



Solubility of gallic acid in liquid mixtures of (ethanol + water) from (293.15 to 318.15) K

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ABSTRACT

The solubility of gallic acid in (water + ethanol) binary solvents was determined from (293.15 to 318.15) K at atmospheric pressure using a thermostatted reactor and UV/vis spectrophotometer analysis. The effects of binary solvents composition and temperature on the solubility were discussed. It was found that gallic acid solubility in (water + ethanol) mixed solvents presents a maximum-solubility effect. Two empirical equations were proposed to correlate the solubility data. The calculated solubilities show good agreement with the experimental data within the studied temperature range. Using the experimentally measured solubilities, the thermodynamic properties of dissolution of the gallic acid such as Gibbs energy ($\Delta_{\text{sol}}G^\circ$), molar enthalpy of dissolution ($\Delta_{\text{sol}}H^\circ$), and molar entropy of dissolution ($\Delta_{\text{sol}}S^\circ$) were calculated.

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1. Introduction

Gallic acid (3,4,5-trihydroxybenzoic acid) and its derivatives are phenolic compounds contained in Olive Mill WasteWater (OMWW) [1,2] and present in some plants, such as green and black teas [3], and oak [4]. They are industrially important chemicals widely used in organic synthesis, pharmaceutical, food, and integrated circuit manufacturing. These phenolic compounds are antioxidant [5,6], anti-inflammatory [7], phytotoxic and toxic to bacteria and used in common biological wastewater treatment [8–10]. The solubility of solid compounds in pure solvents and mixed solvents plays a key role in all crystallization processes [11]. Moreover, solubility data in pure solvents and mixed solvents can be considered as helpful in the extraction and purification processes of organic compounds from different matrices.

In some recent works [12,13], solubilities of gallic acid in several pure solvents have been measured as a function of temperature. However, in our knowledge no experimental or theoretical study concerning the solubility of gallic acid in binary solvents has been reported in the literature. As a continuation of our earlier studies concerning the phenolic compounds solubility [14–17], we report here on the effects of binary solvents composition and temperature on the solubility of gallic acid. The solubilities of gallic acid in (water + ethanol) mixed solvents with ethanol mole fractions of 0.0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, and 1.0, on a

solvent-free basis were determined at $T = (293.15, 298.15, 303.15, 308.15, 313.15, \text{ and } 318.15) \text{ K}$. Obtained data were used to calculate appropriate dissolution thermodynamic properties.

2. Materials and methods

Gallic acid ($\text{C}_7\text{H}_6\text{O}_5$, MW = 170.12, >98% pure) and ethanol were purchased from Sigma–Aldrich (Germany). Gallic acid was used without prior treatment, but stored in a desiccator with P_2O_5 once the bottle has been opened. Absolute Ethanol was of analytical grade ($\text{C}_2\text{H}_6\text{O}$, purity = 99.8%, MW = 46.07, $d = 0.79 \text{ g/mL}$). Distilled water (conductivity around $1.5 \mu\text{S} \cdot \text{cm}^{-1}$) has been used.

Binary solvent were prepared by mass using a Sartorius CP225D analytical balance with an accuracy of $\pm 0.01 \text{ mg}$. The uncertainty in the mole fraction of mixed solvents was estimated to be within ± 0.0003 . 25 g of binary solvents were introduced into a double jacketed reactor thermostatted at $(T \pm 0.1) \text{ K}$ (Polystat Huber CC2). An excess of the solid gallic acid was added to the liquid phase and the solution was continuously stirred with a magnetic stirrer. To check the equilibrium, a sample of the liquid phase was taken through a $0.2 \mu\text{m}$ pore syringe filter and the concentration was measured by UV/visible spectrophotometry (Beckman Coulter UV/vis spectrophotometer model DU-520) at 300 nm. The wavelength of 300 nm was determined to be the most adequate for gallic acid quantification because of the maximum absorbance at this wavelength. When the concentration of gallic acid in the liquid phase remained constant, it was assumed that the system was at equilibrium. Several equilibrium times have been tested (from 1

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to 6 h), and it appears that 3 h is always sufficient to reach equilibrium. Two independent experiments were realised to determine the gallic acid solubility. The reproducibility of the data was within 0.5%.

