

Ultrasonic Velocity and Density Studies of Solutions of Maleic Acid and Tartaric Acid in Water at $T = (298.15 \text{ and } 308.15) \text{ K}$

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Abstract Ultrasonic-velocity and density measurements of aqueous solutions of maleic acid and tartaric acid have been made as a function of molality, at $T = (298.15 \text{ and } 308.15) \text{ K}$, and at atmospheric pressure. A molality range has been studied from $(0.2603 \text{ to } 2.6309) \text{ mol}\cdot\text{kg}^{-1}$ and $(0.4451 \text{ to } 2.6621) \text{ mol}\cdot\text{kg}^{-1}$ for maleic and tartaric acids, respectively. The experimental data have been correlated with molality using a polynomial equation. Furthermore, apparent molar volume, partial molar volume, apparent-specific molar volume, isentropic compressibility, apparent molar isentropic compressibility, limiting apparent molar isentropic compressibility, and isentropic apparent-specific compressibility values have been calculated from experimental values of densities and ultrasonic velocities. The calculated parameters have been interpreted in terms of solute–solvent interactions, solute–solute interactions, structure making/breaking behavior of acids, and their taste quality in water.

Keywords Aqueous solution · Density · Maleic acid · Tartaric acid · Ultrasonic velocity

1 Introduction

Maleic acid is a dibasic acid. It is used as an acidulant in the food and beverage industry. Its main use is in syrup, ready tea, orange juice, sports beverages, and other fortified syrup and food. Maleic acid is also used in the manufacture of unsaturated polyester resin [1], pesticides, tartaric acid, fumaric acid, succinic acid, dyeing [1], and auxiliary and preservative grease, and also as a raw material for the production

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of glyoxylic acid by ozonolysis. Its main use is in making maleate salts of bulk drugs like pheneramine maleate and chloropheneramine maleate. The maleate ion is the ionized form of maleic acid. The maleate ion is useful in biochemistry as an inhibitor of transaminase reactions. Maleic acid is also used to prevent rancidity in oils. Tartaric acid is a dibasic acid. It is widely used in food, medicine, light industry, in the industry of electroplating and chemical fertilizers, and applied in tanning photo, glass, enamel, and telecom materials industries. Tartaric acid is also used in pharmaceuticals.

Due to the wide use of maleic and tartaric acids in various technologies and industries, it is necessary to know accurate physicochemical properties of aqueous solutions of maleic and tartaric acids. Hayvarinen et al. [2] undertook the measurements of densities of aqueous solutions of maleic acid as a function of mole fraction at 298.15 K. Manzurola and Apelblat [3] reported partial molar volumes of maleic and tartaric acids in water at 298.15 K. Densities and partial molar volumes of tartaric acid are also reported by Hoiland and Vikingstad [4]. A literature survey reveals that there are no data on ultrasonic velocities of aqueous solutions of maleic and tartaric acids. The aim of this study is to generate accurate data of densities and ultrasonic velocities of solutions of maleic and tartaric acids in water and to gain a better understanding about solute–solvent interactions and the taste quality of these acids in water. This article reports densities and ultrasonic velocities of aqueous solutions of maleic acid and DL (\pm)-tartaric acid at $T = (298.15 \text{ and } 308.15) \text{ K}$ and at atmospheric pressure.

2 Experimental

Maleic and tartaric acids (A. R. Grade, purity >99.5%, Research Chemical Lab., India) were used as-received. To prepare the solutions of maleic and tartaric acids in water, a weight-by-weight method was used. All the solutions were prepared in an air-tight stoppered glass bottle by using triply distilled water. Masses were recorded on an electronic Dhona balance (India) with a precision of $\pm 1 \times 10^{-7} \text{ kg}$. Density

Table 1 Experimental and literature values of density ρ and ultrasonic velocity u of water and toluene at $T = (298.15 \text{ and } 308.15) \text{ K}$

Liquid	Temperature T (K)	Density $\rho \times 10^{-3} \text{ (kg} \cdot \text{m}^{-3}\text{)}$		Ultrasonic velocity $u \times 10^{-3} \text{ (m} \cdot \text{s}^{-1}\text{)}$	
		Experimental	Literature	Experimental	Literature
Water	298.15		0.9970 [7]	1.4968	1.4967 [7] 1.4969 [6]
	308.15		0.9940 [7]	1.5196	1.5198 [7] 1.5203 [6]
Toluene	298.15	0.8622	0.8622 [8,9]	1.3028	1.3029 [9] 1.3040 [10]
	308.15	0.8530	0.8530 [9]	1.2608	1.2606 [9] 1.2590 [11]

measurements were made by using a 15 cm³ bicapillary pycnometer, in a transparent glass-walled water bath with a thermal stability of 0.01 K. The uncertainties in density and temperature measurements were $\pm 1 \times 10^{-7}$ kg · cm⁻³ and ± 0.01 K, respectively. Comparisons between experimental and literature values of densities of toluene are given in Table 1.

