Studii şi Cercetări Ştiinţifice
Chimie şi Inginerie Chimică, Biotehnologii, Industrie Alimentară

2016, **17**(4), pp. 417 – 426

Scientific Study & Research
Chemistry & Chemical Engineering, Biotechnology, Food Industry

ISSN 1582-540X

ORIGINAL RESEARCH PAPER

MATHEMATICAL MODELLING OF $CaCl_2$ AQUEOUS SOLUTIONS THERMOPHYSICAL PROPERTIES

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Received: December, 5, 2016
Accepted: December, 15, 2016

Abstract: Calcium chloride is an inorganic salt used in its solid state or as aqueous solutions in a multitude of research and industrial areas going from chemical industry to food, cosmetics or drugs production.
This paper was directed to establish mathematical models relating its density and dynamic viscosity with factors such as temperature and concentration since it is known that these properties present a high influence on appropriate transport pipes dimensions and industrial equipment choosing and / or designing processes.
The assessment and selection of the adequate models were realized by testing various software's and equations. The obtained results revealed that calcium chloride aqueous solutions studied thermodynamic properties can accurately be expressed by quadratic (density) and respectively by linear equations (dynamic viscosity) or by more complex equations. Correlation coefficient, relative error and ANOVA values revealed no significant differences between experimental and calculated data.

Keywords: *calcium chloride, density, mathematical model, viscosity*

INTRODUCTION

Calcium chloride is a nontoxic inorganic salt which can be easily obtained at accessible prices. It is characterized by a high hygroscopicity and an important sorption capacity. It has also a good thermal conductivity, a high fusion latent heat and a low volume modification in phase transition [1].
Its properties make it suitable for uses in various fields such as chemistry, food products, cosmetics etc. In chemical industry, pure calcium chloride is used for producing different compounds such as calcium carbonate, calcium phosphate or calcium chromate [2, 3]. In food industry, this salt can be incorporated in coating materials [4 – 7]; it can reduce the non-desirable storage effects (softening, discoloration etc.) of fruits and vegetable by offering a good protection against enzymes action [5, 6, 8, 9]; it enhance meat products properties [10, 11] etc. In cosmetic production processes, calcium chloride, mixed with other ingredients, can be used for example for bigels manufacturing [12]. In pharmaceutics development, calcium chloride presents synergic effects on *in vitro* drugs skin permeation [13] or it can serve to formulate products aimed to improve oral delivery of low soluble drugs [14]. Due to its cooling properties, calcium chloride is also used to obtain selective water composite adsorbents [15, 16], liquid desiccants [17 – 19] or cooling agents [1].
In many of the above mentioned utilizations examples, calcium chloride is often employed as aqueous solutions whose thermophysical properties (namely density, dynamic viscosity, thermal conductivity etc.) present high importance in the appropriate transport pipes dimensions and industrial equipment choosing and / or designing processes.
As consequence, over the time, several studies were aimed to establish values for the cited calcium chloride properties and to correlate them with various parameters such as pressure, temperature, solute concentration different data being nowadays available [20 – 22]. These data are frequently expressed in tabular or graphical form fact that makes their employment rather complicate. A more elegant, adequate and easy to implement method consist in transforming the existing data in mathematical equations.
Considered a suitable alternative in different other areas such as juice production [23], study of flour and dough rheological properties [24], study of dough dynamic viscosity evolution [25] etc., this data interpretation way was successfully applied also to other fluids thermodynamic properties studies like aqueous solutions of sodium chloride [26], glucose [27] or glycerol [28, 29] etc.
Therefore, the present work was focused on the study of the behavior of calcium chloride aqueous solutions density and dynamic viscosity both known as significant in characterizing the behavior when passing through pipes as well as in heat and mass transfer in many processing processes.
Data from existing publications were used for establish mathematical correlations for these properties when influenced by parameters such as temperature (both in positive and negative ranges) and solutions concentration.
The obtained equations have a high similarity with the experimental data fact that confirms the accuracy of the mathematical models.

MATERIALS AND METHODS

Data shown on Tables 1 – 3 provided by different scientific publications [30] on density and dynamic viscosity evolution of aqueous calcium chloride solutions with their concentration and temperature were introduced in various software's in order to establish accurate mathematical equation.