3. Results and discussion

The solubility of gallic acid was measured in pure water, ethanol and (water + ethanol) mixtures, at different temperatures ranging from (293.15 to 318.15) K. This complementary study permits us to extend our solubility data basis of gallic acid [14–17].

To validate the method of solubility determination, the solubility of gallic acid in pure solvents was determined from (293.15 to 318.15) K. As shown in figure 1, solubilities values are similar to those reported in the literature. Compared to pure water, the solubility of gallic acid in pure ethanol is the highest and is more dependent on temperature. This solubility temperature dependence was fitted to the empirical equation proposed by Heidman *et al.*[18]:

$$\ln x_{GA} = A + \frac{B}{T/K} + C \ln(T/K), \quad (1)$$

where x_{GA} is the mole fraction solubility of gallic acid in pure solvents, T is the absolute temperature (K), and A , B , and C are empirical parameters.

The values of empirical parameters A , B and C are given in table 1 together with the corresponding root-mean-square deviations (rmsd's) calculated according to the formula:

$$rmsd = \left[\frac{1}{n} \sum_{i=1}^n (x_{GA}^{cal} - x_{GA}^{exp})^2 \right]^{1/2}, \quad (2)$$

where n is the number of experimental points, and x_{GA}^{cal} and x_{GA}^{exp} represent the solubility of gallic acid calculated from equation (1) and the experimental solubility value, respectively. Calculated and experimental solubilities in pure water, shown in figure 1, demonstrate a very good agreement.

Experimental solubilities were used to calculate the mole fraction solubility x_{GA} in different (water + ethanol) mixtures based on the following equation:

$$x_{GA} = 0.001 \times m_{GA} \times (x_W \times M_W + x_E \times M_E), \quad (3)$$

where m_{GA} , x_W and x_E represent the molality of the gallic acid, the mole fraction of water and ethanol, respectively, and M_{GA} , M_E and M_W are the molecular weights of the gallic acid, ethanol and water, respectively.

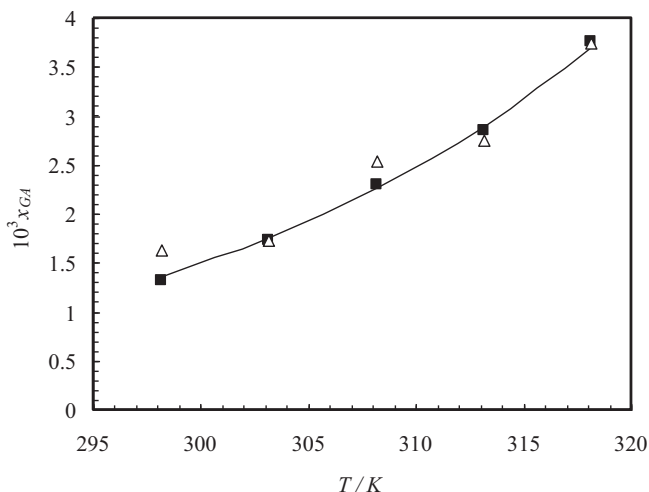


FIGURE 1. Mole fraction solubility of gallic acid in water at different temperatures. ■. This work; literature values¹³ △; line calculated by equation (1).

TABLE 1

Parameters of equation (2) correlated from experimental molar fraction of solubility of gallic acid in pure solvents.

Solvent	A	B/K	C	10 ⁵ rmsd
Water	-93.9147	-2.6453	15.3271	0.0125
Ethanol	-16.0367	-0.6321	2.3083	0.0010

TABLE 2

Experimental (x_{GA}^{exp}) solubility of gallic acid in different (ethanol + water) mixed solvents at various temperatures and atmospheric pressure.