Ultrasonic velocities were measured by using a multifrequency ultrasonic interferometer (M-83, Mittal Enterprises, India) at 2 MHz. The ultrasonic interferometer was calibrated with triply distilled deionized water (conductivity, 0.054 μ S · cm⁻¹ [5] or resistivity, 18.18 M Ω [6]). For measurement of ultrasonic velocities of aqueous solutions of maleic and tartaric acids at different temperatures, water at different temperatures was circulated through the measuring cell of the ultrasonic interferometer by a pump. The estimated uncertainty in ultrasonic velocity measurements was 0.03 %. The experimental and literature values of ultrasonic velocities of triply distilled deionized water and toluene are given in Table 1.

Table 2 Density ρ , apparent molar volume $V_{\phi,m}$, partial molar volume $V_{\phi,m}^0$, and solute–solute interaction parameter V_S of maleic and tartaric acids in water at $T=(298.15$ and $308.15)$ K

m (mol · kg ⁻¹)	$T = 298.15$ K		$T = 308.15$ K	
	$\rho \times 10^{-3}$ (kg · m ⁻³)	$V_{\phi,m} \times 10^6$ (m ³ · mol ⁻¹)	$\rho \times 10^{-3}$ (kg · m ⁻³)	$V_{\phi,m} \times 10^6$ (m ³ · mol ⁻¹)
Maleic acid				
0.0000	0.9970		0.9940	
0.2603	1.0072	76.228	1.0039	77.515
0.6112	1.0198	77.137	1.0162	78.271
0.9261	1.0304	77.549	1.0266	78.576
1.4956	1.0483	77.913	1.0440	78.972
1.9153	1.0605	78.101	1.0559	79.142
2.4319	1.0746	78.238	1.0694	79.379
	$V_{\phi,m}^0 \times 10^6 = 75.546$ m ³ · mol ⁻¹		$V_{\phi,m}^0 \times 10^6 = 76.816$ m ³ · mol ⁻¹	
	$V_S \times 10^6 = 1.851$ m ³ · mol ^{-3/2} · kg ^{1/2}		$V_S \times 10^6 = 1.707$ m ³ · mol ^{-3/2} · kg ^{1/2}	
Tartaric acid				
0.0000	0.9970		0.9940	
0.4450	1.0255	83.718	1.0222	84.462
0.5918	1.0343	84.991	1.0306	85.262
1.3419	1.0762	84.456	1.0711	86.161
1.6891	1.0936	84.791	1.0884	86.241
1.9277	1.1053	84.810	1.0998	86.265
2.6621	1.1387	84.922	1.1324	86.354
	$V_{\phi,m}^0 \times 10^6 = 82.955$ m ³ · mol ⁻¹		$V_{\phi,m}^0 \times 10^6 = 83.621$ m ³ · mol ⁻¹	
	$V_S \times 10^6 = 1.294$ m ³ · mol ^{-3/2} · kg ^{1/2}		$V_S \times 10^6 = 1.883$ m ³ · mol ^{-3/2} · kg ^{1/2}	

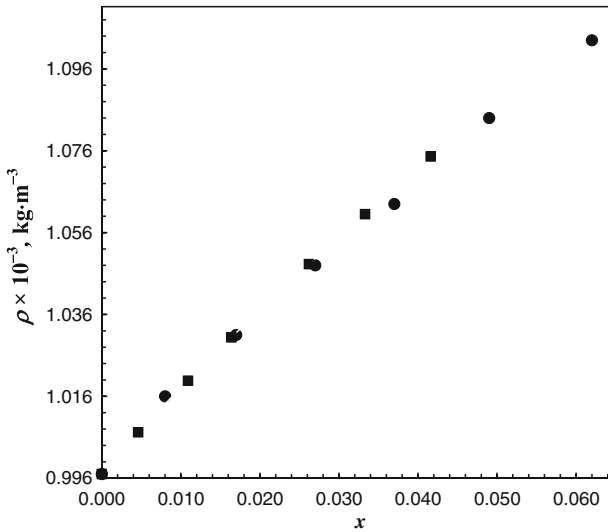


Fig. 1 Densities ρ of maleic acid in water as a function of mole fraction x at $T=298.15$: ●, Hayvarinen et al. [2]; ■, this work

