MATHEMATICAL MODELLING OF TEN VEGETABLE OILS THERMOPHYSICAL PROPERTIES. STUDY OF DENSITY AND VISCOSITY

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Abstract
Scattered data and the behaviour determined by state parameters of vegetable oil thermo-physical properties imposed the need to develop mathematical models that could cover with precision the values from an extended range of these properties. As consequence, this work was focused on finding a possibility to correlate density and viscosity of 10 vegetable oils (olive, peanuts, sesame, almond, cotton, corn germ, sunflower, soybean, grape seed and flaxseed) with temperature and oil tabular hierarchy number.
The available data were analysed and used as inputs in various software which helped to generate several mathematical models. In the case of vegetable oil density four polynomial equations were developed in order to fit the existing literature data value, while for dynamic viscosity variation a polynomial and the Arrhenius equation or "vapour pressure" model gave the best results.
The adequacy of the new mathematical models was verified by using correlation coefficient, relative error and ANOVA test and their evaluation revealed a very good fit with the experimental data.
The proposed mathematical models can be loaded in the widespread PC software for storing, organizing and manipulating data available both for industrial and academic users and they could be valuable for designing or evaluating handling and processing systems and equipment that are involved in the storage, handling and utilization of vegetable oils.

Keywords: oil, density, dynamic viscosity, kinematic viscosity, mathematical modelling

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1. INTRODUCTION

Public interest in foods, cosmetics and pharmaceuticals quality and methods of production has increased significantly in the recent decades, due in part to changes in eating habits, consumer behaviour, and to the increased industrialization and globalization of products supply chains. Demand for high levels of production quality and safety requires high standards compliance and a severe process control (Luna *et al.*, 2013).

Vegetable oils consist of triglycerides, which are glycerol molecules with three long chain fatty acids attached at the hydroxyl groups via ester linkages (Shashidhara and Jayaram, 2010). They can be involved directly or indirectly in different technological aspects (raw and secondary materials, by-product, finished product etc.) in various processes (Rodenbush *et al.*, 1999). The importance of vegetable oils in industries such as foods, soaps, cosmetics, and pharmaceuticals has been well documented (Bockisch, 1998). Some vegetable oils such as olive one are accepted as possessing health beneficial properties (Park *et al.*, 2010; Gouveia de Souza *et al.*, 2004). Numerous evidences are supporting the relationship between olive oil and cancer, with most of the attention being directed toward its fat and phenolic content. One of these studies (Ismail *et al.*, 2013) sustains that olive oil potentiates the effects of aromatase inhibitors via glutathione depletion in estrogen receptor-positive human breast cancer (MCF-7) cells. Other findings support the concept that peanuts oil may possess an unexpectedly atherogenic effect especially due to its content of arachidic and behenic acids (Kritchevsky and Tepper, 1971). Peanut oil may be used also as a natural, non-toxic, cost-effective and biodegradable extractant for decontamination of polycyclic aromatic hydrocarbon contaminated soil. Extraction efficiency was established at more than 90% when peanut oil at concentrations of 2.5–20% was used to remove anthracene from garden soil (Pannu *et al.,* 2004). Sesame oil is receiving considerable attention across the world for its potential health benefits in relation to neurological disorders (Ahmad *et al.,* 2006). It is effective against various diseases including atherosclerosis, hypertension and anti-aging effects. It contains a good amount of phenol, sesaminol, sesamin, sesamol and sesamolin and relatively small amounts of tocopherol which contributes to its superior oxidative stability. Sesame oil, in comparison to other dietary oils such as ground nut and sunflower, offers better protection against increased blood pressure, hyperlipidemia and lipid peroxidation by increasing enzymatic and non-enzymatic antioxidants. Studies have demonstrated sesame oil and its active ingredient sesamol to be a strong antitumor promoting agent when compared with resveratrol and sunflower oil (Ahmad *et al.,* 2006). It is used as a cooking oil, in shortening and margarine, as a soap fat, in pharmaceuticals and as a synergist for insecticides (Doker *et al.,* 2010). Almond oil presents anti-inflammatory, immunity-boosting and anti-hepatotoxicity effects. Associations have also been made with reductions in the incidence of colonic cancer. Almond oil is successfully used in the cosmetic industry for its penetrating, moisturizing and restructuring properties (Ahmad, 2010). Thermally oxidized sunflower oil was found to improve lipid metabolism by lowering the level of postprandial lipemia in healthy subjects and especially in younger individuals (Manning *et al.,* 2013). Sunflower oil modified hyperbranched polyurethane and linear polyurethane were synthesized and can be used as a suitable material for thin film applications which has the potential to replace or minimize the use of non-biodegradable and petroleum based material (Das *et al.,* 2013). Soybean oil is a surface-active agent which may accelerate the oxygen transfer in the production of tetracycline (Jia *et al.,* 1999). Grape seed oil is indicated for human consumption and in particular for infants and elderly people: its pharmaceutical activities concern the ability to contrast free-radicals, cardiovascular diseases, cholesterol (Fiori, 2007). Flax oil is considered a bioactive lipid, as it is rich in ω-3 fatty acids, specifically α-linolenic acid, which has been associated with various health benefits (Comin, *et al.,* 2012). It is often encapsulated or protected in food polymers (Comin *et al.,* 2012) and other materials such as zein (Quispe-Condori *et al.,* 2011) to ensure its delivery in good quality and prevent degradation, to preserve their beneficial properties. Combined with antioxidants flax oil conducted to significant improvements on blood fatty acids composition and behavior in children with Attention Deficiency Hyperactivity Disorder (Joshi *et al.,* 2006). The flax oil is also added into infant formula and various food products and available as nutraceutical supplements in many countries (Bozan and Temelli, 2008). Vegetable oils are known also as sustainable sources for energy production. There are many applications where these biofuels are used directly in engines. Vegetable oils are important alternatives as fossil fuels replacement. Several authors explain vegetable oils potential and characteristics and their use in internal combustion engines (Toscano *et al.,* 2012; Aworanti *et al.,* 2012) a wide variety of studies being focused on biodiesel production Russo *et al.,* 2012; Jimenez-Lopez *et al.,* 2011; Kocar and Civas, 2013).

