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ORIGINAL RESEARCH PAPER

MODELING OF THE THERMO-PHYSICAL PROPERTIES OF GRAPES JUICE III. VISCOSITY AND HEAT CAPACITY♦

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Abstract: When optimizing the process of grapes juice manufacture not just to obtain an excellent quality, but also to develop a data base is essential to know the evolution of physical properties, such as viscosity or heat capacity elevation. These properties are affected by temperature and dry matter content. The aim of this study was to establish a mathematical relation between these variables. In order to assess and select a suitable mathematical model the known data were fitted in different equations. Tests results have shown that at constant dry matter content the temperature and the viscosity are related by an exponential equation while between temperature and heat capacity variation a "heat capacity" model seems to be the most appropriate.

Keywords: *food industry, grapes juice, mathematical modeling, thermo-physical properties*

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INTRODUCTION

The fruit juice processing industry is one of the world's major agro-based businesses. The processing of fruit into juice products requires knowledge of the interplay of many variables. Rationally designed equipment needs a thoroughly knowledge of physical and thermodynamic properties of materials involved: raw materials, additives, intermediate and finished products.
Knowledge at any time of the thermo physical properties, such as viscosity and heat capacity variation, is important not only for process design but also for the prediction and control of various changes that occur in juice during processing [1].
A major part of the existing data is presented in graphical form and they are improper for use in algorithms and mathematical models of spread sheet type.
This paper presents simple and reliable mathematical relationships between the grapes juice viscosity respectively heat capacity variation with temperature and dry matter content.

MATERIALS AND METHODS

Different experimental extrapolated equations (Equations 1-5) and graphic representations (Figure 1 [2]), presented in literature, concerning the variation of viscosity and heat capacity of grapes juice with dry matter and temperature were used as primary data for the regression analysis [2, 3]. These data were selected due to their wide use in research, development and design of the grapes juice processing industry.
The most common mathematical model given in literature [4-6], for calculating the viscosity of grapes juice is based on Arrhenius equation:

$$\mu = \mu_0 \cdot e^{-\frac{E_a}{R \cdot T}} \tag{1}$$

where: μ – viscosity [mPa·s, cP]; μ_0 – constant [mPa·s, cP]; E_a – activation energy [kcal/mol]; R – universal gas constant [1.987×10^{-3} kcal/mol]; T – absolute temperature [K].
Two mathematical models based on Arrhenius equation that provides the best correlation between experimental data and calculated values of viscosity of grapes juice whit these, are the model of Zuritz et al. [3] combined with equation (1):

$$\mu_0 = -\exp(2.0365 - 2.0933 \cdot 10^{-02} \cdot {}^{\circ}Bx + 4.3614 \cdot 10^{-04} \cdot {}^{\circ}Bx^2) \tag{2}$$

$$E_a / R = -\exp(7.6346 - 1.0455 \cdot 10^{-02} \cdot {}^{\circ}Bx + 3.6897 \cdot 10^{-04} \cdot {}^{\circ}Bx^2) \tag{3}$$

where: °Bx – degrees Brix [g sugar/100 g solution], and Bayindirli model [4]:

$$\mu = \mu_{water} \cdot \exp \frac{[-0.24 + (1821.45/T)^{\circ}Bx]}{100 - [(0.86 + 0.000441 \cdot T)^{\circ}Bx]} \tag{4}$$

In order to calculate heat capacity of grapes juice, an equation with a good accuracy of calculation is [3]:

$$C_p = 1386 + 21.6 \cdot (100 - X) + 2 \cdot T \tag{5}$$

where: X represents dry matter content [%, w/w].

