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Investigation of Solubility Behavior, Data Correlations and Thermodynamic Study of Succinic Acid in Water + Methanol Mixtures at various Temperatures

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Abstract

By using a gravimetric approach, the solubility of succinic acid in water, methanol, and water + methanol binary solvent was determined at temperatures between 305.15 and 313.15 K in the local atmosphere. The findings of the experiment demonstrated a positive correlation between temperature rise and succinic acid solubility. In order to better correlate and match the experimental results, the thermodynamic models van't Hoff and Apelblat were used. Additionally, the DFT investigation was conducted to look into how solvent-solute interactions affected solubility. In addition, the dissolving process's thermodynamic characteristics were computed, and the findings suggested that it was an entropy-driven, spontaneous, endothermic process.

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Keywords

Apelblat Equation;
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Introduction

With the chemical formula $C_4H_6O^4$, succinic acid (SA) is a dicarboxylic acid that is also known as butanedioic acid or amber acid. It is used as a precursor in the synthesis of several fine and bulk compounds, such as vitamins, antibiotics, medications, and biodegradable polymers like polyamides and polybutyrate succinate, as well as food additives, ion chelators, and surfactants.[1-5] Studies on the solubility of succinic acid in a variety of organic solvents, including isopropanol,[6] water + methanol and water + ethanol mixtures,[7] binary mixtures of cyclohexanone, cyclohexanol, and cyclohexane,[8] binary mixtures of methanol, ethanol, and propanol,[9] and fumaric acid's solubility in n-propanol, isopropanol, ethanol, and acetone,[10] are all reported in the literature. With the use of a gravimetric technique, the solubility of succinic acid

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in water, methanol, and water + methanol binary solvent was investigated at temperatures between 305.15 and 313.15 K in the local environment. Apelblat and van der Hoff equations are two examples of thermodynamic models with which obtained experimental solubilities have been related. Furthermore, the van der Hoff equation has been used to evaluate the energies of solution in order to obtain a better understanding of the phenomena of the succinic acid dissolving process.

Materials and Methods

Triple-distilled water was employed in these studies. MERCK provided 99.5% pure succinic acid, whereas Jiangyin Huaxi International Trade Co. (China) provided 99.9% pure methanol. The equipment and procedures utilized to determine solubility have previously been thoroughly discussed.[11–13]

In this experiment, a specifically built 100 mL double jacketed flask was used to generate binary solvent mixtures by weight (Shimadzu, Auxzzo) with an uncertainty of ± 0.1 mg. An excess of succinic acid was added to the mixes. Water was pumped between the flask's inner and outer sides at a steady temperature.

A thermostat was used to regulate the water's temperature, keeping it within (±0.1) K. In order to ensure equilibrium, the solution was constantly stirred with a magnetic stirrer for a duration of about one hour. Following this, the solution was allowed to stand for one hour. The supernatant liquid was then taken out of the flask and placed in a weighing container in a predetermined quantity using a pipette that is hotter than the solution. This sample was weighed, and it was then maintained at 343 K in an oven until all of the solvent had evaporated. Weighing twice or three times to get a consistent weight allowed us to confirm this.

Weight of solution and solute have been used to calculate solubility. Every solubility value measured is the mean of at least three experiments.

Using standard Eqs. 1 and 2, the saturated mole fraction solubility (X_b), starting methanol mole fraction (X_C^0), and initial water mole fraction (X_A^0) were determined.

$$X_b = (m_b / M_b) / (m_b / M_b + m_A / M_A + m_C / M_C) \quad ...(1)$$

$$X_C^0 = (m_C / M_C) / (m_A / M_A + m_C / M_C) \text{ and } X_A^0 = (m_A / M_A) / (m_A / M_A + m_C / M_C) \quad ...(2)$$

In this case, the molecular weights of the solute, water, and methanol are represented by the letters M_b, M_A, and M_C respectively, while the masses of the solute, water, and methanol are denoted by the letters m_b, m_A, and m_C.

Results and Discussion

Table 1 lists the solubility values (X_b) of SA in pure water and methanol between 305.15 and 313.15 K, and Figure 1 illustrates how these values change with temperature. It has been noted that temperature has a direct correlation with SA's solubility in pure solvents. In water + methanol solvent mixes at T = 305.15 to 313.15 K, Table 2 displays the mole fraction solubilities of SA along with the solubilities that were estimated using corresponding models. Figures 2 and 3, respectively, illustrate the solubility of SA in binary solvent mixtures as a function of temperature and the initial mole fraction of solvent.