T/K	10 ³ x _{GA} ^{exp}			
	x _E = 0.100	x _E = 0.200	x _E = 0.300	x _E = 0.400
293.15	2.486 ± 0.010	5.893 ± 0.015	12.977 ± 0.021	22.477 ± 0.027
298.15	3.177 ± 0.009	7.276 ± 0.014	16.018 ± 0.026	25.581 ± 0.029
303.15	4.180 ± 0.012	9.487 ± 0.016	18.804 ± 0.024	30.622 ± 0.045
308.15	5.479 ± 0.016	12.002 ± 0.019	22.193 ± 0.026	35.142 ± 0.048
313.15	7.079 ± 0.014	14.719 ± 0.023	26.186 ± 0.029	40.060 ± 0.029
318.15	8.995 ± 0.013	19.294 ± 0.025	31.712 ± 0.031	46.515 ± 0.058
	x _E = 0.500	x _E = 0.600	x _E = 0.700	x _E = 0.800
293.15	30.433 ± 0.052	30.836 ± 0.036	29.001 ± 0.034	28.006 ± 0.033
298.15	35.418 ± 0.043	36.750 ± 0.043	33.788 ± 0.044	32.158 ± 0.041
303.15	39.815 ± 0.038	40.733 ± 0.029	37.321 ± 0.048	36.371 ± 0.058
308.15	45.917 ± 0.042	47.990 ± 0.051	45.258 ± 0.046	41.938 ± 0.033
313.15	52.140 ± 0.054	55.602 ± 0.061	51.986 ± 0.056	48.854 ± 0.053
318.15	59.261 ± 0.057	63.033 ± 0.062	59.282 ± 0.065	55.324 ± 0.061
	x _E = 0.900	x _E = 0.950		
293.15	31.750 ± 0.041	37.972 ± 0.031		
298.15	36.855 ± 0.029	43.797 ± 0.034		
303.15	44.006 ± 0.045	55.489 ± 0.075		
308.15	50.470 ± 0.061	63.203 ± 0.071		
313.15	58.611 ± 0.057	74.612 ± 0.064		
318.15	67.394 ± 0.074	87.011 ± 0.077		

Gallic acid solubilities in a binary (water + ethanol) mixture are listed in table 2 and also shown in figure 2. We find maxima and minima in ranges of ethanol mole fractions respectively.

A fifth-order polynomial equation was proposed to correlate the solubility data of gallic acid in the binary mixed solvents as a function of x_E as follows:

$$\ln x_{GA} = B_0 + B_1 x_E + B_2 x_E^2 + B_3 x_E^3 + B_4 x_E^4 + B_5 x_E^5, \quad (4)$$

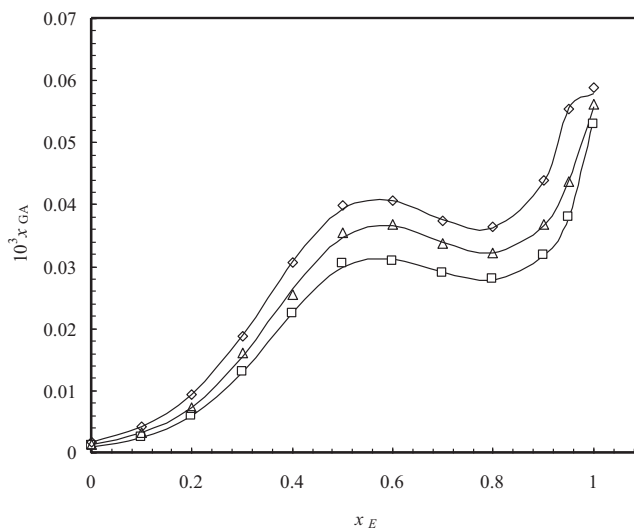


FIGURE 2. Solubility of gallic acid vs the mole fraction of ethanol (0.0 to 1) on a solute-free basis in ethanol + water at different temperatures/K. □, 293.15; △, 298.15; ◇, 303.15; line calculated by equation (6).

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