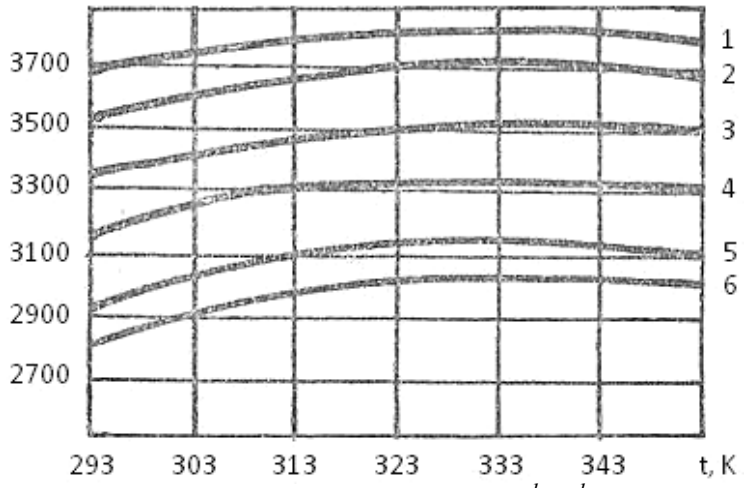


Figure 1. *Variation of grapes juice heat capacity ($J.kg^{-1}.K^{-1}$) with temperature (T) and water content (W): 1 – 85%, 2 – 80%, 3 – 70%, 4 – 60%, 5 – 50%, 6 – 42%* [2]

Because most of thermo-physical properties of grapes juice in the food industry are not presented as numerical data but in graphics forms, xyExtract Graph Digitizer.v2.3 software was used to extract numerical data from graphical representations.
The experimental data were plotted in *Temperature – Thermo-physical property, Dry matter content – Thermo-physical property* coordinates and linear regression techniques, involving the method of least squares were used to reveal the best-fit equation. Microsoft Excel™ 2007 spreadsheets, CurveExpert® and TableCurve 3D® v.4 software were used to establish the equations.

RESULTS AND DISCUSSIONS

Viscosity

Using Microsoft Excel™ 2007 spreadsheets and CurveExpert® software, a logarithmic correlation between temperature and viscosity, at constant dry matter content has been established:

$$\log_{10}(\mu) = A \cdot T + B \tag{6}$$

The *A* and *B* values are presented in Table 1. The regression coefficients are greater than 0.99, thus indicating a very good correlation of variables.
In order to correlate *A* and *B* coefficients with soluble solids content, more models were used in CurveExpert® software (1st, 2nd and 3rd degree polynomial equations, "vapor pressure" model, "heat capacity" model etc.). The best mathematical model final form is represented by the 2nd degree polynomial equation (Table 2):

$$Coefficient = a \cdot^{\circ} Bx^2 + b \cdot^{\circ} Bx + c \tag{7}$$

Combining the equations (6) and (7) and replacing the coefficients with numeric values in order to correlate the viscosity of grapes juice with temperature and soluble solids, the final form of the proposed model is:

$$\log_{10}(\mu) = (-7.9E-06\cdot^{\circ}Bx^2 + 0.00041498\cdot^{\circ}Bx - 0.013978429)\cdot T + \\ + 0.003047\cdot^{\circ}Bx^2 - 0.14689869\cdot^{\circ}Bx + 4.651530818 \quad (8)$$

Table 1. *Coefficients for equation (6)*

°Brix [g/100 g]	*A*	*B*	R^2
22.9	-0.00823	2.754363	0.9971
25.5	-0.00847	2.860423	0.9971
31.0	-0.00915	3.150672	0.9971
34.0	-0.00943	3.296351	0.9971
41.0	-0.01049	3.842687	0.9971
45.0	-0.01131	4.212523	0.9971
51.0	-0.01298	4.928617	0.9971
53.4	-0.01435	5.450880	0.9971
60.7	-0.01766	6.822610	0.9971
67.0	-0.02219	8.643872	0.9971

Table 2. *Coefficients for equation (7)*

Coefficient	*a*	*b*	*c*	R^2
A	-7.9E-06	0.000414	-0.01397	0.9943
B	0.003047	-0.14689	4.65153	0.9961

If measurement unit for temperature is changed in °C in the mathematical model, after following the same steps, equation (8) becomes:

$$\log_{10}(\mu) = (-8E-06\cdot^{\circ}Bx^2 + 0.0004\cdot^{\circ}Bx - 0.0139)\cdot t + \\ + 0.0009\cdot^{\circ}Bx^2 - 0.0336\cdot^{\circ}Bx + 0.8353 \quad (9)$$

To calculate the viscosity if is known the percentage of dry substance (X) instead of the value °Bx, the proposed mathematical model is equation (10) if the temperature is measured in °C and equation (11) if the temperature is measured in K.

