.58
0.089	243.65	−2.07	250.71	−0.13	257.30	1.40	261.16	2.66
0.120	245.65	−1.76	252.93	0.16	259.36	1.65	264.45	3.00
0.148								

Note: The standard uncertainties (u) for density, speed of sound, temperature, pressure, molality of thiamine and DES are $u(d)$ = 0.05 kg·m^{-3}; $u(u)$ = 0.5 m·s^{-1}; $u(T)$ = 0.01 K; $u(p)$ = 1050 hPa; $u(m)$ = 0.002, respectively. The uncertainty for apparent molar volumes and apparent molar isentropic compressibility are $u(V)$ = 0.06 × 10^{-6} m^3·mol^{-1} and $u_c(k_\varphi)$ = 0.02 × 10^{-14} m^3·mol·Pa^{-1}, respectively. m_1 is the molal concentration of thiamine hydrochloride in the solution of DES + water. m_2 is the molal concentration of DES in water.

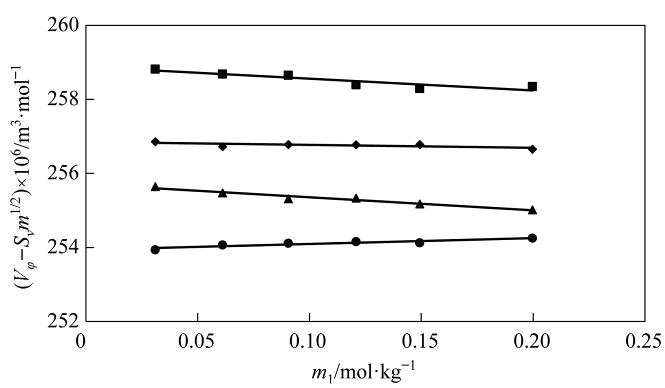

Fig. 1. Apparent molar volume of thiamine hydrochloride, V_φ, *versus* its molality, m_1, in aqueous DESs solution at m_2 = 0.3 mol·kg^{-1} at T = 298.15 K: ●, (Thiamine + water); ■, (Thiamine + (ChCl/Urea) + water); ▲, (Thiamine + (ChCl/EG) + water); ◆, (Thiamine + (ChCl/G) + water).

dilution are negligible, important information on solute–solvent interactions is provided by standard partial molar volumes. The values of V_φ^0, S_v and b_v together with its standards deviation of the V_φ^0 values are reported in Table 4. It is noteworthy that the all values of V_φ^0 as a criterion of the solute–solvent interactions are positive and increase with an increase in each DES concentrations and temperature. This is due to the low electrostriction of water and strong interactions between solute and solvent molecules. The larger valuesat high temperature and in presence of DES are probably refers to the release of the solvent molecules to the bulk. The positive b_v values are indicative of dominance of hydrophilic hydrogen-bonding interactions over hydrophobic interactions [25–28].

Temperature dependence of V_φ^0 values can be defined by following equation:

$$V_\varphi^0 = A + BT + CT^2 \quad (3)$$

Table 4
Standard partial molar volumes (V_φ^0), adjustable parameters of Eq. 2 (S_v and B_v), transfer volume ($\Delta_{tr}V_\varphi^0$), and standard deviations ($\sigma(V_\varphi^0)$) for thiamine in aqueous solutions of DESs at different temperatures