Table 1. Experimental of mole fraction solubility (X_b) of SA in pure water & methanol at T= (305.15 -313.15 K)

T/K	X_b	
	Water	Methanol
305.15	0.0164	0.0628
308.15	0.0184	0.0665
310.15	0.0203	0.0716
313.15	0.0227	0.0730

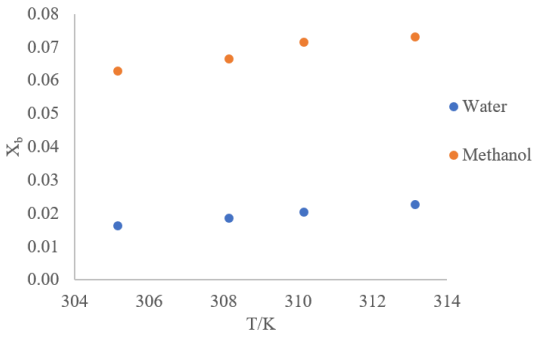

Fig. 1: Mole fraction solubility (Xb) variation with temperatures for water (♦), methanol (■)

It is evident that when temperature rises, SA becomes more soluble in all binary solvent mixes. Additionally, when the initial mole fraction of methanol increases, so does SA's solubility in the water + methanol mixture.

Table 2: Experimental (X_b) and calculated mole fraction solubility of SA in various initial mole fraction X_C^0 of methanol at T = 305.15 to 313.15 K

T/K	X_C^0	X_b^{Expt}	X_b^{Apel}	$X_b^{van't}$	T/K	X_b^{Expt}	X_b^{Apel}	$X_b^{van't}$
305.15	0.0000	0.0164	0.0164	0.0164	310.15	0.0203	0.0203	0.0201
	0.0588	0.0188	0.0188	0.0190		0.0247	0.0245	0.0240
	0.1232	0.0225	0.0225	0.0225		0.0282	0.0282	0.0282
	0.1942	0.0278	0.0278	0.0283		0.0361	0.0363	0.0358
	0.2726	0.0328	0.0328	0.0329		0.0399	0.0399	0.0398
	0.3599	0.0398	0.0398	0.0399		0.0474	0.0476	0.0475
	0.4575	0.0501	0.0499	0.0494		0.0551	0.0544	0.0550
	0.5675	0.0510	0.0513	0.0503		0.0566	0.0581	0.0593
	0.6922	0.0562	0.0562	0.0563		0.0644	0.0644	0.0643
	0.8350	0.0614	0.0615	0.0612		0.0677	0.0681	0.0684
	1.0000	0.0628	0.0625	0.0631		0.0716	0.0704	0.0697
308.15	0.0000	0.0184	0.0184	0.0185	313.15	0.0227	0.0227	0.0227
	0.0588	0.0220	0.0221	0.0219		0.0269	0.0270	0.0274
	0.1232	0.0258	0.0258	0.0258		0.0321	0.0321	0.0321
	0.1942	0.0332	0.0331	0.0326		0.0405	0.0404	0.0411
	0.2726	0.0370	0.0370	0.0369		0.0445	0.0445	0.0446
	0.3599	0.0445	0.0444	0.0443		0.0527	0.0526	0.0527
	0.4575	0.0515	0.0521	0.0527		0.0590	0.0592	0.0586
	0.5675	0.0558	0.0544	0.0555		0.0672	0.0667	0.0653
	0.6922	0.0612	0.0611	0.0610		0.0694	0.0694	0.0695
	0.8350	0.0655	0.0651	0.0655		0.0736	0.0735	0.0731
	1.0000	0.0665	0.0676	0.0670		0.0730	0.0733	0.0740