$$\log_{10}(\mu) = (-8.53E-06\cdot X^2 + 0.000516\cdot^{\circ}X - 0.0164)\cdot t + \\ + 0.000972\cdot X^2 - 0.047867\cdot X + 1.1506 \quad (10)$$

$$\log_{10}(\mu) = (-8.522E-06\cdot X^2 + 0.0005177\cdot X - 0.0164344)\cdot T + \\ + 0.003298\cdot X^2 - 0.189222\cdot X + 5.63733 \quad (11)$$

To quantify the deviation from experimental data between measured and calculated viscosity, the relative error (12a) and average relative error (12b) were used:

$$\varepsilon = \left|\frac{\mu_{measured} - \mu_{calculated}}{\mu_{measured}}\right|\cdot 100 \quad [\%] \quad (12a)$$

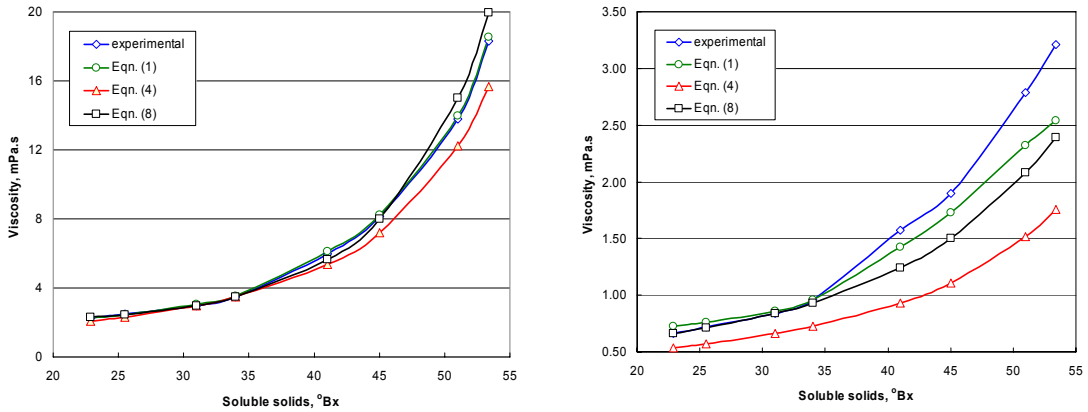
$$\tilde{\varepsilon} = \frac{1}{n}\sum_{i=1}^{n}\varepsilon_i \quad (12b)$$

Table 3. *Comparison between experimental measured values and calculated values with equation developed by Zuritz et al, Bayindirli and proposed model for viscosity of grapes juice*