T/K	$V_\varphi^0 \times 10^6$/m^3·mol^{-1}	$S_v \times 10^6$/m^3·mol^{-2}·kg	$B_v \times 10^6$/m^3·mol^{-2}·kg	$\Delta_{tr}V_\varphi^0 \times 10^6$/m^3·mol^{-1}	$\sigma(V_\varphi^0) \times 10^6$/m^3·mol^{-1}
Thiamine hydrochloride + water					
m_2 = 0.000 mol·kg^{-1}					
288.15	229.63	−3.41	−7.23	–	0.18
298.15	237.11	−1.01	−18.61	–	0.16
308.15	241.7	2.86	−26.98	–	0.14
318.15	246.17	5.59	−37.31	–	0.05
Thiamine hydrochloride + (ChCl/EG) + water					
m_2 = 0.100 mol·kg^{-1}					
288.15	233.70	11.48	39.22	4.07	0.29
298.15	239.94	1.70	69.35	2.83	0.28
308.15	244.00	13.31	54.82	2.30	0.17
318.15	249.72	5.83	69.57	3.55	0.37
m_2 = 0.300 mol·kg^{-1}					
288.15	237.78	−7.85	85.57	8.15	0.28
298.15	242.86	−1.98	90.00	5.75	0.57
308.15	246.63	12.78	70.63	4.93	0.52
318.15	253.37	−4.67	98.24	7.20	0.14
m_2 = 0.500 mol·kg^{-1}					
288.15	240.68	−7.60	88.58	11.05	0.13
298.15	245.31	8.31	71.89	8.20	0.27
308.15	250.02	9.10	81.76	8.32	0.28
318.15	255.79	1.21	90.71	9.62	0.24
Thiamine hydrochloride + (ChCl/G) + water					
m_2 = 0.100 mol·kg^{-1}					
288.15	236.88	−2.10	76.35	7.25	0.26
298.15	242.65	−0.75	88.17	5.54	0.56
308.15	246.54	14.21	67.49	4.84	0.51
318.15	252.02	5.79	79.89	5.85	0.22
m_2 = 0.300 mol·kg^{-1}					
288.15	239.12	−7.02	86.01	9.49	0.16
298.15	244.09	3.10	81.10	6.98	0.36
308.15	248.22	11.62	75.11	6.52	0.40
318.15	254.57	−1.52	94.07	8.40	0.17
m_2 = 0.500 mol·kg^{-1}					
288.15	241.12	−1.27	80.28	11.49	0.25
298.15	246.17	12.67	67.79	9.06	0.29
308.15	251.37	9.35	84.00	9.67	0.18
318.15	257.86	−1.31	95.54	11.69	0.29
Thiamine hydrochloride + (ChCl/Urea) + water					
m_2 = 0.100 mol·kg^{-1}					
288.15	230.12	21.52	12.95	0.49	0.07
298.15	238.05	−11.64	80.76	0.94	0.12
308.15	242.11	−1.36	67.01	0.41	0.22
318.15	246.73	3.00	67.36	0.56	0.18
m_2 = 0.300 mol·kg^{-1}					
288.15	231.87	19.67	18.46	2.24	0.54
298.15	238.70	1.82	61.90	1.59	0.48
308.15	242.56	14.45	45.40	0.86	0.49
318.15	248.25	8.61	59.34	2.08	0.48
m_2 = 0.500 mol·kg^{-1}					
288.15	235.74	1.93	62.16	6.11	0.08
298.15	241.17	1.39	77.16	4.06	0.32
308.15	245.24	13.34	62.53	3.54	0.21
318.15	251.68	−0.36	85.52	5.51	0.24

where A, B and C are empirical constants which are calculated by the least-square fitting of standard partial molar volume at studied temperatures [29,30]:

On the basis of the derivative of standard partial molar volume V_φ^0 from Eq. (3) as function of temperature and at constant pressure, the standard apparent molar expansibilities E_φ^0 were computed which are given in Table 5. The values of E_φ^0 are positive for the studied drug in aqueous DESs solutions. Positive expansibility is a characteristic property of aqueous solutions of hydrophobic hydration. This would increase the solution volume a little more rapidly than that of the pure water and so E_φ^0 would be positive.

The E_φ^0 values are positive and increase with increasing in concentration of DES and temperature. This may indicate that systems are sensitive to temperature and molecular motilities are increased at higher temperature [31,32].