Table 1. *Variation of $CaCl_2$ aqueous solutions density with temperature and concentration [30]*

Concentration, $C_{\%}$ [%, w/w]	Density, ρ [kg·m^{-3}] Temperature, T [K]									
	268.15	***273.15***	***293.15***	***303.15***	***313.15***	***333.15***	***353.15***	***373.15***	***393.15***	***413.15***
2	-	1017.1	1014.8	1012	1008.4	999.4	988.1	974.8	959.6	942.8
4	-	1034.6	1031.6	1028.6	1024.9	1015.8	1004.6	991.5	976.5	960.1
8	1070.8	1070.3	1065.9	1062.6	1058.6	1049.2	1038.2	1025.7	1011.1	995.4
12	1108.3	1107.2	1101.5	1097.8	1093.7	1084.0	1073.0	1061.0	1046.6	1031.7
16	1147.1	1145.4	1138.6	1134.5	1130.1	1120.2	1109.2	1097.3	1083.5	1069.1
20	1187.4	1185.3	1177.5	1173.0	1168.4	1158.1	1147.1	1135.2	1121.9	1108.0
25	-	1237.6	1228.4	1223.6	1218.6	1207.9	1196.5	1184.6	-	-
30	-	1292.2	1281.6	1276.4	1270.9	1259.7	1247.8	1235.9	-	-
35	-	-	1337.3	1331.6	1325.5	1313.7	1301.3	1289.3	-	-
40	-	-	1395.7	1389.5	1382.6	1370	1357.1	1345	-	-

Table 2. *Variation of $CaCl_2$ aqueous solutions density with temperature and concentration [30]*

Concentration, $C_{\%}$ [%, w/w]	Density, ρ [kg·m^{-3}] Temperature, T [K]					
	288.15	***273.15***	***263.15***	***253.15***	***243.15***	***233.15***
15	1132	1137	1140	-	-	-
16	1142	1147	1150	-	-	-
17	1151	1157	1160	-	-	-
18	1161	1167	1170	-	-	-
19	1171	1177	1180	-	-	-
20	1181	1187	1190	-	-	-
21	1191	1197	1201	1205	-	-
22	1201	1207	1211	1215	-	-
23	1211	1218	1222	1226	-	-
24	1222	1228	1233	1237	-	-
25	1232	1239	1244	1248	-	-
26	1243	1250	1254	1259	1263	-
27	1252	1261	1266	1270	1275	-
28	1264	1272	1277	1282	1287	-
29	1275	1283	1288	1293	1298	1303
30	1286	1294	1298	1304	1310	1315

Microsoft Excel™ 2013, CurveExpert® and TableCurve 3D® v.4 software's were employed for mathematical models development and for data 2D and 3D plotting.

Table 3. *Variation of $CaCl_2$ aqueous solutions dynamic viscosity with temperature and concentration [30]*

Concentration, $C_{\%}$ [%, w/w]	**Dynamic viscosity, μ [mPa·s]**												
	Temperature, T [K]												
	293.15	***283.15***	***273.15***	***268.15***	***263.15***	***258.15***	***253.15***	***248.15***	***243.15***	***238.15***	***233.15***	***228.15***	***223.15***
9.4	12.36	15.49	21.58	25.48	-	-	-	-	-	-	-	-	-
14.7	14.91	18.63	25.6	30.4	40.6	-	-	-	-	-	-	-	-
18.9	18.04	22.36	29.91	34.32	46.68	61.49	-	-	-	-	-	-	-
20.9	20.01	24.52	32.75	38.25	50.7	65.9	-	-	-	-	-	-	-
23.8	23.54	28.73	38.15	44.13	59.23	75.51	94.73	115.72	-	-	-	-	-
25.7	26.28	32.17	42.56	-	66.78	83.65	105.62	129.15	148.08	-	-	-	-
27.5	29.32	36.09	48.05	-	75.22	-	118.66	147.1	171.61	215.74	-	-	-
28.4	31.38	-	51.19	-	80.22	-	126.51	159.85	188.29	245.17	304.01	-	-
29.4	34.03	-	54.92	-	86.3	-	138.27	-	212.8	254.97	323.62	402.07	490.33
29.9	35.11	-	56.88	-	90.43	-	144.16	-	225.55	284.39	353.04	431.49	509.95

RESULTS AND DISCUSSION

Density

10 different second order polynomial simple correlations (Equation 1) have been developed on Microsoft Excel software by representing density ρ [kg·m^{-3}] values as temperature T [K] functions at constant calcium chloride concentrations $C_{\%}$ [% w/w].

$$\rho = A_1 + A_2 T + A_3 T^2 \qquad (1)$$

The A_1, A_2 and A_3 coefficients values are presented in Table 4. For all of them, the regression coefficients R^2 are superior of 0.999 indicating a good correlation of variables.