Temperature, [K]	Soluble solids, °Bx [g/100 g]	Viscosity, μ [mPa·s]							
		Experimental data	calculated with equations:						
			(2), (3) in (1)		(4)		(8)		
			μ	ε [%]	μ	ε [%]	μ	ε [%]	
293	22.9	2.24	2.267	1.223	2.061	7.970	2.286	2.064	
	25.5	2.48	2.464	0.639	2.299	7.296	2.430	1.998	
	31.0	2.96	3.061	3.438	2.974	0.474	2.984	0.820	
	34.0	3.50	3.532	0.916	3.483	0.463	3.486	0.397	
	41.0	5.98	6.105	2.090	5.358	10.392	5.644	5.607	
	45.0	8.16	8.258	1.202	7.192	11.861	8.013	1.800	
	51.0	13.80	13.997	1.429	12.213	11.496	15.012	8.782	
	53.4	18.30	18.545	1.340	15.668	14.377	19.971	9.134	
Average relative error at 293 K, %				**1.5346**	**-**	**8.0411**	**-**	**3.8253**	
303	22.9	1.70	1.817	6.902	1.545	9.083	1.861	9.476	
	25.5	1.88	1.962	4.378	1.708	9.129	1.979	5.319	
	31.0	2.30	2.394	4.112	2.164	5.907	2.415	5.010	
	34.0	2.66	2.741	3.051	2.502	5.905	2.798	5.190	
	41.0	4.47	4.604	3.019	3.719	16.778	4.386	1.858	
	45.0	5.86	6.093	3.983	4.878	16.742	6.064	3.490	
	51.0	9.41	9.874	4.936	7.953	15.473	10.798	14.754	
	53.4	11.90	12.609	5.958	10.013	15.853	14.024	17.853	
Average relative error at 303 K, %				**4.5424**	**-**	**11.8588**	**-**	**7.8688**	
313	22.9	1.58	1.477	6.497	1.198	24.166	1.515	4.112	
	25.5	1.66	1.585	4.711	1.313	21.075	1.613	3.062	
	31.0	1.93	1.902	1.428	1.631	15.480	1.954	1.279	
	34.0	2.11	2.162	2.469	1.864	11.655	2.245	6.438	
	41.0	3.44	3.536	2.810	2.682	22.024	3.409	0.889	
	45.0	4.23	4.584	8.377	3.442	18.625	4.589	8.507	
	51.0	6.83	7.123	4.216	5.399	21.006	7.767	13.642	
	53.4	8.30	8.786	5.867	6.676	19.557	9.848	18.655	
Average relative error at 313 K, %				**4.5469**	**-**	**19.1985**	**-**	**7.0730**	
323	22.9	1.27	1.216	4.215	0.951	25.101	1.233	2.890	
	25.5	1.33	1.298	2.388	1.034	22.226	1.314	1.196	
	31.0	1.52	1.533	0.862	1.261	17.028	1.581	4.077	
	34.0	1.63	1.730	6.172	1.424	12.579	1.802	10.581	
	41.0	2.72	2.760	1.505	1.988	26.881	2.649	2.583	
	45.0	3.38	3.510	3.856	2.500	26.032	3.473	2.774	
	51.0	5.15	5.243	1.814	3.780	26.589	5.587	8.490	
	53.4	6.12	6.261	2.318	4.596	24.894	6.915	13.00	
Average relative error at 323 K, %				**2.8913**	**-**	**22.6663**	**-**	**5.6989**	

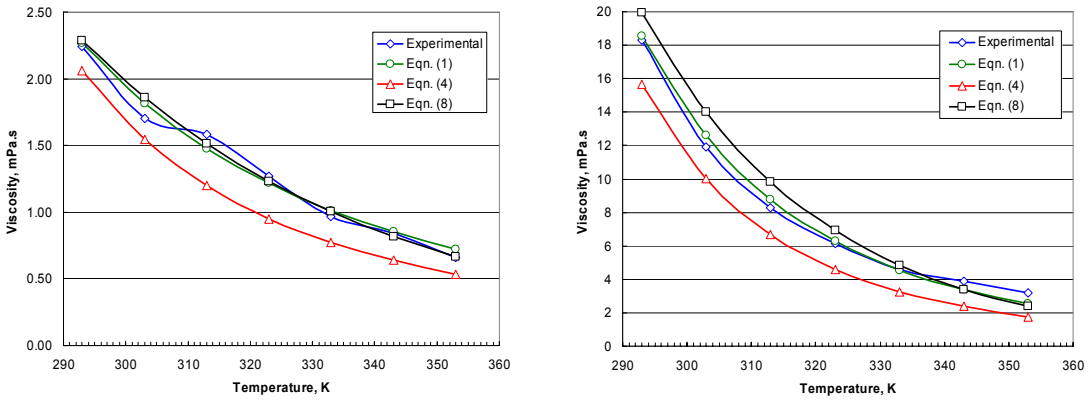
333	22.9	0.97	1.013	4.474	0.771	20.457	1.003	3.500
	25.5	1.09	1.075	1.304	0.832	23.596	1.070	1.784
	31.0	1.27	1.251	1.448	0.997	21.437	1.280	0.812
	34.0	1.39	1.403	0.997	1.115	19.761	1.440	4.089
	41.0	2.11	2.187	3.680	1.512	28.327	2.059	2.402
	45.0	2.66	2.731	2.683	1.864	29.915	2.629	1.163
	51.0	3.76	3.931	4.558	2.722	27.585	4.019	6.888
	53.4	4.61	4.554	1.210	3.257	29.343	4.856	5.345
Average relative error at 333 K, %				**2.5443**	-	**25.0526**	-	**3.2479**
343	22.9	0.84	0.853	0.622	0.638	24.681	0.817	3.625
	25.5	0.91	0.901	0.850	0.684	24.69	0.872	4.055
	31.0	1.03	1.033	0.383	0.806	21.685	1.036	0.601
	34.0	1.21	1.152	4.727	0.892	26.231	1.161	4.025
	41.0	1.81	1.757	2.922	1.177	34.941	1.600	11.577
	45.0	2.30	2.156	6.235	1.424	38.048	1.989	13.488
	51.0	3.39	2.997	11.575	2.013	40.610	2.890	14.721
	53.4	3.88	3.374	13.034	2.371	38.875	3.410	12.106
Average relative error at 343 K, %				**5.0435**	-	**31.2201**	-	**8.0248**
353	22.9	0.66	0.725	8.769	0.535	19.673	0.665	0.257
	25.5	0.72	0.762	4.907	0.570	21.539	0.710	2.269
	31.0	0.84	0.863	2.061	0.661	21.788	0.838	0.873
	34.0	0.96	0.957	1.007	0.725	25.009	0.932	3.609
	41.0	1.57	1.428	8.986	0.932	40.633	1.243	20.775
	45.0	1.90	1.725	9.173	1.108	41.683	1.505	20.741
	51.0	2.79	2.320	16.811	1.516	45.631	2.079	25.465
	53.4	3.21	2.542	20.782	1.760	45.150	2.394	25.396
Average relative error at 353 K, %				**9.0620**	-	**32.6383**	-	**12.4231**
Avg. rel. error at 293 – 353 K, %				**4.3093**	-	**21.5250**	-	**6.8802**