The thermal expansion coefficient, α, was calculated by the values of the standard partial molar volume, using Eq. (4) [33]:

Table 5
Standard apparent molar expansibility, E_φ^0, coefficient of thermal expansion, α, Hepler's constant expansion, $(\partial^2 V_\varphi^0/\partial T^2)_p$ of thiamine in water and in the aqueous solutions of DESs at T = (288.15–318.15) K

T/K	(E_φ^0)/$m^3\cdot mol^{-1}\cdot K^{-1}$	$\alpha \times 10^3$/K^{-1}	10^4 $(\partial^2 V_\varphi^0/\partial T^2)$p
Thiamine hydrochloride + water			
m_2 = 0.000 mol·kg^{-1}			
288.15	0.7823	3.407	−0.015
298.15	0.6322	2.666	
308.15	0.4823	1.995	
318.15	0.3323	1.350	
Thiamine hydrochloride + (ChCl/EG) + water			
m_2 = 0.100 mol·kg^{-1}			
288.15	0.4387	1.2539	−0.002
298.15	0.4587	1.4951	
308.15	0.4787	1.7799	
318.15	0.4987	2.0567	
m_2 = 0.300 mol·kg^{-1}			
288.15	0.3002	1.2625	0.008
298.15	0.3802	1.5655	
308.15	0.4602	1.8660	
318.15	0.5402	2.1321	
m_2 = 0.500 mol·kg^{-1}			
288.15	0.5049	2.0978	0.009
298.15	0.5649	2.3028	
308.15	0.6249	2.4994	
318.15	0.6849	2.6776	
Thiamine hydrochloride + (ChCl/G) + water			
m_2 = 0.100 mol·kg^{-1}			
288.15	0.4828	1.9157	−0.0020
298.15	0.5028	2.0394	
308.15	0.5228	2.1545	
318.15	0.5428	2.2915	
m_2 = 0.300 mol·kg^{-1}			
288.15	0.3909	1.6349	0.0068
298.15	0.4589	1.8801	
308.15	0.5269	2.1228	
318.15	0.5949	2.3370	
m_2 = 0.500 mol·kg^{-1}			
288.15	0.4357	1.8069	0.0072
298.15	0.5077	2.0623	
308.15	0.5797	2.3061	
318.15	0.6517	2.5272	
Thiamine hydrochloride + (ChCl/Urea) + water			
m_2 = 0.100 mol·kg^{-1}			
288.15	0.4136	1.0569	−0.012
298.15	0.4236	1.4498	
308.15	0.4436	1.8344	
318.15	0.4536	1.9385	
m_2 = 0.300 mol·kg^{-1}			
288.15	0.4217	1.6987	−0.002
298.15	0.4417	1.8210	
308.15	0.4617	1.9342	
318.15	0.4817	2.0774	
m_2 = 0.500 mol·kg^{-1}			
288.15	0.4361	1.8497	0.005
298.15	0.4861	2.0154	
308.15	0.536	2.1859	
318.15	0.5861	2.3286	

$$\alpha = \frac{E_\varphi^0}{V_\varphi^0} \tag{4}$$

The values of thermal expansion coefficients, α, for the investigated systems are listed in Table 5. This parameter is basically a balance to evaluate the reaction of the solutions to temperature variations.

The sign of the second derivative of V_φ^0 with respect to temperature leads to obtaining the qualitative information about solute