Table 4. *Coefficients for equation no. 1*

Concentration, $C_{\%}$ [%, w/w]	**Equation 1 coefficients**			
	A_1	A_2	A_3	R^2
2	839.99	1.4414	-0.0029	0.9994
4	879.7	1.3054	-0.0027	0.9996
8	950.78	1.084	-0.0024	0.9997
12	1034.3	0.8028	-0.002	0.9999
16	1116.6	0.5376	-0.0016	1
20	1196	0.3018	-0.0012	1
25	1291.6	0.0461	-0.0009	1
30	1393.9	-0.2327	-0.0005	1
35	1494.9	-0.4864	-0.0002	1
40	1616.8	-0.846	0.0003	0.9999

A_1, A_2 and A_3 coefficients were associated with concentrations $C_{\%}$ [% w/w] by using CurveExpert® software. Quadratic equation for A_1 (Equation 2) and linear equation

(Equation 2) for A_2 and A_3 coefficients with good regression coefficients (Table 5) have been established.

$$Coefficient = a_1 + a_2 C_{\%} + a_3 C_{\%}^2 \tag{2}$$

$$Coefficient = a_1 + a_2 C_{\%} \tag{3}$$

Table 5. *Coefficients for equations no. 2 and 3*

Equation 1 coefficient	**Equations 2 and 3 coefficients**			
	a_1	a_2	a_3	R^2
A_1	803.55498	18.65180	0.03781	0.99971
A_2	-0.05916	1.53134	-	0.99827
A_3	- 0.00301	0.00008	-	0.99571

Equations 1, 2 and 3 were combined in order to obtain the general model expressed by Equation 4:

$$\rho = (a_1 + a_2 \cdot C_{\%} + a_3 \cdot C_{\%}{}^2)_{A_1} + (a_1 + a_2 \cdot C_{\%})_{A_2} \cdot T + (a_1 + a_2 \cdot C_{\%})_{A_3} \cdot T^2 \tag{4}$$

A parallel between data calculated with the mathematical model and the experimental ones was realized by the mean of relative error equation (5).

$$\varepsilon = \left| \frac{Data_{\text{experimental}} - Data_{calculated}}{Data_{calculated}} \right| \cdot 100[\%] \tag{5}$$

An average of 0.630% (in absolute value) was obtained. The regression coefficient R^2 of the proposed model was 0.9946 for densities values presented in Table 1 and higher (0.9999) for those given in Table 2. In both cases the absolute value of the relative error was of 0.524%.

The ANOVA (Two-Factor with Replication) test was used also to compare tabular and calculated density values. The results revealed sample *P-values* of 0.900567 and 0.941336, greater than the targeted alpha 0.05, and *Fcrit* values of 3.89364 superior to *F-test* values (0.015655 and 0.005431) for calcium chloride aqueous solutions density values variation in positive (Table 1) and negative (Table 2) temperature ranges. In this case, the null hypothesis cannot be rejected and no statistical difference between tabular and calculated data was registered.

Density data were introduced in TableCurve 3D® v.4 software which served to generate two different polynomial equations (6 and 7): one classified Rank 49 (Eqn. 313) with a precision of R^2 = 0.9999966601, *FitSdErr* = 0.2326931809, *Fstat.* = 2.46181E+06 for values shown in Table 1 and one of Rank 16 (Eqn. 310) with R^2 = 0.9999560422, *FitSdErr* = 0.3701979907, *Fstat.* = 139015.96133 for values exposed in Table 2.

$$\begin{aligned} \rho = {} & b_1 + b_2 \cdot C_{\%} + b_3 \cdot \ln(T) + b_4 \cdot C_{\%}^2 + b_5 \cdot (\ln T)^2 + b_6 \cdot C_{\%} \cdot \ln(T) + b_7 \cdot T^3 + \\ & + b_8 \cdot (\ln T)^3 + b_9 \cdot C_{\%} \cdot (\ln T)^2 + b_{10} \cdot C_{\%}{}^2 \cdot \ln T \end{aligned} \tag{6}$$

$$\begin{aligned} \rho = {} & b_1 + b_2 \cdot C_{\%} + b_3 \cdot T + b_4 \cdot C_{\%}^2 + b_5 \cdot T^2 + b_6 \cdot C_{\%} \cdot T + b_7 \cdot C_{\%}{}^3 + b_8 \cdot T^3 + \\ & + b_9 \cdot C_{\%} \cdot T^2 + b_{10} \cdot C_{\%}{}^2 \cdot T \end{aligned} \tag{7}$$

Coefficients of the equations 6 and 7 are presented in Tables 9 and 10.