As shown in Table 3, greatest errors are given by Bayindirli equation (4), with an average value of 21.525%. The proposed model, depicted by equation (8) gives average relative errors below 10% at all temperatures excepting that of 353 K. The average relative error calculated for all temperatures is 6.88%, comparable to the 4.31% given by Zuritz et al. model (Eqns. 1-3).



a) T = 293 K b) T = 353 K

Figure 2. *Viscosity of grapes juice vs. soluble solids*



a) 22.9°Bx b) 53.4°Bx

Figure 3. *Viscosity of grapes juice vs. temperature*

At low concentrations of dry matter and low temperatures (Figures 2a and 3a) the Zuritz model (Eqns. 1-3) and the proposed model (Eqn. 8) are in good concordance with experimental data. At high dry solids and low temperatures (Figure 3b) the Zuritz model is closer to reality than other models. At low dry solids and high temperatures (Figure 2b and 3a) proposed model and Zuritz model have the same accuracy. In the field of high temperatures and high solids concentrations (Figure 2b and 3b) all models give relative errors greater than 10%.

By plotting experimental data for grapes juice viscosity in TableCurve 3D® v.4 software (Figure 4) two equations for the response function were generated, chosen due to the accuracy, respectively, the simplicity of formulation:

$$\mu = \frac{4.812689 + 0.3380235 \cdot \ln T + 1.6606316 \ln {}^{\circ}Bx - 0.24942578 \cdot (\ln {}^{\circ}Bx)^2}{1 - 0.2191656 \cdot \ln T + 0.058441335 \cdot \ln {}^{\circ}Bx} \quad (13)$$

$$\ln \mu = -4.7989371 + \frac{463834.09}{T^2} + 0.00019800219 \cdot {}^{\circ}Bx^2 \cdot \ln {}^{\circ}Bx \quad (14)$$