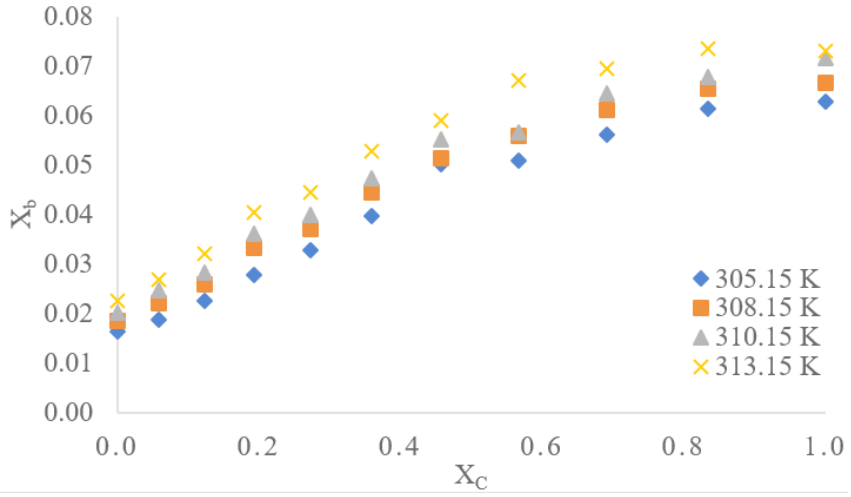

Fig. 2: Mole fraction solubility (X_b) variation with Initial mole fraction (X_C^0) of methanol, at various temperatures (◆T=305.15 K, ■T=308.15 K; ▲T=310.15 K; ×T=313.15 K)

Apelblat[14-15] and the van't Hoff model[16] are used to link the observed X_b values of SA with temperature. Table 2 provide the computed values of SA in binary solvents for X_b^{Apel} and $X_b^{van't}$. It is discovered that there is relatively little variation between the calculated and experimental solubilities of these three models. In terms of correlation coefficient (R^2) values, the X_b values of SA are correlated with the X_b^{Apel} and $X_b^{van't}$ values of SA in all binary solvents.

The correlation coefficient (R^2) values are nearly equal to one. Based on the Apelblat Model and van't Hoff model, the estimated solubility and the measured mole fraction solubility show extremely high agreement in these results. As a result, the solubility data fits the van der Hoff equation and the Apelblat Model extremely well. Table 3 and 4 presents the values of the parameters derived from the van der Hoff equation and the Apelblat model, together with R^2. The correlation coefficient data ($R^2 \approx 1$) show a strong relationship between the experimental mole fraction solubility, the Apelblat Model, and the van der Hoff model.

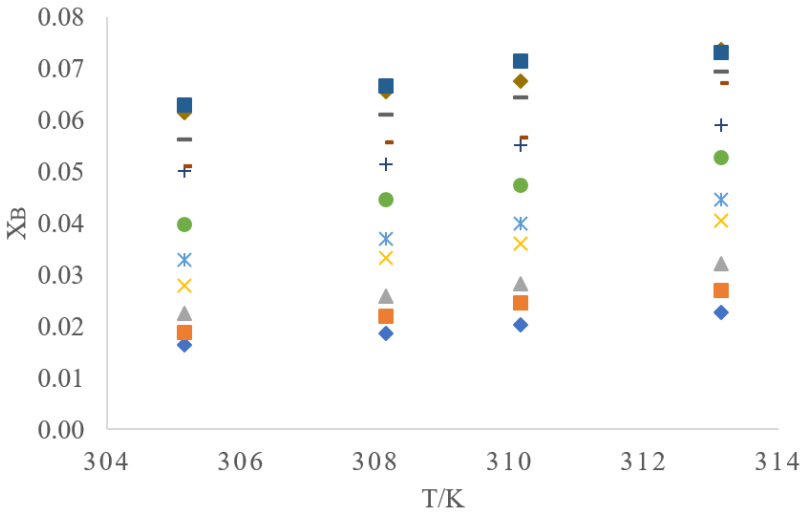

Fig. 3: Mole fraction solubility (X_b) variation with temperature at initial mole fraction (X_c^0) of methanol (◆= wt. fraction 0.0; ■ = 0.1; ▲= 0.2; × = 0.3; × = 0.4; ● = 0.5; + = 0.6; - = 0.7; — = 0.8 and ◇ = 0.9; □ =1)

Table 3: Model parameters and correlation coefficient of the Apelblat equation

Solvent	X_c^0	Parameters			R^2
		A	B	C	
Methanol	0.0000	-2169.7	92508.69	325.6688	0.9999
	0.0588	663.7504	-33461.7	-97.4497	0.9986
	0.1232	266.753	-16044.3	-38.1014	0.9999
	0.1942	2587.085	-122710	-382.557	0.9994
	0.2726	163.0504	-10732.8	-22.9503	0.9999
	0.3599	116.2437	-8315.35	-16.1196	0.9991
	0.4575	-1888.83	84837.85	281.0471	0.9791
	0.5675	-3597.38	162342.2	535.3105	0.9631
	0.6922	275.6835	-14924.4	-40.1436	0.9999
	0.8350	-926.877	40618.35	138.2637	0.9964
	1.0000	1529.997	-71975.1	-226.699	0.9581