Table 9. *Coefficients for equation 6*

Coefficient	Value	Coefficient	Value
b_1	10406.05291	b_6	-109.516623
b_2	325.6324771	b_7	6.15222E-05
b_3	-7007.51748	b_8	-116.551335
b_4	0.203129712	b_9	9.436791193
b_5	1604.133205	b_{10}	-0.02778971

Table 10. *Coefficients for equation 7*

Coefficient	Value	Coefficient	Value
b_1	810.4786129	b_6	-0.09129409
b_2	22.6640109	b_7	-0.00028682
b_3	1.070837953	b_8	-5.4388E-06
b_4	0.05461806	b_9	0.000134079
b_5	0.000191949	b_{10}	5.71015E-05

The graphical representation of the equations 6 and 7 is presented in Figure 1.

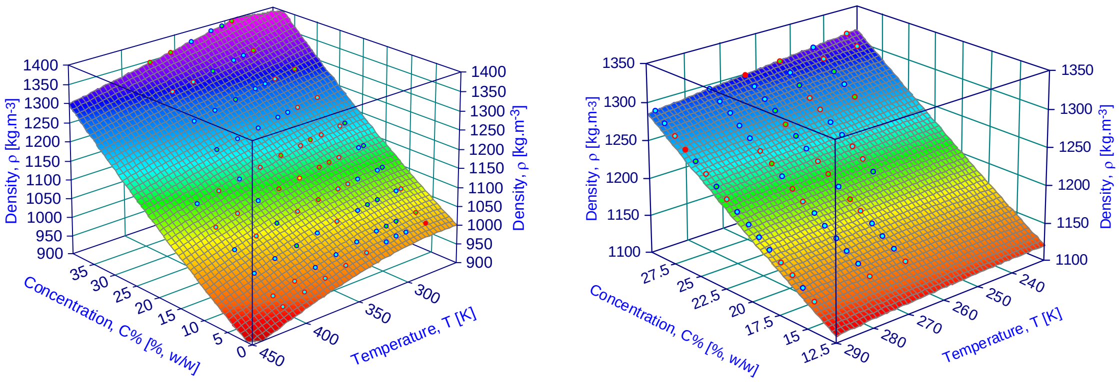

Figure 1. *$CaCl_2$ aqueous solutions density values plotted in TableCurve 3D and fitted with polynomial type (Equations 6 and 7) and its residuals*

Dynamic viscosity

The same calculus algorithm was followed for calcium chloride aqueous solutions dynamic viscosity data. In this case, 10 quadratic correlations expressed by equation 8 have been established by plotting the logarithmic values of dynamic viscosity μ [mPa·s] vs temperature T [K] at fixed $CaCl_2$ concentrations.

$$log(\mu) = A_1 + A_2T + A_3T \tag{8}$$

Table 10 depicts A_1, A_2 and A_3 coefficients values.
A_1, A_2 and A_3 coefficients were correlated with $CaCl_2$ concentrations $C_{\%}$ [% w/w], and various mathematical models (1^{st}, 2^{nd} and 3^{rd} degree polynomial equations, "vapour pressure" model, "heat capacity" model etc.) were generated.
Among them the best fit was given by the quadratic equation 9 whose coefficients values are shown in Table 11.

$$Coefficient = a_1 + a_2C_{\%} + a_3C_{\%}^2 \tag{9}$$

***Table 10.** Coefficients for equation 8*

Concentration, $C_{\%}$ [%, w/w]	Equation 8 coefficients			
	A_1	A_2	A_3	R^2
9.4	0.000165	0.10509	17.75579	0.9992
14.7	0.000152	0.09875	17.01874	0.9974
18.9	0.000137	0.09013	15.87924	0.9993
20.9	0.000135	0.08849	15.65616	0.9971
23.8	0.000113	0.07694	14.16871	0.9978
25.7	0.000094	0.06653	12.79728	0.9966
27.5	0.000076	0.05687	11.56793	0.9968
28.4	0.000066	0.05173	10.98032	0.9972
29.4	0.000054	0.04563	10.18695	0.9948
29.9	0.000048	0.0423	9.79841	0.9955

