MODELLING OF THE THERMOPHYSICAL LACTIC ACID AQUEOUS SOLUTIONS. DENSITY AND VISCOSITY

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Abstract: *Lactic acid is an industrially important product with a large and rapidly expanding market due to its attractive and valuable multi-function properties. It is widely used in various fields such as food industry, in pharmaceuticals and cosmetics etc. and knowing and predicting the evolution of its thermophysical properties at any moment may be very useful.*
In this paper various mathematical relations between lactic acid concentrations and temperature with density and dynamic viscosity were established. The known data were fitted in different equations in order to assess and select a suitable mathematical model. Taking in consideration the level of precision and the simplicity of formulation several equations were generated for each thermophysical property. For density, two equations were formulated with average relative errors (absolute value) of 0.062% and respectively R^2 = 0.9999 and average relative errors of 0.055% for intervals of temperature of 298.15 to 353.15 K and dry matter concentration range between 9.16 to 85.32%.
For the dynamic viscosity an equation based on Arrhenius mathematical model with average relative errors of 1.11% and an equation with other mathematical formulation with R^2 = 0.9997 were generated in the same range of temperature and citric acid concentration.
The obtained equations can be uploaded in computer software for storing, organizing and manipulating data available both for industrial and academic users and so facilitating the sizing and optimization calculations of various technological processes and equipments.

Keywords: *lactic acid, thermo-physical properties, mathematical modelling*

1. Introduction

Lactic acid is an organic acid found in many products of natural origin. The first reports on isolation of lactic acid from milk can be found in as early as 1780 and the solidification by self-esterification some years later [1]. Lactic acid can be obtained via chemical synthesis [2-4] or carbohydrate fermentation [5-6] with the help of microorganisms such as *Lactobacillus rhamosus* [7], *Lactococcus lactis* [8] or *Lactobacillus helveticus* [9]. This last technique has a significant advantage in that by choosing a strain of microorganism able to produce only one enantiomer, an optically pure product can be obtained, whereas synthetic production results in a racemic mixture [10]. One of the most important steps of lactic acid production lies in the separation process which is needed to recover and purify the product from the fermentation broth. Different methods are available to this purpose. Among them, solvent extraction followed by centrifugal short path distillation [11], extraction with aliphatic amines [12] or liquid ions [13], nanofiltration [14-15], vapor-permeation assisted esterification [16], reverse osmosis downstream process [17], chromatography [18], electrodialysis [19-21], adsorption technology [22] conducted to satisfactory results. Lactic acid and its derivatives are widely used in the food [23-24], pharmaceutical leather, and textile industries [1]. Recently, there has been an increased interest in lactic acid as a raw material for production of polylactic acid [25-27], a polymer used as a specialty medical and environmental-friendly biodegradable plastic as an alternative for substituting conventional plastic produced from petroleum oil. This new material can be employed as film barrier on intra-abdominal adhesion formation [28], as colloidal drug delivery system [29] as membrane for periodontal guided tissue regeneration [30] etc.

For proper and adequate process development it is necessary to use a lactic acid with high purity. Its production and further employment depend on some of its thermophysical properties. Among them density and viscosity are two of the most important ones. As for other types of products [31-32], the evolution of these lactic acid characteristics are related to parameters such temperature and solution concentrations. Different data are available in literature in this field but their use is rather difficult. As consequence, this work intended to establish mathematical relations between aqueous solutions of lactic acid density and viscosity and the above mentioned parameters. Relative error, ANOVA test and correlation coefficient were used in order to verify the similarity between the experimental data and that proposed by the obtained mathematical models.

2. Experimental

Experimental data provided by the scientific publications (Tables 1 and 2) concerning the variation of aqueous lactic acid solutions density and dynamic viscosity with concentration and temperature were used as primary data for the regression analysis.