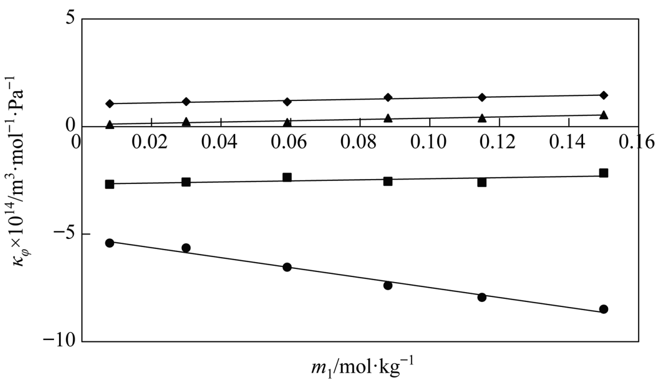

Fig. 2. Apparent molar isentropic compressibility of thiamine hydrochloride, K_φ *versus* its molality, m_1, in aqueous DESs solutions at m_2 = 0.3 mol•kg^{-1} at T = 298.15 K: ●, (Thiamine + water); ■, (Thiamine + (ChCl/Urea) + water); ▲, (Thiamine + (ChCl/EG) + water); ◆, (Thiamine + (ChCl/G) + water).

ability as a structure maker or structure breaker in solutions by using the following expression [34]:

$$\left(\frac{\partial E_\varphi^0}{\partial T}\right)_p = \left(\frac{\partial^2 V_\varphi^0}{\partial T^2}\right)_p = 2C \tag{5}$$

The values of $(\partial^2 V_\varphi^0/\partial T^2)_p$ for studied systems are given in Table 5. Negative value seems to associate with a structure-breaking solute and positive one or closer to zero is associated with a structure-making solute [35]. It is noticed that the values of $(\partial^2 V_\varphi^0/\partial T^2)_p$ are negative and closer to zero for aqueous solutions of thiamine hydrochloride, and become positive with increasing of DES concentration which implies that drug essentially acts as a structure maker.

The partial molar volume of transfer, $\Delta_{tr}V_\varphi^0$, for thiamine from water to the aqueous DESs solutions have been calculated as [36]:

$$\Delta_{tr}V_\varphi^0 = V_\varphi^0(\text{DES} + \text{water}) - V_\varphi^0(\text{water}) \tag{6}$$

The values of $\Delta_{tr}V_\varphi^0$ for the present solutions are given in Table 4. The $\Delta_{tr}V_\varphi^0$ values are positive at all temperatures and increase with increasing in concentration of each DES. The type of interactions possible between solute (thiamine hydrochloride) and co-solute (DES) in ternary solutions (thiamine + DES + water) are: (i), ion–hydrophilic and hydrophilic–hydrophilic interactions (between ions of thiamine hydrochloride with DESs polar groups) (ii) hydrophilic interactions between ions of thiamine hydrochloride and polar groups of DESs (iii) hydrophobic-hydrophobic interactions (between alkyl groups of drug and hydrophobic groups of DESs) (iv) hydrophilic–hydrophobic interactions between the cation and anion parts of the thiamine hydrochloride and hydrophilic groups of DESs. Considering co-sphere overlap model [37,38] two types of interactions including ion–hydrophilic and hydrophilic–hydrophilic have positive effects on $\Delta_{tr}V_\varphi^0$ values whereas, other types result in negative transfer volumes. The positive values of $\Delta_{tr}V_\varphi^0$ express that hydrophilic interactions overcome to the other types of interactions and this trend becomes stronger by increasing in concentration of each DES [39,40].

3.2. Ultrasonic properties

Experimentally measured speeds of sound, u, for thiamine hydrochloride in water and aqueous solutions of DESs at different temperatures are listed in Table 2. The Laplace–Newton's equation

Table 6
Partial molar isentropic compressibility (K_φ^0), adjustable parameters of Eq. (9) (S_k and B_k), transfer isentropic compressibility ($\Delta_{tr}K_\varphi^0$), and standard deviations ($\sigma(K_\varphi^0)$) for thiamine in aqueous solutions of DESs at different temperatures