3 Results and Discussion

3.1 Density

Densities of aqueous solutions of maleic and tartaric acids are summarized in Table 2. Hayvarinen et al. [2] reported densities of aqueous solutions of maleic acid as a function of mole fraction at 298.15 K. Figure 1 illustrates comparisons of experimental densities with those reported by Hayvarinen et al. [2]. The observed percentage deviation is 0.79%. From Fig. 1, it is inferred that the density varies nonlinearly with the mole fraction of the solute (also with molality of the solution). From Table 2, it is observed that the density of the solution decreases with an increase in the temperature at fixed concentration. Densities of solutions of tartaric acid in water are graphically presented in Fig. 2. The effect of concentration and temperature on densities of solutions of tartaric acid is similar to those of solutions of maleic acid. To calculate apparent molar volumes ($V_{\phi,m}$) from experimental values of ρ , the following equation [12,13] was used:

$$V_{\phi,m} = (M/\rho) - [(\rho - \rho_0)/(m\rho\rho_0)] \quad (1)$$

M is the molar mass of the maleic or tartaric acid and ρ_0 is the density of water. $V_{\phi,m}$ values are reported in Table 2. The estimated uncertainties in the apparent molar volumes were $0.742 \times 10^{-6} \text{ m}^3 \cdot \text{mol}^{-1}$ for maleic acid and $0.428 \times 10^{-6} \text{ m}^3 \cdot \text{mol}^{-1}$ for tartaric acid. The uncertainty was estimated by the use of the propagation of errors method and at the lowest concentration. The partial molar volume $V_{\phi,m}^0$ has been calculated by using Masson's equation [14],

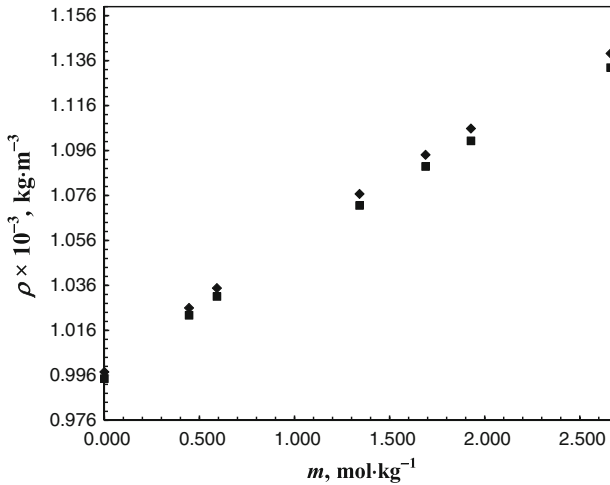


Fig. 2 Densities ρ of tartaric acid in water as a function of molality m at different temperatures: \blacklozenge , $T=298.15$ K; \blacksquare , $T=308.15$ K

$$V_{\phi,m} = V_{\phi,m}^0 + V_S m^{1/2} \quad (2)$$

where $V_{\phi,m}^0$ and V_S are solute, solvent, and temperature-dependent empirical parameters. $V_{\phi,m}^0$ and V_S have been estimated by a least-squares fit of the apparent molar volume data with Eq. 2. Table 2 compiles $V_{\phi,m}^0$ and V_S values for both maleic and tartaric acids. The observed values for $V_{\phi,m}^0$ of maleic and tartaric acids at 298.15 K are $75.546 \times 10^{-6} \text{ m}^3 \cdot \text{mol}^{-1}$ and $82.955 \times 10^{-6} \text{ m}^3 \cdot \text{mol}^{-1}$, respectively. The reported values of $V_{\phi,m}^0$ of maleic and tartaric acids at 298.15 K by Manuzurolo and Apelblat [3] are $75.65 \times 10^{-6} \text{ m}^3 \cdot \text{mol}^{-1}$ and $82.23 \times 10^{-6} \text{ m}^3 \cdot \text{mol}^{-1}$, respectively. Good agreement has been found between estimated and reported values of $V_{\phi,m}^0$ of maleic and tartaric acids at 298.15 K. At the lowest concentration, the estimated uncertainties in the partial molar volumes were $0.764 \times 10^{-6} \text{ m}^3 \cdot \text{mol}^{-1}$ and $0.4443 \times 10^{-6} \text{ m}^3 \cdot \text{mol}^{-1}$ for maleic and tartaric acids, respectively. Information regarding solute–solvent interactions and solute–solite interactions can be obtained from $V_{\phi,m}^0$ and V_S values. The positive values of $V_{\phi,m}^0$ at all temperatures indicate strong solute–solvent interactions [15]. V_S values are positive and smaller than $V_{\phi,m}^0$, suggesting the dominance of solute–solvent interactions over solute–solite interactions. $V_{\phi,m}^0$ increases and V_S decreases with an increase in temperature.