The appropriate use of these oils is related to their thermodynamic properties which influence equally the quality of products in which they are incorporated and the adequate exploitation of process equipment such as heat exchangers, reactors, process piping and storage tanks (Fasina and Colley, 2008). Two of the more important thermophysical properties of vegetable oils are represented by density and viscosity. Even though several studies have reported different expressions for vegetable oils thermophysical properties (Rodenbush *et al.,* 1999; Toscano *et al.,* 2012; Aworanti *et al.,* 2012; Halvorsen *et al.,* 1993; Neelamegam and Krishnaraj, 2011; Esteban *et* *al.*, 2013; Noureddini *et al.*, 1992;) there is still a necessity of expanding the databank gathered from literature. In most cases, previous papers present polynomial correlations or specific equations (like Rackett equation for density and Vogel equation for viscosity) (Sales-Cruz *et al.*, 2000) generated by adjusting each set of experimental data to a specific compound. In contrast to traditional methods based on temperature-dependence correlations, in this work, is presented a new generalized method useful to find mathematical correlations between temperature and oil type and the two aforementioned thermodynamic properties for ten vegetable oils namely olive, peanuts, sesame, almond, cotton, corn germ, sunflower, soybean, grape seed and flax. The use of correlation coefficient, relative error and ANOVA test has successfully showed that the new mathematical models can describe with high accuracy the data existing in graphical and tabular form which are rather difficult to apply in practice.

2. MATERIAL AND METHODS

Experimental data provided by the scientific publications (Tables 1 and 2) concerning the density and dynamic viscosities variations of different types of oil with temperature were used as primary data for the regression analysis.
In order to develop mathematical models that could predict the thermophysical property tendency, for a certain level of complexity in generating equation models after data integration and plotting, Microsoft Excel™ 2010 software was employed. ANOVA analysis tool integrated also in this software was able to compare the experimental with the calculated data generated by the proposed mathematical models. More complex models generated in 2D (“vapour pressure” model, “heat capacity” model etc.) were performed with CurveExpert® software and 3D representations as a surface response were fitted and analysed in TableCurve 3D® v.4 software.
Because a second parameter that could influence the thermo-physical properties evolution except the temperature (concentration of certain fat acids, saponification index, melting point etc.) was difficult to be integrated in the mathematical model a simpler methodology was developed. For each type of oil (the present mathematical model being applied and verified on only 10 types) a number from 1 to 10 was attributed.
Thermo-physical property vs. Temperature, *Thermo-physical property vs. oil number* were plotted and different types of regression techniques, involving the method of least squares. Relative error ε (Equation 1) and ANOVA were used also to reveal the best-fit equation.