Table 1.
Variation of lactic acid aqueous solutions density with temperature and lactic acid content [1]

Lactic acid concentration, *C* [% w/w]	**Density, ρ [kg/m^3]**							
	Temperature, *T* [K]							
	293.15	298.15	303.15	313.15	323.15	333.15	343.15	353.15
9.16	1019.5	1018.1	1015.8	1011.3	1006.7	1000.7	995.04	988.99
24.35	1056.7	1054.4	1051.8	1047.1	1041.4	1035.1	1029.5	1022.6
45.48	1109.8	1105.3	1101.8	1094.2	1087.0	1079.2	1072.1	1063.9
64.89	1155.2	1151.8	1147.2	1139.8	1132.0	1123.5	1115.3	1099.6
75.33	1178.6	1174.8	1170.1	1161.3	1152.6	1142.5	1134.0	1125.1
85.32	1198.9	1194.8	1190.1	1181.3	1171.8	1163.1	1153.6	1144.3

Table 2.
Variation of lactic acid aqueous solutions dynamic viscosity with temperature and lactic acid content [1]

Lactic acid concentration, *C* [% w/w]	**Dynamic viscosity, $\mu \cdot 10^3$ [Pa · s]**						
	Temperature, *T* [K]						
	298.15	303.15	313.15	323.15	333.15	343.15	353.15
9.16	1.150	1.030	0.809	0.671	0.572	0.473	0.416
24.35	1.670	1.460	1.130	0.918	0.746	0.632	0.532
45.48	3.090	2.740	2.030	1.590	1.260	1.020	0.843
64.89	6.960	6.010	4.220	3.120	2.380	1.850	1.470
75.33	13.03	10.55	7.080	4.980	3.570	2.730	2.080
85.32	28.50	22.60	13.91	9.400	6.400	4.590	3.400

Microsoft Excel™ 2010 software was employed for typical data integration, graphical representations and ANOVA analysis. ANOVA analysis tool is able to compare the experimental and the calculated data generated by the established mathematical models.
Complex and atypical data plotting in 2D (“vapour pressure” model, “heat capacity” model etc.) were performed with CurveExpert® software and 3D representations as a surface response were fitted and analyzed in TableCurve 3D® v.4 software.
Thermo-physical property vs. Temperature, Thermo-physical property vs. Lactic acid concentration in aqueous solutions were plotted and different types of regression techniques, involving the method of least squares, relative error ε (Equation 1) and ANOVA were used to reveal the best-fit equation.

$$\varepsilon = \left| \frac{Data_{experimental} - Data_{calculated}}{Data_{calculated}} \right| \cdot 100[\%] \quad (1)$$

3. Results and discussions

3. 1. Density

Using Microsoft Excel™ 2010 spreadsheets and CurveExpert® software, 8 quadratic correlations (taking in consideration the best fit and simplicity in formulation) between lactic acid concentrations *C* [% w/w] and density *ρ* [kg/m^3], at constant temperature *T*, [K] have been established:

$$\rho = a_1 + a_2 C + a_3 C^2 \quad (2)$$

The a_1 and a_2 values are presented in Table 3 and the regression coefficients R^2 are greater than 0.99, thus indicating a good correlation of variables.
In order to correlate a_1, a_2 and a_3 coefficients with temperature *T*, [K], several models were uploaded in CurveExpert® software (1st, 2nd and 3rd degree polynomial equations, “vapor pressure” model, “heat capacity” model etc.). The best fit model is the quadratic equation with good regression coefficients (Table 4).

$$Coefficient = b_1 + b_2 T + b_3 T^2 \quad (3)$$

Table 3.
Coefficients for equation no. 2

Temperature, *T* [K]	Equation 2 coefficients			
	a_1	a_2	a_3	R^2
293.15	994.6371	2.6771	-0.0032	0.999
298.15	994.1297	2.5713	-0.0024	0.999
303.15	992.4598	2.5202	-0.0023	0.999
313.15	989.0405	2.4335	-0.0020	0.999
323.15	984.9243	2.3689	-0.0020	0.999
333.15	980.2156	2.2684	-0.0014	0.999
343.15	975.2492	2.2243	-0.0016	0.999
353.15	969.7177	2.1592	-0.0013	0.999

Table 4.
Coefficients for equation no. 3

Equation 2 coefficients	Equation 3 coefficients			
	b_1	b_2	b_3	R^2
a_1	- 0.0574246013	0.0003189759	-4.54E-07	0.9044
a_2	11.8306041781	- 0.0506144371	5.8356E-05	0.9912
a_3	813.3102959923	1.4872913211	-0.0029584854	0.9993

Combining the equations 2 and 3 and, the final form of proposed equation model (Equation 4) is:

$$\rho = (b_{1a3} + b_{2a3} \cdot T + b_{3a3} \cdot T^2) + (b_{1a2} + b_{2a2} \cdot T + b_{3a2} \cdot T^2) \cdot C + (b_{1a1} + b_{2a1} \cdot T + b_{3a1} \cdot T^2) \cdot C^2 \quad (4)$$

Using the relative error equation the calculated data given by the density mathematical model and the existing experimental data were compared (Table 5) obtaining a final average of 0.062%.