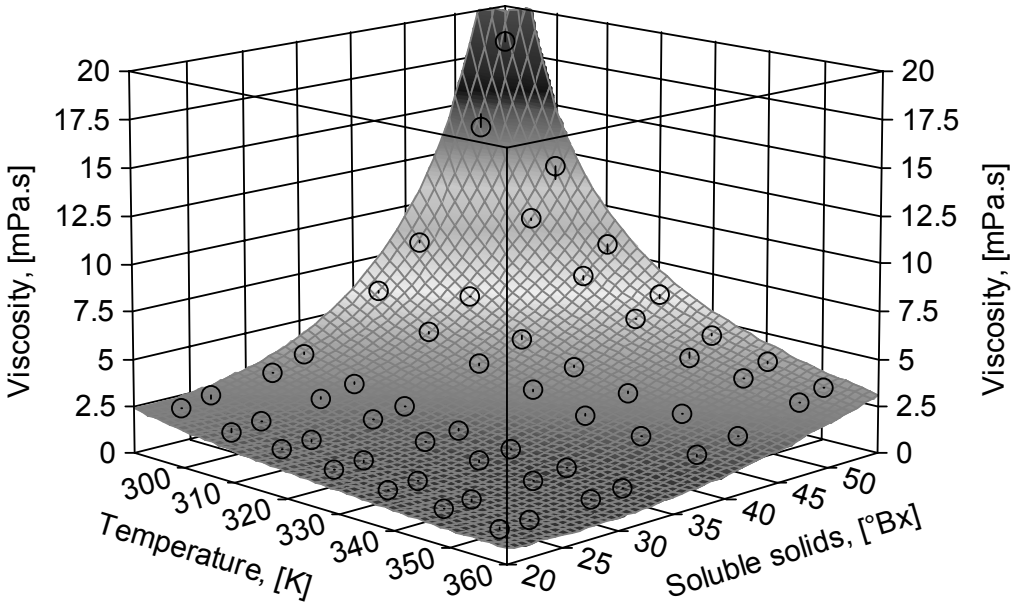
Equation (13), has a better precision (R^2 = 0.9971), meanwhile equation (14) is more simple, but has a lower precision (R^2 = 0.9919). Both equations have an $R^2 > 0.99$, thus being more accurate than equations (1-3), (4) or (8).

Using the models developed for the calculation of dynamic viscosity and the mathematical model (15) developed for calculating the density of the grapes juice [8-11], according to equation (16) can be calculated kinematic viscosity of grapes juice:

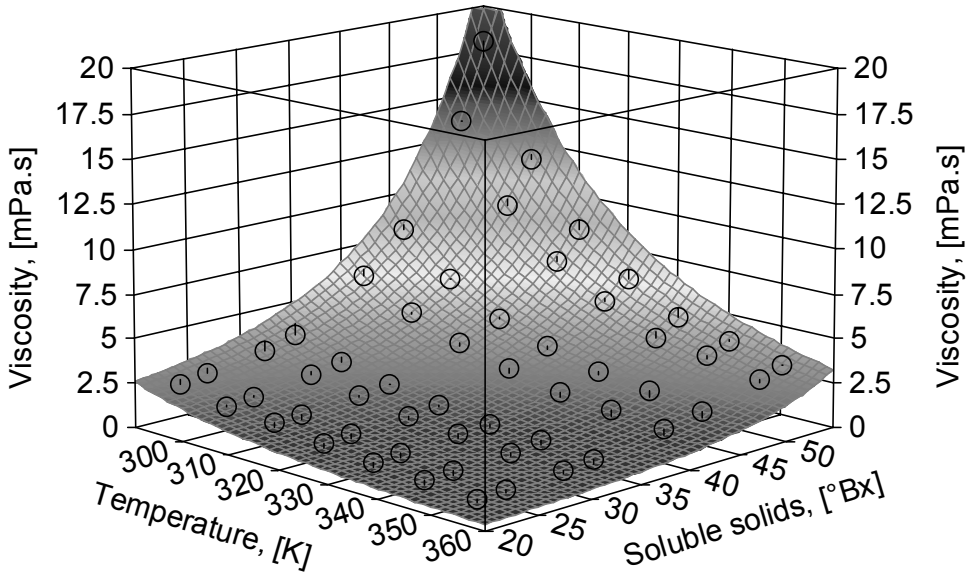
$$\rho = (1842.7 + 8.8595 \cdot X) + (-153.69 - 0.6631 \cdot X) \cdot \ln(T) \quad (15)$$

$$\nu = \frac{\mu}{\rho} \quad (16)$$

where v is the kinematic viscosity [m^2/s] and X the dry matter content [%, w/w].



a



b

Figure 4. *Grapes juice viscosity values plotted in TableCurve 3D and fitted equations with residuals: a) equation (13); b) equation (14)*

Heat capacity

Using CurveExpert® software, a "heat capacity" model correlation between temperature and heat capacity, at constant dry matter content has been established:

$$C_p = M + N \cdot T + \frac{S}{T^2} \tag{17}$$

The *M*, *N* and *S* values are presented in Table 4. The regression coefficients are equal to 0.999, thus indicating a good correlation of variables.