3.2 Ultrasonic Velocity

Ultrasonic velocities (u) of aqueous solutions of maleic acid at 298.15 K and 308.15 K are presented in Table 3. The graphical representation of variations of ultrasonic velocities with molality at (298.15 and 308.15) K is shown in Fig. 3. From Table 3 and Fig. 3, it is revealed that the more concentrated solution of maleic acid at fixed temperature

Table 3 Ultrasonic velocity u , isentropic compressibility K_S , and apparent molar isentropic compressibility $K_{\phi,S}$ of maleic and tartaric acids in water at $T=(298.15$ and $308.15)$ K

m (mol · kg ⁻¹)	$T = 298.15$ K			$T = 308.15$ K		
	$u \times 10^{-3}$ (m · s ⁻¹)	$K_S \times 10^{10}$ (Pa ⁻¹)	$K_{\phi,S} \times 10^{15}$ (m ³ · mol ⁻¹ · Pa ⁻¹)	$u \times 10^{-3}$ (m · s ⁻¹)	$K_S \times 10^{10}$ (Pa ⁻¹)	$K_{\phi,S} \times 10^{15}$ (m ³ · mol ⁻¹ · Pa ⁻¹)
Maleic acid						
0.0000	1.4968	4.477		1.5196	4.357	
0.2603	1.5024	4.399	3.353	1.5240	4.289	7.027
0.6112	1.5088	4.307	5.421	1.5288	4.210	8.872
0.9261	1.5128	4.241	7.297	1.5320	4.150	10.195
1.4956	1.5196	4.131	8.989	1.5380	4.049	11.307
1.9153	1.5232	4.064	10.130	1.5416	3.985	12.019
2.4319	1.5268	3.992	11.233	1.5448	3.918	12.976
Tartaric acid						
0.0000	1.4968	4.477		1.5196	4.357	
0.4451	1.5120	4.265	-11.96	1.5344	4.155	-10.464
0.5918	1.5172	4.200	-11.62	1.5392	4.096	-9.458
1.3419	1.5400	3.918	-8.684	1.5616	3.829	-6.609
1.6891	1.5524	3.794	-8.359	1.5700	3.727	-5.331
1.9277	1.5612	3.712	-8.320	1.5770	3.656	-5.019
2.6621	1.5896	3.475	-8.216	1.5988	3.455	-4.253

shows a higher ultrasonic velocity. At higher temperatures, the solution shows higher values of the ultrasonic velocity. u values of solutions of tartaric acid are included in Table 3. Figure 4 presents plots of u against the molality of solutions. Similar effects of concentration and temperature on ultrasonic velocities of tartaric acid solutions have been observed. u values have been used to calculate the values of isentropic compressibility K_S by using the Laplace equation [16],

$$K_S = 1/(u^2 \rho) \quad (3)$$

K_S , u , and ρ are the isentropic compressibility (Pa⁻¹), ultrasonic velocity (m · s⁻¹), and density (kg · m⁻³) of the solution, respectively. Table 3 shows that the K_S values are positive and become smaller at higher concentrations of both maleic and tartaric acids. The uncertainty in isentropic compressibility was estimated at lowest concentration by using the method of propagation error. The estimated values are 1.023×10^{-13} Pa⁻¹ for maleic acid and 0.981×10^{-13} Pa⁻¹ for tartaric acid.