Table 5.
For densities of citric acid aqueous solutions the absolute value of relative errors for calculated data versus tabular data

Lactic acid concentration, C [% w/w]	Density, ρ [kg/m³]											
	Temperature, T [K]											
	293.15		ε, %	298.15		ε, %	303.15		ε, %	313.15		ε, %
	ED*	CD*		ED	CD		ED	CD		ED	CD	
9.16	1019.5	1019.1	0.044	1018.1	1017.3	0.082	1015.9	1015.3	0.051	1011.4	1011.1	0.029
24.35	1056.7	1057.8	0.103	1054.5	1055.3	0.079	1051.8	1052.6	0.076	1047.2	1047.1	0.009
45.48	1109.8	1109.5	0.022	1105.4	1106.1	0.068	1101.8	1102.6	0.073	1094.3	1095.5	0.113
64.89	1155.2	1154.7	0.046	1151.8	1150.7	0.098	1147.2	1146.6	0.053	1139.9	1138.4	0.130
75.33	1178.6	1178.1	0.042	1174.8	1173.8	0.085	1170.1	1169.5	0.052	1161.3	1160.8	0.042
85.32	1198.9	1199.8	0.081	1194.8	1195.4	0.052	1190.1	1190.9	0.070	1181.3	1181.9	0.047
	Average ε, %		0.057	Average ε, %		0.077	Average ε, %		0.062	Average ε, %		0.061
	Temperature, T [K]											
	323.15		ε, %	333.15		ε, %	343.15		ε, %	353.15		ε, %
	ED	CD		ED	CD		ED	CD		ED	CD	
9.16	1006.7	1006.4	0.038	1000.8	1001.2	0.040	995.0	995.5	0.043	989.0	989.3	0.032
24.35	1041.5	1041.2	0.029	1035.1	1034.9	0.019	1029.6	1028.4	0.116	1022.6	1021.5	0.106
45.48	1087.0	1088.2	0.108	1079.3	1080.7	0.137	1072.2	1073.1	0.082	1064.0	1065.2	0.117
64.89	1132.1	1130.0	0.177	1123.6	1121.6	0.088	1115.3	1113.0	0.029	1099.6	1104.3	0.064
75.33	1152.6	1152.0	0.053	1142.5	1143.1	0.050	1134.1	1134.0	0.004	1125.1	1124.9	0.022
85.32	1171.8	1172.7	0.073	1163.1	1163.3	0.019	1153.6	1153.9	0.023	1144.3	1144.3	0.003
	Average ε, %		0.079	Average ε, %		0.058	Average ε, %		0.049	Average ε, %		0.057

* ED – experimental data, CD – calculated data

The ANOVA analysis was used to compare the values of density experimental and calculated data at 6 different concentrations in 8 temperatures variation. The results presented in Table 6 showed that the sample *P*-value is 0.991263 greater than the targeted alpha 0.05 and the *F crit* value is larger than the *F*-test value and as consequence the null hypothesis is not rejected indicating that is not a statistical difference between tabular and calculated data.

By plotting experimental data for aqueous lactic acid solutions in TableCurve 3D® v.4 software (Figure 1) an equation for the response function was generated, chosen due to the accuracy and simplicity of formulation. The Equation 5 is a simple equation, Rank 33, Eqn. 1033 in TableCurve 3D® v.4 library with a precision of R^2 = 0.999967394, FitSdErr =

1.2166463, Fstat. = 20950.495. The coefficients values are presented in Table 7.

$$\ln\rho = \frac{a_1 + a_2 \cdot C + a_3 \cdot T}{1 + a_4 \cdot C + a_5 \cdot T + a_6 \cdot T^2 + a_7 \cdot T^3} \quad (5)$$