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

3. RESULTS AND DISCUSSIONS

Density

Using Microsoft Excel™ 2010 and CurveExpert® softwares, after a preliminary hierarchy in the sense of the increasing average value of density for each studied oil, 10 quadratic correlations (considering the best regression coefficient and the equation complexity) between temperatures *T*, [K] or *t*, [°C] and density *ρ* [kg/m^3], have been established and are expressed by the following equation:

$$\rho = a_1 + a_2 T + a_3 T^2 \qquad (2)$$

The a_1, a_2 and a_3 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 the hierarchy number *No. oil*, several models were generated in available softwares (1st, 2nd and 3rd degree polynomial equations, “vapour pressure” model, “heat capacity” model etc.). The best fit model is the logarithmic equation (Table 4).

$$Coefficient = b_1 + b_2 \ln(No.oil) \qquad (3)$$

Combining the equations 2 and 3 (when the temperature is measured both in Kelvin, T and Celsius degrees, t) the final form of proposed equation model (Equation 4) is:

$$\rho = (b_{1a1} + b_{2a1} \ln(No.oil)) + (b_{1a2} + b_{2a2} \ln(No.oil)) \cdot T + (b_{1a3} + b_{2a3} \ln(No.oil)) \cdot T^2 \quad (4)$$

Table 1. Density variation for different types of oil with temperature (Macovei, 2000)

Temperature, T [K]	Flax seed	Soybean	Corn germ	Sunflower seed	Sesame seed	Grape seed	Cotton seed	Peanut	Olive
	Density, ρ [kg/m^3]								
253.15	958.0	948.0	947.0	946.1	946.0	946.0	944.6	941.8	940.0
263.15	951.1	941.1	940.0	939.5	938.3	940.0	938.0	934.4	933.1
273.15	944.2	934.0	933.1	932.8	931.2	933.1	930.7	927.5	926.0
283.15	937.5	927.0	927.0	926.0	924.3	926.0	924.2	920.5	918.9
288.15	934.8	923.1	923.2	922.4	921.0	923.0	921.2	917.2	915.3
293.15	931.3	919.4	920.0	918.9	917.5	919.1	917.7	913.7	911.8
298.15	927.8	916.1	916.5	915.4	914.0	916.2	914.2	910.2	908.3
303.15	924.3	912.6	913.0	911.7	910.5	912.8	910.7	906.7	904.8
308.15	920.8	909.1	909.1	908.9	907.0	909.1	907.2	903.2	901.3
313.15	917.3	905.6	906.0	904.9	903.5	906.0	903.7	899.7	897.8
318.15	913.8	902.1	900.1	901.4	900.0	900.0	900.2	896.2	894.3
323.15	910.3	898.6	898.6	897.9	896.5	898.0	896.7	892.7	890.8
333.15	903.3	891.6	893.1	890.9	889.5	892.0	889.7	885.7	883.8
343.15	896.3	884.6	885.7	883.9	882.7	885.2	882.7	878.7	876.8
353.15	889.3	877.6	879.0	876.9	875.5	878.1	875.7	871.7	869.8
363.15	882.3	870.6	872.0	869.9	868.5	872.0	868.7	864.7	862.8
373.15	875.3	863.6	865.1	862.9	861.5	865.0	861.7	857.7	855.8
393.15	861.6	849.2	852.0	848.5	847.2	851.1	847.4	843.6	841.4
413.15	847.5	835.5	838.1	835.0	833.0	838.0	833.2	830.0	827.5
423.15	840.3	828.6	831.0	827.9	826.5	831.0	826.7	822.7	820.8
473.15	805.3	793.6	795.0	792.9	791.5	795.0	791.7	787.7	785.8
523.15	770.3	758.6	760.0	797.7	756.5	760.0	756.7	752.7	750.8