Table 4. *Coefficients for equation (17)*

Soluble solids, *X* [%]	***M***	***N***	***S***	**R^2**
20	15784.47	-24.2248	-443308	0.9990
30	11993.47	-16.6931	-323151	0.9990
40	13713.08	-20.8092	-381959	0.9990
50	14744.78	-23.2384	-429346	0.9990
60	15269.68	-24.3640	-456607	0.9990

In order to correlate *M*, *N* and *S* coefficients with soluble solids content, more models were used in CurveExpert® software. The best mathematical model is represented by the linear equation (Table 5):

$$Coefficient = m \cdot X + n \tag{18}$$

Table 5. *Coefficients for equation (18)*

Coefficient	***m***	***n***	**R^2**
M	108.60339	9043.1067	0.9427
N	-0.254421	-9.827303	0.9355
S	-4477562.5	-196276020	0.9755

Combining equations (17) and (18) and replacing the coefficients with numeric values in order to correlate the heat capacity of grapes juice with temperature and soluble solids, the final form of the proposed model is:

$$C_p = 9043.1067 + 108.60339 \cdot X - (0.254421 \cdot X + 9.827303) \cdot T - \\ - \frac{(4477562.5 \cdot X + 196276020)}{T^2} \tag{19}$$

To quantify the deviation from experimental data, between measured and calculated heat capacity, the relative error (20a) and average relative error (20b) were used:

$$\varepsilon = \left| \frac{C_{p\,measured} - C_{p\,calculated}}{C_{p\,measured}} \right| \cdot 100 \quad [\%] \tag{20a}$$

$$\tilde{\varepsilon} = \frac{1}{n} \sum_{i=1}^{n} \varepsilon_i \tag{20b}$$

Table 6 presents the comparison between measured values (digital recovered from the graph depicted in Figure 1) and the values calculated with the literature equation (5) [2] and the proposed model, equation (19). By analyzing the obtained values it can be observed that the induced relative error for the proposed equation model is only 0.19% compared with those obtained using equation (5), which gives an average error of 0.9%.

Table 6. *Comparison between digital recovered values from graphic and calculated values with equation (5) and proposed model for heat capacity of grapes juice*

Temperature, [K]	Dry matter, [%]	Heat capacity, C_p [J/(kg·K)]				
		Digital recovered values from graphic (Figure 1)	calculated with equation:			
			(5)		(19)	
			C_p	ε [%]	C_p	ε [%]
293	20	3528	3700	4.6486	3515.44	0.3570
	30	3342	3484	4.0757	3334.46	0.2259
	40	3157	3268	3.3965	3153.48	0.1114
	50	2921	3052	4.2922	2972.50	1.7326
	60	2810	2836	0.9167	2791.52	0.6620
303	20	3610	3720	2.9569	3582.42	0.7696
	30	3410	3504	2.6826	3409.86	0.0040
	40	3260	3288	0.8515	3237.29	0.7012
	50	3040	3072	1.0416	3064.73	0.8069
	60	2910	2856	1.8907	2892.16	0.6166
313	20	3670	3740	1.8716	3629.02	1.1290
	30	3470	3524	1.5323	3461.68	0.2402
	40	3310	3308	0.0604	3294.34	0.4753
	50	3111	3092	0.6144	3126.99	0.5116
	60	2999	2876	4.2767	2959.65	1.3293
323	20	3715	3760	1.1968	3657.72	1.5659
	30	3500	3544	1.2415	3492.80	0.2061
	40	3325	3328	0.0901	3327.87	0.0864
	50	3116	3112	0.1285	3162.95	1.4845
	60	3020	2896	4.2817	2998.03	0.7327
333	20	3725	3780	1.4550	3670.64	1.4809
	30	3530	3564	0.9539	3505.66	0.6941
	40	3330	3348	0.5376	3340.68	0.3199
	50	3117	3132	0.4789	3175.71	1.8488
	60	3030	2916	3.9094	3010.73	0.6398
343	20	3710	3800	2.3684	3669.58	1.1012
	30	3520	3584	1.7857	3502.37	0.5032
	40	3324	3368	1.3064	3335.15	0.3345
	50	3116	3152	1.1421	3167.93	1.6395
	60	3030	2936	3.2016	3000.72	0.9756
353	20	3671	3820	3.9005	3656.13	0.4066
	30	3505	3604	2.7469	3484.72	0.5816
	40	3310	3388	2.3022	3313.32	0.1004
	50	3110	3172	1.9546	3141.92	1.0161
	60	3010	2956	1.8267	2970.52	1.3288