Table 6.
The ANOVA test summary

SUMMARY	Lactic acid concentration, *C* [% w/w]						Total
	9.16	24.35	45.48	64.89	75.33	85.32	
Experimental data							
Count	8	8	8	8	8	8	48
Sum	8056.42	8338.99	8713.71	9064.73	9239.16	9397.9	52810.91
Average	1007.053	1042.374	1089.214	1133.091	1154.895	1174.738	1100.227
Variance	126.4038	153.4055	271.2956	373.6544	387.4041	397.5284	3960.208
Calculated data							
Count	8	8	8	8	8	8	48
Sum	8055.094	8338.821	8721.028	9059.337	9236.239	9402.199	52812.72
Average	1006.887	1042.353	1090.128	1132.417	1154.53	1175.275	1100.265
Variance	116.92	174.5367	260.073	337.1056	375.7415	409.9365	3956.772

ANOVA						
Source of Variation	*SS**	*df**	*MS**	*F*	*P-value**	*F crit*
Sample	0.034015	1	0.034015	0.000121	0.991263	3.954568
Columns	348403.1	5	69680.62	247.094	1.13E-48	2.323126
Interaction	6.930834	5	1.386167	0.004915	0.999995	2.323126
Within	23688.04	84	282.0004			

* *SS* – sum of squares, *df* – degrees of freedom, *MS* – mean square, *P-value* – level of significance

Table 7.
Coefficients for equation no. 5

Coefficient	*Value*	Coefficient	*Value*
a_1	1004.2716	a_5	0.0032418557
a_2	3.6820806	a_6	-8.8833524E-07
a_3	2.8476715	a_7	3.4556043E-08
a_4	0.000950369	R^2	0.9999

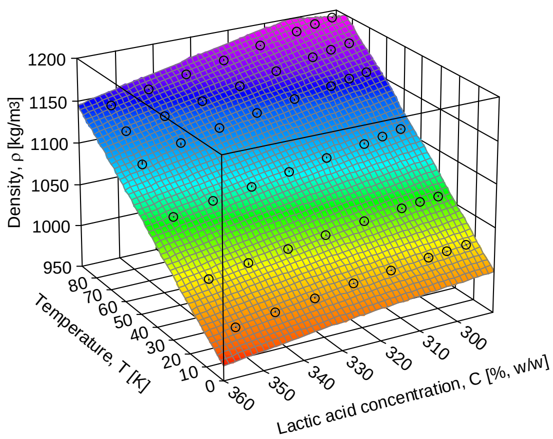

Figure 1. Lactic acid aqueous solutions density values plotted in TableCurve 3D and fitted with simple equation type (Equation 5) with residuals

2. Dynamic viscosity

A common mathematical model, for fitting the viscosity values is based on the equation of Arrhenius because it creates a good correlation between experimental and calculated values.

$$\mu = \mu_0 \cdot e^{-\frac{E_a}{R \cdot T}} \quad (6)$$

where:

μ – dynamic viscosity [Pa·s,], μ_0 – water dynamic viscosity [Pa·s,], *Ea* – activation energy [kcal/mol], *R* – universal gas constant [1.987×10^{-3} kcal/mol],

T – absolute temperature [K].

Taking logs of equation (6), it gets (7):

$$\log \mu = \log \mu_0 - \frac{E_a}{2.303 \cdot R} \cdot \frac{1}{T} \qquad (7)$$

and:

$$\frac{E_a}{R} = 2.303(\log \mu_0 - \log \mu) \cdot T \qquad (8)$$

By plotting in TableCurve 3D® v.4 software the results obtained from Equation 8, Figure 2, which make correlations between experimental data from Table 1 and water viscosity an equation for the response surface was generated (Equation 9). The Equation 9 is a polynomial equation, Rank 29, Eqn. 1049 in TableCurve 3D® v.4 library with a precision of R^2 = 0.9998, FitSdErr = 4.6371739, Fstat. = 36032.

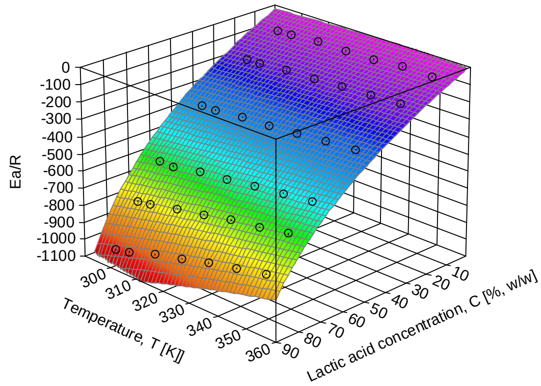

Figure 2. *Ea/R* values plotted in TableCurve 3D and fitted with polynomial equation type (Equation 9) with residuals

$$\frac{E_a}{R} = \frac{a_1 + a_2 \cdot C + a_3 \cdot C^2 + a_4 \cdot T}{1 + a_5 \cdot C + a_6 \cdot T} \qquad (9)$$