Table 4: **Model parameters and correlation coefficient of the Van't Hoff equation**

Solvent	X_c^0	Parameters		R^2
		A	B	
Methanol	0.0000	8.7734	-3932.27	0.9984
	0.0588	10.3223	-4358.59	0.9856
	0.1232	10.1882	-4266.85	0.9998
	0.1942	11.0468	-4459.11	0.9874
	0.2726	8.5093	-3638.71	0.9999
	0.3599	7.6989	-3332.68	0.9991
	0.4575	3.6632	-2035.70	0.9492
	0.5675	7.2535	-3125.86	0.9185
	0.6922	5.3673	-2515.70	0.9995
	0.8350	4.1533	-2119.88	0.9894
	1.0000	3.4659	-1900.85	0.9361

Table 5: **Thermodynamic parameters relative to solution process of SA at Thm= 309.15K**

X_c^0	ΔH^0_{sol} KJK^{-1} mol^{-1}	ΔG^0_{sol} KJK^{-1} mol^{-1}	$T\Delta S^0_{sol}$ KJK^{-1} mol^{-1}	ζH%	ζTS%
0.0000	32.69	9.94	22.75	58.97	41.03
0.0588	36.24	9.51	26.73	57.56	42.44
0.1232	35.48	9.10	26.37	57.36	42.64
0.1942	37.07	8.51	28.56	56.48	43.52
0.2726	30.25	8.22	22.04	57.86	42.14
0.3599	27.71	7.76	19.95	58.14	41.86
0.4575	16.93	7.36	9.57	63.89	36.11
0.5675	25.99	7.20	18.79	58.04	41.96
0.6922	20.92	6.98	13.94	60.01	39.99
0.8350	17.63	6.81	10.81	61.98	38.02
1.0000	15.80	6.76	9.05	63.60	36.40

The thermodynamic study of solubility of SA is carried out in order to evaluate dissolution pattern of SA in pure as well as binary solvent combinations. Thermodynamic parameters (ΔH^0_{soln}, ΔS^0_{soln}, ΔG^0_{soln}) of dissolution are calculated for this purpose. Using Eq., the ΔH^0_{soln} values at the mean harmonic temperature (T_{hm}) of 309.15 K are calculated using van't Hoff analysis.[17,18]

$$\Delta H^0_{sol} = -R\left(\frac{\partial \ln x_b}{\partial(1/T)}\right) = -R\left[\frac{\partial \ln x_b}{\partial\left(\frac{1}{T}-\frac{1}{T_{mean}}\right)}\right] \quad ...(4)$$

$$\Delta G^0_{soln} = -R.T \times intercept \quad ...(5)$$

$$\Delta S^0_{soln} = \frac{\Delta H^0_{soln} - \Delta G^0_{soln}}{T_{mean}} \quad ...(6)$$

Table 5 displays the values of ΔG^0_{soln}, ΔH^0_{soln}, and ΔS^0_{soln}, for the SA dissolution in water + methanol. The findings indicate that when the initial mole fraction of alcohols increases, the, ΔH^0_{soln} values for SA dissolution in all mixes decrease. Every mixture's

SA dissolving process is endothermic, as indicated by the positive ΔH^0_{soln} values. In pure water, the value of ΔH^0_{soln} is 32.69 $kJK^{-1}mol^{-1}$, whereas in methanol, it is 15.80 $kJK^{-1}mo^{-1}$. This shows that dissolving SA in water takes more energy than dissolving it in methanol. The total organic acid solubility and the Gibbs free energy data show a similar trend: the greater the solubility values, the lower the Gibbs free energy.

Conclusion

The mole fraction solubility of succinic acid (SA), a significant industrial chemical, in pure and binary solvent mixtures has been experimentally evaluated within the temperature range of 303.15 to 313.15K. Investigations are done on the impact of temperature and solvent composition. It has been noted that solubility constantly rises with temperature, indicating that all solvent mixes have a positive dissolving enthalpy.

As the initial mole fraction of methanol increases, so does SA's solubility in the water + methanol combination. Using the Apelblat and van der Hoff models, the experimental solubility values of SA are extremely strongly associated with temperature. Thermodynamic characteristics demonstrate that the SA dissolving process is endothermic.

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Conflict of interest

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Data Availability Statement

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Ethics Statement

This research did not involve human participants, animal subjects, or any material that requires ethical approval.