The apparent molar isentropic compressibility ($K_{\phi,S}$) values are calculated by using the following equation [17]:

$$K_{\phi,S} = (1/m\rho_0)(K_S - K_{\text{Solvent}}) + K_S V_{\phi,m} \quad (4)$$

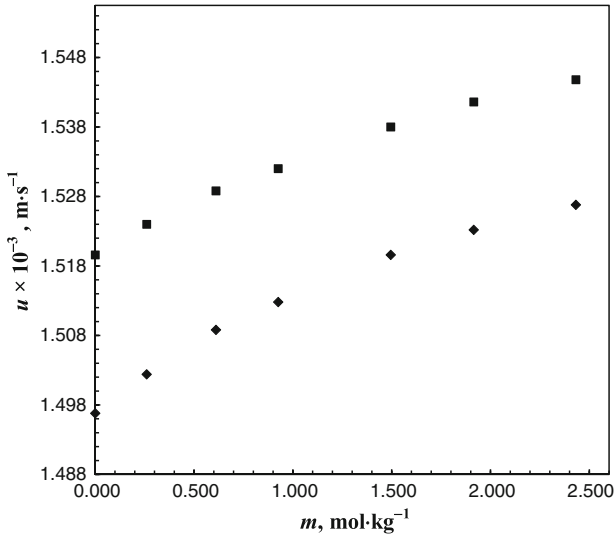


Fig. 3 Ultrasonic velocities u of maleic acid in water as a function of molality m at different temperatures: \blacklozenge , $T = 298.15$ K; \blacksquare , $T = 308.15$ K

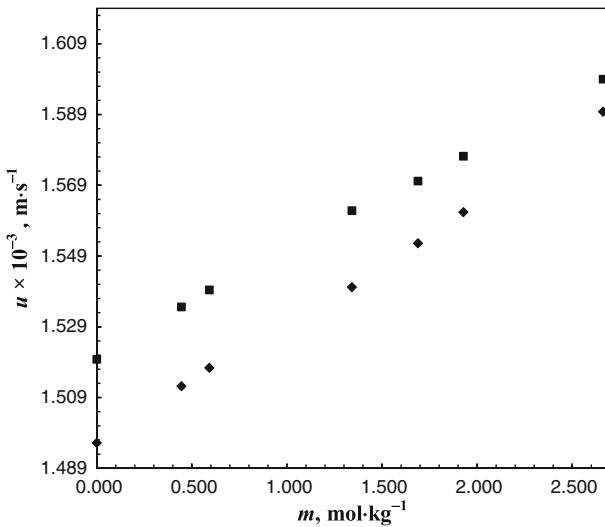


Fig. 4 Ultrasonic velocities u of tartaric acid in water as a function of molality m at different temperatures: \blacklozenge , $T = 298.15$ K; \blacksquare , $T = 308.15$ K

K_{Solvent} is the isentropic compressibility of water. To calculate the isentropic compressibility of water, experimental values of ultrasonic velocities and literature values of densities [7] of water were used. The positive values of $K_{\phi,S}$ suggest that the water molecules around the maleic and tartaric acids' molecules are more compressible than water molecules in the bulk [18, 19]. The uncertainties in apparent molar

Table 4 Least-squares fitted values of parameters of Eq. 5 and apparent isentropic-specific compressibility (AISC) for maleic acid and tartaric acid in water at $T=(298.15$ and $308.15)$ K

$T(K)$	$K_{\phi,S}^0 \times 10^{15}$ (Pa^{-1})	$a \times 10^{15}$ ($\text{m}^3 \cdot \text{mol}^{-2} \cdot \text{Pa}^{-1} \cdot \text{kg}$)	$b \times 10^{15}$ ($\text{m}^3 \cdot \text{mol}^{-3} \cdot \text{Pa}^{-1} \cdot \text{kg}^2$)	$AISC \times 10^{17}$ (Pa^{-1})
Maleic acid				
298.15	-1.675 ± 0.678^a	10.446 ± 1.410^a	-1.387 ± 0.672^a	-1.443
308.15	2.915 ± 0.598^a	8.953 ± 1.245^a	-1.647 ± 0.593^a	2.511
Tartaric acid				
298.15	-21.111 ± 1.398^a	17.127 ± 2.664^a	-5.654 ± 1.180^a	-14.062
308.15	-19.090 ± 0.999^a	15.513 ± 1.903^a	-3.910 ± 0.843^a	-12.719

^aStandard error

isentropic compressibilities were $4.19 \times 10^{-16} \text{ m}^3 \cdot \text{mol}^{-1} \cdot \text{Pa}^{-1}$ for maleic acid and $2.51 \times 10^{-16} \text{ m}^3 \cdot \text{mol}^{-1} \cdot \text{Pa}^{-1}$ for tartaric acid.