Table 8.
Coefficients for equation no. 9

Coefficient	Value	Coefficient	Value
a_1	-230.01384	a_4	0.67832811
a_2	-9.7502949	a_5	-0.01242596
a_3	0.044803796	a_6	0.0019299232

Combining the equations 6 and 9 and replacing the coefficients with numeric values, the final form of proposed equation model (Equation 10).

$$\mu = \mu_0 \cdot e^{-\frac{a_1 + a_2 \cdot C + a_3 \cdot C^2 + a_4 \cdot T}{(1 + a_5 \cdot C + a_6 \cdot T)T}} \qquad (10)$$

Appling the relative error equation the calculated data generated with the dynamic viscosity final equation and the existing tabular data were compared (Table 9) obtaining a final average of 1.11%.

Table 9.
For dynamic viscosities of citric acid aqueous solutions the absolute value of relative errors for calculated data versus tabular data

Temperature, *T* [K]	**Dynamic viscosity, $\mu \cdot 10^3$ [Pa · s]**								
	Lactic acid concentration, *C* [% w/w]								
	9.16		ε, %	24.35		ε, %	45.48		ε, %
	ED*	CD*		ED	CD		ED	CD	
298.15	1.15	1.16	0.83	1.67	1.68	0.39	3.09	3.14	1.66
303.15	1.03	1.02	0.62	1.46	1.47	0.35	2.74	2.69	1.77
313.15	0.81	0.82	1.45	1.13	1.15	1.98	2.03	2.04	0.53
323.15	0.67	0.67	0.32	0.92	0.92	0.51	1.59	1.58	0.64
333.15	0.57	0.56	2.45	0.75	0.76	1.51	1.26	1.26	0.24

Temp., *T* [K]	**Dynamic viscosity, $\mu \cdot 10^3$ [Pa · s]**								
	Lactic acid concentration, *C* [% w/w]								
	9.16		ε, %	24.35		ε, %	45.48		ε, %
	ED*	CD*		ED	CD		ED	CD	
343.15	0.47	0.47	0.31	0.63	0.63	0.35	1.02	1.02	0.31
353.15	0.42	0.41	1.83	0.53	0.54	1.17	0.84	0.85	0.34
	Average ε, %		1.11	Average ε, %		0.89	Average ε, %		0.78
	Lactic acid concentration, *C* [% w/w]								
	64.89		ε, %	75.33		ε, %	85.32		ε, %
	ED	CD		ED	CD		ED	CD	
298.15	6.96	7.00	0.51	13.03	12.84	1.46	28.50	28.80	1.04
303.15	6.01	5.82	3.21	10.55	10.39	1.47	22.60	22.37	1.01
313.15	4.22	4.17	1.13	7.08	7.10	0.26	13.91	14.18	1.95
323.15	3.12	3.07	1.54	4.98	5.00	0.46	9.40	9.36	0.41
333.15	2.38	2.34	1.88	3.57	3.66	2.45	6.40	6.46	0.96
343.15	1.85	1.82	1.42	2.73	2.76	1.02	4.59	4.63	0.89
353.15	1.47	1.45	1.24	2.08	2.13	2.27	3.40	3.41	0.43
	Average ε, %		1.56	Average ε, %		1.34	Average ε, %		0.95

* ED – experimental data, CD – calculated data

The ANOVA analysis was used to compare the values of dynamic viscosity tabular and calculated data at 10 different concentrations in 4 temperatures variation. The results presented in Table 10 showed that the sample *P*-value is 0.9997 greater than the targeted alpha 0.05 and the *F crit* value is larger than the *F*-test value and as consequence the null hypothesis is not rejected indicating that is not a statistical difference between tabular and calculated data.