Informed Consent Statement

This study did not involve human participants, and therefore, informed consent was not required

Clinical Trial Registration

This research does not involve any clinical trials.

Authors' Contribution

The sole author was responsible for the conceptualization, methodology, data collection, analysis, writing, and final approval of the manuscript.

References

1. Zeikus J.G., Jain M.K., Elankovan P. Biotechnology of succinic acid production and markets for derived industrial products. *Appl. Microbiol Biotechnol.* 1999;51:545-552.
2. Song H., Lee S.Y. Production of succinic acid by bacterial fermentation. *Enzyme and Microbioal Technology.* 2006;39:352-361.
3. Willke, T., Vorlop, K.D. Industrial bioconversion of renewable resources as an alternative to conventional chemistry. *Appl Microbiol Biotechnol.* 2004;66:131–142.
4. Hong, S.H. Systems approaches to succinic acid-producing microorganisms. *Biotechnol. Bioprocess Eng*. 2007;12:73–79.
5. Barros M. d., Freitas S., Padilha G. S., Alegre R. M. Biotechnological Production of Succinic Acid by Actinobacillus Succinogenes Using Different Substrate. *Chemical Engineering Transaction.* 2013;32:985-990.
6. Davey R. J., Mullin J. W., Whiting M. J. L. Habit modification of succinic acid crystals grown from different solvents, *J. Cryst. Growth.* 1982; 58:304–312.
7. Jiang X., Hu Y., Meng Z., Yang W., Shen F. Solubility of succinic acid in different aqueous solvent mixtures: Experimental measurement and thermodynamic modeling. *Fluid Phase Equilib*. 2013;341:7–11.

8. Sheng X., Luo W., Wang Q. Determination and Correlation for the Solubilities of Succinic Acid in Cyclohexanol + Cyclohexanone + Cyclohexane Solvent Mixtures. *J. Chem. Eng. Data*. 2018;63:801–811.
9. Bancroft W.D., Butler F.J.C. Solubility of Succinic Acid in Binary Mixtures. *J. Phys. Chem.* 1932;36:2515–2520.
10. Dang L., Du W., Black S., Wei H. Solubility of Fumaric Acid in Propan-2-ol, Ethanol, Acetone, Propan-1-ol, and Water. *J. Chem. Eng. Data.* 2009;54:3112–3113.
11. Pawar R. R., Golait S. M., Hasan M., Sawant A. B. Solubility and Density of Potassium Iodide in a Binary Propan-1-ol−Water Solvent Mixture at (298.15, 303.15, 308.15, and 313.15) *K. J. Chem. Eng. Data.* 2010;55:1314-1316.
12. Pawar R. R., Aher C. S., Pagar J. D., Nikam S. L., Hasan M. Solubility, Density and Solution Thermodynamics of NaI in Different Pure Solvents and Binary Mixtures. *J. Chem. Eng. Data.* 2012;57:3563-3572.
13. Pawar R. R., Nahire S. B. Investigation Correlation and DFT study for solubility of Malonic acid in water + methanol and water + ethanol binary solvents at T = 293.15 to 313.15 K. *Research J. Pharm. and Tech.* 2021;14(3):1226-1232.
14. Apelblat A., Manzurola E. J. Solubilities of o-acetylsalicylic, 4-aminosalicylic, 3,5-dinitrosalicylic, and p-toluic acid, and magnesium-DL-aspartate in water from T = (278 to 348) *K. Chem. Thermodyn.* 1999;31:85-91.
15. Yang Z. H., Shao D. F., Zhou G. Q. Solubility parameter of lenalidomide for predicting the type of solubility profile and application of thermodynamic model. *J. Chem. Thermodyn.* 2019;132:268-275.
16. Prausnitz J.M., Lichtenthaler R.N., Azevedo E.G. Molecular Thermodynamics of Fluid-Phase Equilibria, 3rd ed., Englewood Cliffs, NJ, Prentice- Hall, 1999.
17. Yu Y., Zhang F., Gao X. Q., Xu L., Liu G. J. Experiment, correlation and molecular simulation for solubility of 4-methylphthalic anhydride in different organic solvents from T = (278.15 to 318.15) *K Mol. Liq.* 2019;275:768-777.
18. Hildebrand J. H., Prausnitz J. M., Scott R. L. Regular and Related Solutions,Van Nostrand - Reinhold, NewYork,1970.