***Table 11.** Coefficients for equation 9*

Equation 8 coefficient	Equation 9 coefficients			
	a_1	a_2	a_3	R^2
A_1	0.0001293054	0.0000063231	-0.0000003008	0.9940
A_2	0.0869560277	0.0032994350	-0.0001596278	0.9956
A_3	-0.0203417811	0.4191380278	15.4856071276	0.9962

The last mentioned two equations (8 and 9) were combined leading to the following final model:

$$log(\mu) = (a_1 + a_2 \cdot C_{\%} + a_3 \cdot C_{\%}{}^2)_{A_1} + (a_1 + a_2 \cdot C_{\%} + a_3 \cdot C_{\%}{}^2)_{A_2} \cdot T + + (a_1 + a_2 \cdot C_{\%} + a_3 \cdot C_{\%}{}^2)_{A_3} \cdot T^2 \quad (10)$$

The error of this model was of 0.897% (2.488% in absolute value) and its regression coefficient was of 0.996.

The ANOVA test revealed a sample *P-value* of 0.9647 (superior to targeted alpha of 0.05) and an *Fcrit* value of 3.881505 (larger than the *F-test* value of 0.001956). The same conclusion as for density values analysis was drawn, namely that there is no statistical difference between tabular and calculated data.

Multiple equations were generated also with TableCurve 3D® v.4 software but only one was selected due to its high precision (Rank 33, Eqn. 254122972). This equation has a correlation coefficient of 0.9990438298 and its *FitSdErr* and *Fstat.* values were 6922.0578888 and 3.2745062671 respectively. The established equation (11) has the coefficients presented in Table 14 and the graphical form pictured in Figure 2.

$$ln(\mu) = b_1 + b_2 \cdot C_{\%}^2 \cdot ln(C_{\%}) + b_3 \cdot C_{\%}{}^{2.5} + b_4 \cdot C_{\%}{}^3 + b_5 \cdot T + b_6 \cdot T^{0.5} \cdot ln(T) + + b_7 \cdot ln(T)^2 + b_8 \cdot T^{0.5} + b_9 \cdot ln(T) \quad (11)$$

***Table 14.** Coefficients for equation 11*

Coefficient	Value	Coefficient	Value
b_1	-1.3076e+06	b_6	-76142.224
b_2	0.028660749	b_7	-142669.246
b_3	-0.02505711	b_8	716967.2378
b_4	0.001365614	b_9	117931.3782
b_5	1268.065255		

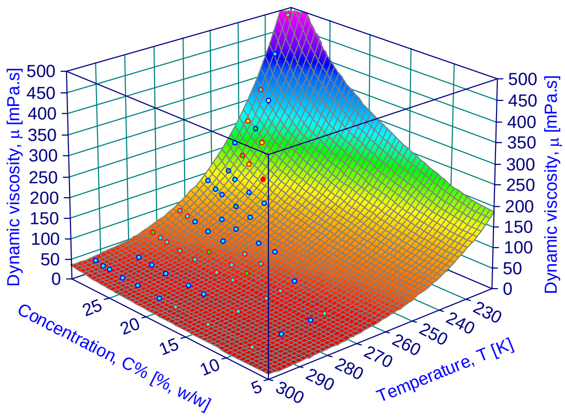

Figure 2. *$CaCl_2$ aqueous solutions dynamic viscosity values plotted in TableCurve 3D and fitted with linear type (Equation 11) and its residuals*

Kinematic viscosity

Using the data obtained for density and dynamic viscosity of calcium chloride aqueous solutions, with the help of equation 12, one can calculate also the kinematic viscosity (v).

$$v = \frac{\mu}{\rho} \ [m^2 \cdot s^{-1}] \qquad (12)$$

Compared to results presented in other studies on the same thematic [20, 21], the mathematical models developed in this work present a higher degree of accuracy. In some cases, they are also simpler and therefore easier to use.

CONCLUSION

This paper was able to establish mathematical models relating density and dynamic viscosity of calcium chloride aqueous solutions with their temperature and concentration. The results released with the help of Excel Microsoft software showed that a second order polynomial equation can be considered a suitable mathematical expression of density while the dynamic viscosity can be presented as a linear model. More complex equations were obtained with CurveExpert and TableCurve 3D softwares. Correlation coefficient, relative error and ANOVA values revealed no significant differences between the experimental and calculated data. The high degree of accuracy registered for all the mathematical models developed in this study recommends their use for data base enrichment but also for equipment and processes sizing and optimization.

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