Table 10.
The ANOVA test summary (for dynamic viscosity, *μ* [Pa · s])

SUMMARY	**Lactic acid concentration, *C* [% w/w]**						**Total**
	9.16	24.35	45.48	64.89	75.33	85.32	
Experimental data							
Count	7	7	7	7	7	7	42.00
Sum	0.005121	0.007088	0.012573	0.02601	0.04402	0.0888	0.183612
Average	0.000732	0.001013	0.001796	0.003716	0.006289	0.012686	0.004371714
Variance	7.76E-08	1.84E-07	7.43E-07	4.45E-06	1.73E-05	9.2E-05	3.46139E-05
Calculated data							
Count	7	7	7	7	7	7	42
Sum	0.005114	0.007146	0.012579	0.025668	0.043878	0.089219	0.18360409
Average	0.000731	0.001021	0.001797	0.003667	0.006268	0.012746	0.004371526
Variance	8.02E-08	1.84E-07	7.51E-07	4.4E-06	1.65E-05	9.27E-05	3.47515E-05

ANOVA						
Source of Variation	*SS**	*df**	*MS**	*F*	*P-value**	*F crit*
Sample	7.45E-13	1	7.45E-13	3.9E-08	0.999843	3.973897
Columns	0.001468	5	0.000294	15.3644	2.93E-10	2.341828
Interaction	2.26E-08	5	4.51E-09	0.000236	1	2.341828
Within	0.001376	72	1.91E-05			

* *SS* – sum of squares, *df* – degrees of freedom, *MS* – mean square, *P-value* – level of significance

By plotting directly the tabular data for the dynamic viscosity in TableCurve 3D® v.4 software an equation for the response surface was generated (Figure 3).

The Equation 11 is a linear equation, Rank 3, Eqn. 1071 in TableCurve 3D® v.4 library with a precision of R^2 = 0.99973191, FitSdErr = 0.00010903979, Fstat. = 13258.82 and the coefficients are presented in table 11.

$$\mu=\frac{b_1+b_2\cdot C+b_3\cdot C^2+b_4\cdot T+b_5\cdot T^2+b_6\cdot T^3}{1+b_7\cdot C+b_8\cdot C^2+b_9\cdot C^3+b_{10}\cdot T} \quad (11)$$

Table 11.
Coefficients for equation no. 11

Coefficient	Value	Coefficient	Value
b_1	0.1095492	b_6	-2.4739564E-09
b_2	-4.7563422E-06	b_7	-0.027799358
b_3	1.9479985E-08	b_8	0.00022424967
b_4	-0.00092119128	b_9	-6.1981778E-07
b_5	2.6073585E-06	b_{10}	0.00051576641

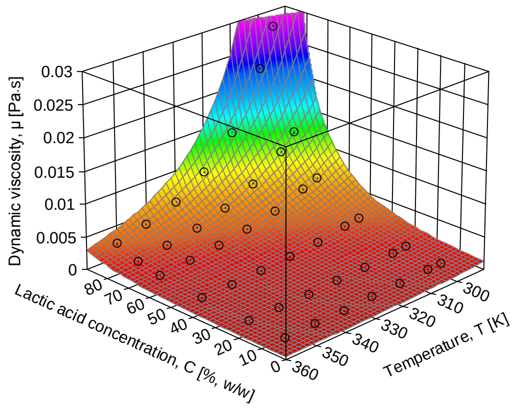

Figure 3. Lactic acid aqueous solutions dynamic viscosity values plotted in TableCurve 3D and fitted with linear equation type (Equation 11) with residuals

Combining the models developed for the calculation of dynamic viscosity and density of citric acid aqueous solutions, the kinematic viscosity (ν) can be calculated using equation 12:

$$\nu=\frac{\mu}{\rho}\ [m^2/s] \quad (12)$$

4. Conclusion

For density two equations were formulated with average relative errors (absolute value) of 0.062% (Equation 4) and respectively R^2 = 0.9999 and average relative errors of 0.055% (Equation 5) for intervals of temperature of 298.15 to 353.15 K and dry matter concentration range between 9.16 to 85.32%.

For the dynamic viscosity an equation based on Arrhenius mathematical model with average relative errors of 1.11% (Equation 10) and for a direct fitting of the experimental data in TableCurve 3D an equation with R^2 = 0.9997 were generated in the same range of temperature and citric acid concentration.

The proposed mathematical models can be loaded in the widespread PC software for storing, organizing and manipulating data and for targeted concentrations and temperature more precise values of the studied thermophysical properties can be found easier than using the existing experimental data in tabular form or graphic form.

5. References

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