The limiting molar isentropic compressibility $K_{\phi,S}^0$ is estimated by fitting of $K_{\phi,S}$ data with the equation [17],

$$K_{\phi,S} = K_{\phi,S}^0 + a m^{1/2} + bm \quad (5)$$

where $K_{\phi,S}^0$, a , and b values for maleic and tartaric acids are included in Table 4. The uncertainties in limiting molar isentropic compressibilities were $2.09 \times 10^{-16} \text{ Pa}^{-1}$ for maleic acid and $4.27 \times 10^{-16} \text{ Pa}^{-1}$ for tartaric acid. The apparent isentropic-specific compressibility is estimated by dividing the limiting molar isentropic compressibility by the molar mass. Table 4 includes the values of apparent isentropic-specific compressibility for maleic and tartaric acids. The low values of the apparent isentropic-specific compressibility show that the solute is strongly hydrated and therefore hydrophilic. Maleic [20] and tartaric acids are hydrophilic acids. This indicates the presence of strong solute–solvent interactions. The main form of solute–solvent interactions is hydrophilic hydration. This results in the formation of structurally enhanced water from rearrangements of the molecules in an organized fashion with strong hydrogen bonds between them. For a sour taste, the broad range of the apparent isentropic-specific compressibility is $6.31 \times 10^{-17} \text{ m}^3 \cdot \text{mol}^{-1} \cdot \text{Pa}^{-1}$ to $-2.49 \times 10^{-16} \text{ m}^3 \cdot \text{mol}^{-1} \cdot \text{Pa}^{-1}$ [21]. The observed values of the apparent isentropic-specific compressibility suggest the sour taste of maleic and tartaric acids.

Density and ultrasonic velocity data of aqueous solutions of maleic and tartaric acids are correlated with m by using the following equation.

$$Y = a_0 + a_1 m + a_2 m^{1.5} + a_3 m^2 + a_4 m^{2.5} \quad (6)$$

where Y is either the density or ultrasonic velocity of maleic acid or tartaric acid solutions in water. The constants a_0 , a_1 , a_2 and a_3 are calculated by the least-squares method. Table 5 summarizes the values of a_0 , a_1 , a_2 , and a_3 .

Table 5 Least-squares fitted values of parameters of Eq. 6 for maleic and tartaric acids in water at $T=(298.15$ and $308.15)$ K

T (K)	Property	$a_0 \times 10^{-3}$	$a_1 \times 10^{-3}$	$a_2 \times 10^{-3}$	$a_3 \times 10^{-3}$
Maleic acid					
298.15	ρ	0.9971 ± 0.0000^a	0.0419 ± 0.0004^a	-0.0057 ± 0.0006^a	-0.0005 ± 0.0002^a
308.15	ρ	0.9941 ± 0.0001^a	0.0401 ± 0.0006^a	-0.0041 ± 0.0007^a	-0.0011 ± 0.0002^a
Tartaric acid					
298.15	ρ	0.9957 ± 0.0010^a	0.0770 ± 0.0055^a	-0.0160 ± 0.0063^a	0.0010 ± 0.0019^a
308.15	ρ	0.9954 ± 0.0017^a	0.0629 ± 0.0009^a	-0.0022 ± 0.0010^a	-0.0029 ± 0.0003^a
Maleic acid					
298.15	u	1.4966 ± 0.0009^a	0.0286 ± 0.0058^a	-0.0126 ± 0.0074^a	0.0014 ± 0.0025^a
308.15	u	1.5206 ± 0.0009^a	0.0133 ± 0.0059^a	0.0015 ± 0.0076^a	-0.0024 ± 0.0026^a
Tartaric acid					
298.15	u	1.4992 ± 0.0046^a	0.0312 ± 0.0248^a	-0.0057 ± 0.0284^a	0.0046 ± 0.0088^a
308.15	u	1.5121 ± 0.0039^a	0.0822 ± 0.0212^a	-0.0611 ± 0.0243^a	0.0188 ± 0.0075^a

^a Standard error

4 Conclusions

1. At constant temperature, the ultrasonic velocity of aqueous solutions of both maleic and tartaric acids increases with an increase in the molality of the solution.
2. At fixed concentration, the ultrasonic velocity of aqueous solutions of both maleic and tartaric acids increases with an increase in the temperature.
3. Strong solute–solvent interactions exist in aqueous solutions of maleic and tartaric acids.
4. Maleic and tartaric acids show a sour taste in water.

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