T/K	$K_\varphi^0\times10^{14}$/m^3·mol^{-1}·Pa^{-1}	$S_k\times10^{14}$/m^3·mol^{-2}·Pa^{-1}·kg	$B_k\times10^{14}$/m^3·mol^{-2}·Pa^{-1}·kg	$\Delta_{tr}K_\varphi^0\times10^{14}$/m^3·mol^{-1}·Pa^{-1}	$\sigma(K_\varphi^0)\times10^{14}$/m^3·mol^{-1}·Pa^{-1}
Thiamine hydrochloride + water					
m_2 = 0.000 mol·kg^{-1}					
288.15	−6.47	3.38	−4.19		0.06
298.15	−5.16	11.93	−23.05		0.12
308.15	−3.06	7.98	−17.53		0.06
318.15	−1.02	5.56	−14.77		0.07
Thiamine hydrochloride + (ChCl/EG) + water					
m_2 = 0.100 mol·kg^{-1}					
288.15	−2.68	−3.92	11.43	3.79	0.06
298.15	−1.19	−2.21	9.49	3.97	0.08
308.15	0.14	−0.70	6.68	3.20	0.06
318.15	1.79	−1.42	8.20	2.81	0.04
m_2 = 0.300 mol·kg^{-1}					
288.15	−1.15	−2.46	6.56	5.32	0.28
298.15	0.08	0.84	3.04	5.24	0.57
308.15	1.67	−0.69	6.72	4.73	0.52
318.15	2.90	−1.99	8.78	3.92	0.14
m_2 = 0.500 mol·kg^{-1}					
288.15	0.37	0.18	1.66	6.84	0.02
298.15	2.00	−0.24	2.77	7.16	0.04
308.15	3.25	−1.96	9.16	6.31	0.06
318.15	3.90	−2.26	9.70	4.92	0.03
Thiamine hydrochloride + (ChCl/G) + water					
m_2 = 0.100 mol·kg^{-1}					
288.15	−1.38	−0.92	4.03	5.09	0.02
298.15	0.06	1.05	2.66	5.22	0.04
308.15	1.79	−1.46	7.94	4.85	0.05
318.15	2.91	−1.88	8.48	3.93	0.07
m_2 = 0.300 mol·kg^{-1}					
288.15	−0.47	−0.53	3.06	6.00	0.02
298.15	1.04	0.34	2.81	6.20	0.04
308.15	2.45	−1.25	7.80	5.51	0.06
318.15	3.46	−2.50	9.86	4.48	0.05
m_2 = 0.500 mol·kg^{-1}					
288.15	1.24	0.42	1.15	7.71	0.03
298.15	2.56	1.21	0.71	7.72	0.03
308.15	3.46	1.52	3.35	6.52	0.06
318.15	4.00	0.60	4.95	5.02	0.05
Thiamine hydrochloride + (ChCl/Urea) + water					
m_2 = 0.100 mol·kg^{-1}					
288.15	−6.46	5.61	0.03	0.01	0.03
298.15	−5.05	6.64	−1.63	0.11	0.06
308.15	−3.25	6.48	−3.68	0.12	0.07
318.15	−0.87	5.99	−2.16	0.15	0.10
m_2 = 0.300 mol·kg^{-1}					
288.15	−4.07	0.33	5.01	2.4	0.11
298.15	−2.68	2.26	2.53	2.48	0.13
308.15	−1.19	3.10	0.28	1.87	0.12
318.15	0.78	2.00	2.64	1.80	0.04
m_2 = 0.500 mol·kg^{-1}					
288.15	−1.43	−7.24	16.38	5.04	0.04
298.15	0.15	−5.71	14.86	5.31	0.04
308.15	1.32	−3.73	11.92	4.38	0.03
318.15	2.70	−4.28	12.88	3.72	0.03

[41] was used to obtain the isentropic compressibility value, κ_s by following equation:

$$\kappa_s = \frac{1}{d\,u^2} \tag{7}$$

Isentropic compressibility (κ_s) is defined by the participation of two parts κ_{s1}(solvent intrinsic) is due to the compression of the solvent (water or aqueous solutions of DESs), and κ_{s2}(solute intrinsic) is due to the compression layer of solute molecules due to the influence of solvent molecules into the empty space of solute. At low concentration of thiamine hydrochloride, the κ_{s1}(solvent intrinsic) is the dominant contribution to the value of κ_s [42]. The lower values of κ_s with increasing in DES concentration is due to the breaking the three-dimensional structure of water and hydrogen bonds form around thiamine hydrochloride molecules, this leads to reduces the compression of water molecules in bulk.