Table 2. Dynamic viscosity variation for different types of oil with temperature (Macovei, 2000)

Temperature, T [K]	Olive	Peanut	Sesame seed	Almond	Cotton seed	Corn Germ	Sunflower seed	Soybean	Grape seed	Flax seed
	Dynamic viscosity, μ · 10^3 [Pa·s]									
288.15	99.70	96.70	90.20	90.10	85.30	85.10	79.80	72.90	67.20	65.80
293.15	78.10	75.90	71.50	71.20	67.20	66.40	63.30	57.80	53.10	52.70
298.15	62.50	60.50	57.00	56.90	54.60	51.70	51.10	47.10	42.40	43.70
303.15	50.80	49.40	46.60	46.50	44.30	41.10	42.20	38.90	34.10	36.20
308.15	41.50	40.60	38.60	38.60	36.60	33.00	34.90	32.50	27.70	30.40
313.15	31.30	33.90	31.80	31.90	30.70	27.60	29.30	27.70	22.60	25.70
318.15	28.90	28.50	27.00	27.00	25.80	22.40	24.90	23.30	18.00	22.00
323.15	24.50	24.20	23.20	23.10	22.10	18.60	21.30	20.20	14.80	19.00
333.15	18.10	18.00	17.10	17.20	16.50	13.00	16.00	15.10	10.00	14.50
343.15	13.80	13.80	13.20	13.20	12.70	9.11	12.50	11.80	6.97	11.30
353.15	10.80	10.90	10.40	10.40	10.10	6.07	10.00	9.40	4.74	9.10
363.15	8.70	8.70	8.40	8.40	8.20	4.70	8.10	7.70	3.22	7.50
373.15	7.10	7.20	6.90	6.90	6.70	4.07	6.70	6.40	2.60	6.20

Table 3. Coefficients for equation no. 2

Type of oil	Number of hierarchy, *No. oil*	Equation 2 coefficients			
		a_1	a_2	a_3	R^2
Flaxseed	1	944.5484	-0.67895	-0.0001101525	0.9999
Soybean	2	933.8699	-0.70774	0.0000374850	0.9999
Corn germ	3	933.282	-0.68605	0.0000433166	0.9996
Sunflower	4	932.6692	-0.69172	-0.0000530301	0.9999
Sesame seed	5	931.5105	-0.69992	-0.0000109768	0.9999
Grape seed	6	932.8557	-0.68692	-0.0001071007	0.9996
Cotton seed	7	931.1394	-0.68307	-0.0001071007	0.9999
Peanut	8	927.6195	-0.69877	-0.0000007746	0.9999
Olive	9	925.9443	-0.70367	0.0000106771	0.9999

Table 4. Coefficients for equation no. 3

Equation 2 coefficients	Equation 3 coefficients	
	b_1	b_2
Kelvin measure unit for temperature		
a_1	1128.5228457641	-5.0447177362
a_2	0.675549072	-0.0088159248
a_3	0.0000257669	0.0000107955
Celsius degree measure unit for temperature		
a_1	942.04	-6.632
a_2	- 0.687	-0.004
a_3	- 4E-05	7E-06

Table 5. Correction coefficients for equation no. 4

Type of oil	Flax seed	Soybean	Corn germ	Sunflower seed	Sesame seed	Grape seed	Cotton seed	Peanut	Olive
Temperature measured in:	Correction coefficients								
Kelvins	0.287	-0.480	-0.091	0.186	-0.027	0.398	0.237	-0.096	-0.220
Celsius degrees	2.581	-4.446	-1.276	-0.828	-0.783	2.755	1.400	-1.561	-2.684

Using the relative error equation and considering its negative and positive values, the calculated data by the mathematical model and the existing experimental data were compared (Table 9) obtaining a final average of 0.021% (absolute value of relative error of 0.236).

The regression coefficient R^2 was determined for the proposed model of 0.9943 using Kelvin as measure units for temperature.

For equation formed with generated coefficients for the Celsius degrees version were determined a relative error and a regression coefficient of -0.057% (absolute value of relative error 0.266) and respectively 0.9938.

In order to increase the accuracy of the proposed mathematical model, correction coefficients for each type of oil were imposed (Table 5) and by summing them with the equation 4 determined an increases of the regression coefficient at 0.9948 (T, Kelvin) respectively at 0.9964 (t, Celsius degrees).