By analyzing the obtained values it can be observed that the induced relative error for the proposed equation model is only 0.7634% and for equation (5) is 2.0548%.

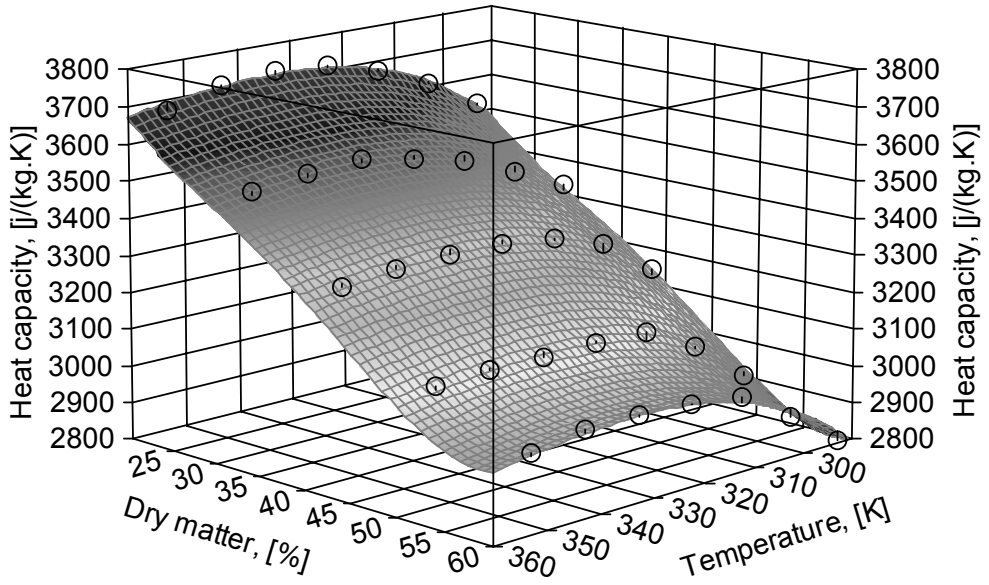


Figure 5. *Grapes juice heat capacity values plotted in TableCurve 3D and fitted linear equation (21) with residuals*

Plotting digital recovered data for grapes juice heat capacity in TableCurve 3D® v.4 software (Figure 5) a polynomial equation for the response function was generated, chosen due to the relative simplicity of formulation and high accuracy ($R^2 = 0.9981$).

$$Cp = -102924.62 + 1144.7527 \cdot T - 4.5367833 \cdot T^2 + 0.0079959596 \cdot T^3 - \\ - 5.3030303E - 06 \cdot T^4 - 161.41429 \cdot X + 6.2425 \cdot X^2 - 0.116 \cdot X^3 + \\ + 0.000775 \cdot X^4 \qquad (21)$$

CONCLUSIONS

The newly proposed equations can be used in the software easier than using existing experimental data in tabular form (taking into consideration that for those values not found tables an interpolation calculus is necessary) or graphic form (taking into account that the digital extraction of those values is compulsory).

The elaborated models have a high degree of accuracy, the heat capacity model is approximate four times more precise than a previous model existing in literature, more simple in formulation and for a wide range of parameters; for viscosity were offered four models based on a simple logarithmic equation as a function of soluble solids (°Bx) or dry matter (X) and temperatures measured in Kelvins or Celsius degrees.

The dedicated software TableCurve 3D® for fitting equations for plotted surfaces, generated equations with coefficients of determination over 0.99 for viscosity and heat capacity of grapes juice.

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