The apparent molar isentropic compressibility, (κ_φ) is obtained from Eq. (8):

$$\kappa_\varphi = \frac{\kappa_s d_0 - \kappa_{s0} d}{m d d_0} + \frac{\kappa_s M}{d} \tag{8}$$

where κ_{s0} and κ_s are isentropic compressibility values of the solvent and solutions, respectively. The calculated κ_φ values for investigated systems are given in Table 2. Also, the apparent molar isentropic compressibility values for thiamine hydrochloride in the aqueous solutions of DESs with concentration of 0.3 mol·kg^{-1} of each DES at 298.15 K are represented in Fig. 2. As can be seen from Table 2, the values of apparent molar isentropic compressibility increase with increasing of temperature and concentration of studied DESs. The variation of apparent molar isentropic compressibility with molality of thiamine hydrochloride can be presented by using Eq. (9):

$$\kappa_\varphi = \kappa_\varphi^0 + S_k m^{1/2} + b_k m \tag{9}$$

where κ_φ^0 is partial molar isentropic compressibility, S_k and b_k have the empirical and similar meaning as in Eq. (2) for apparent molar volumes. The values of κ_φ^0, S_k and b_k are given for the studied solutions along with standard deviations at the experimental temperatures in Table 6. The small b_k values rather than κ_φ^0 predict that solute–solute interactions are negligible in comparison to solute–solvent interactions. The κ_φ^0 values increase from negative values towards positive values by increase in DESs concentration. By increasing DES concentration the formation of ion-pairs cause to suppressed the electrostatic interactions between thiamine hydrochloride and water molecules which leads to become more compressible than that at lower concentration [42].

Partial molar isentropic compressibility of transfer, $\Delta_{tr}\kappa_\varphi^0$ for water and the aqueous solutions of DESs have been calculated using the following equation:

$$\Delta_{tr}k_\varphi^0 = k_\varphi^0(\text{DES} + \text{water}) - k_\varphi^0(\text{in water}) \tag{10}$$

The values of $\Delta_{tr}\kappa_\varphi^0$ for studied solutions are represented in Table 6. The sign of $\Delta_{tr}\kappa_\varphi^0$ is positive in the studied systems and these values increase with a rise in the concentration of DESs. Positive values of κ_φ^0 for thiamine hydrochloride illustrates the dominance of the head charged groups N^+ and COO^- of DESs with the ions of thiamine hydrochloride which increase with a rise in the concentration of DESs. This behavior which observed for partial molar isentropic compressibility of transfer, are in good agreement with volumetric results and supports them.

4. Conclusions

In this study, the thermodynamic properties of thiamine hydrochloride (vitamin B1) as an essential micronutrient in the aqueous solutions of three DESs composed of choline chloride as HBA and ethylene glycol, glycerol, and urea as HBD (ChCl/EG, ChCl/G, and ChCl/U with concentrations of 0.1, 0.3, and 0.5 mol·kg^{-1}) at different temperatures were investigated. The volumetric and ultrasonic properties indicated that the existence of strong interactions between thiamine hydrochloride and studied DESs and dominance of hydrophilic-hydrophilic types of the interactions between solute and solvent. The structure making property of thiamine hydrochloride in aqueous solutions of mentioned DESs are shown by Helper's constant, $(\partial^2 V_\varphi^0/\partial T^2)_p$. Generally, it is concluded that with addition of DES in aqueous solutions of thiamine hydrochloride, the water molecules are released from the hydration layer of solute and finally the interactions between thiamine hydrochloride and DES become stronger.

Data Availability

Data will be made available on request.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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