The ANOVA analysis was used to compare the values of experimental and calculated density data at 9 different types of oils in 22 temperatures variation.

The results presented in Table 6 showed that the sample *P*-value is 0.974192 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 studied types of oils in TableCurve 3D® v.4 software (Figure 1) an equation for the response function was generated, chosen due to the accuracy and simplicity.

The Equation 5 is a polynomial equation, Rank 32, Eqn. 70 in TableCurve 3D® v.4 library with a precision of R^2 = 0.9960, FitSdErr = 3.0075, Fstat. = 4768.2588. The coefficients values are presented in Table 7.

Table 6. The ANOVA test summary (Kelvin for temperature unit measure)

Summary	Density, ρ [kg/m^3]									Total
	Flax seed	Soybean	Corn germ	Sunflower seed	Sesame seed	Grape seed	Grape	Peanut	Olive	
Experimental data										
Count	22	22	22	22	22	22	22	22	22	198
Sum	19742.7	19490.2	19504.6	19512.4	19442.2	19496.7	19443.3	19359	19317.7	175308.8
Average	897.3955	885.9182	886.5727	886.9273	883.7364	886.2136	883.7864	879.9545	878.0773	885.398
Variance	2305.52	2338.891	2256.39	1913.791	2332.503	2244.856	2316.254	2325.565	2336.178	2197.902
Calculated data										
Count	22	22	22	22	22	22	22	22	22	198
Sum	19692.37	19572.56	19520.92	19484.38	19446.45	19428.8	19402.41	19375.13	19355	175278
Average	895.1077	889.6618	887.3145	885.6536	883.9295	883.1273	881.9277	880.6877	879.7727	885.2425
Variance	2296.228	2299.22	2301.11	2302.247	2303.22	2303.978	2304.682	2305.365	2305.915	2230.066

ANOVA						
Source of Variation	*SS**	*df**	*MS**	*F*	*P-value**	*F crit*
Sample	2.392445	1	2.392445	0.001048	0.974192	3.866177
Columns	8965.553	8	1120.694	0.490912	0.862678	1.962913
Interaction	413.9638	8	51.74547	0.022667	0.999997	1.962913
Within	862930.2	378	2282.884			

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

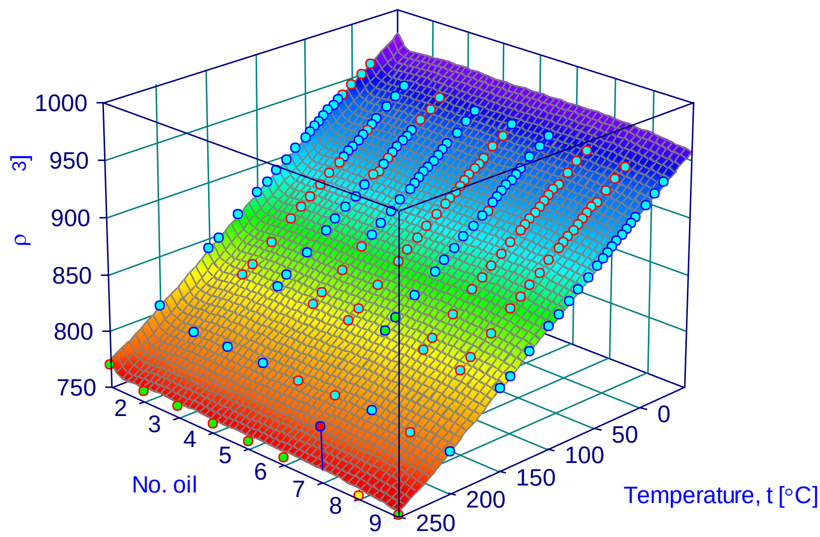

Figure 1. Oils density values plotted in TableCurve 3D and fitted with polynomial equation type (Equation 5 with temperature measured in Kelvin) with residuals

$$\ln \rho = a_1 + a_2 \cdot No.oil + a_3 \cdot No.oil^2 + a_4 \cdot No.oil^3 + a_5 \cdot No.oil^4 + a_6 \cdot No.oil^5 + a_7 \cdot \ln(T) + a_8 \cdot \ln(T)^2 + a_9 \cdot \ln(T)^3 + a_{10} \cdot \ln(T)^4 + a_{11} \cdot \ln(T)^5 \quad (5)$$

Table 7. Coefficients for equation no. 5

Coefficient	*Value*	Coefficient	*Value*
Kelvin measure unit for temperature			
a_1	603.9115819	a_7	-86.3707142
a_2	6.939732598	a_8	208.3981931
a_3	-0.04348015	a_9	-216.069629
a_4	0.000122469	a_{10}	100.5905446
a_5	-1.7056e-07	a_{11}	-17.3573249
a_6	9.39257e-11	R^2	0.9960
Celsius degree measure unit for temperature			
a_1	944.6784216	a_7	-86.3707142
a_2	-0.69046898	a_8	208.3981931
a_3	-0.00033394	a_9	-216.069629
a_4	6.19598e-06	a_{10}	100.5905446
a_5	-4.2279e-08	a_{11}	-17.3573249
a_6	9.39257e-11	R^2	0.9960

Dynamic viscosity

In order to obtain a mathematical model that describes with accuracy the variation of dynamic viscosity of 10 types of oil (with little in common regarding the variation ratio of thermo-dynamical properties with temperature) the equation of Arrhenius was employed because it creates a good correlation between experimental data 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 [J·mol^{-1}], *R* – universal gas constant [8.3144621 J·mol^{-1} ·K^{-1}], *T*–absolute temperature [K].

Table 8. Calculated values for activation energy, Ea

Olive	Peanut	Sesame seed	Almond	Cotton seed	Corn Germ	Sunflower seed	Soybean	Grape seed	Flax seed
1	2	3	4	5	6	7	8	9	10
Ea – activation energy [J·mol^{-1}]									
31875.90	31802.69	31635. 95	31633.29	31502. 11	31496. 48	31342. 40	31125. 69	30930.60	30880.15
32531.40	32461.74	32316.16	32305.91	32164.96	32135.76	32019.21	31797.62	31590.86	31572.43
33201.96	33121.32	32973.57	32969.22	32866.92	32731.60	32702.66	32500.56	32239.91	32314.79
33881.11	33810.66	33663.56	33658.15	33535.96	33346.95	33413.53	33208.26	32876.25	33026.91
34547.55	34491.37	34361.92	34361.92	34225.58	33960.25	34103.70	33921.13	33511.61	33749.96
35192.84	35117.13	34950.60	34958.78	34858.93	34581.72	34737.38	34591.14	34061.24	34395.98
35855.47	35818.60	35675.56	35675.56	35555.28	35181.40	35461.34	35285.62	34602.80	35133.73
36486.50	36453.40	36340.00	36328.39	36209.46	35746.13	36110.38	35967.89	35131.98	35803.31
37795.12	37779.77	37637.67	37653.83	37538.72	36878.21	37453.46	37293.07	36151.33	37180.74
39089.37	39089.37	38962.53	38962.53	38852.33	37904.28	38807.04	38642.59	37140.19	38519.03
40397.34	40424.40	40286.51	40286.51	40200.55	38705.20	40171.32	39989.61	37978.87	39894.35
41717.07	41717.07	41611.10	41611.10	41538.33	39857.51	41501.27	41348.33	38715.42	41268.85
42998.06	43041.46	42909.40	42909.40	42818.12	41271.34	42818.12	42675.97	39880.74	42577.45

Table 9. Oil density absolute value of relative errors for calculated data versus tabular data (Equation 4 with temperature measured in Kelvin)

Temp., T [K]	Density, ρ [kg/m³] Oil type																	
	Flaxseed		Soya bean		Corn germ		Sunflower seed		Sesame seed		Grape seed		Cotton seed		Peanut		Olive	
	CD*	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %
253.15	955.9	0.22	951.3	0.35	948.6	0.17	946.7	0.07	945.3	0.08	944.1	0.21	943.0	0.17	942.2	0.04	941.4	0.15
263.15	949.0	0.22	944.4	0.35	941.7	0.18	939.8	0.03	938.3	0.00	937.1	0.31	936.1	0.20	935.2	0.09	934.4	0.14
273.15	942.1	0.23	937.5	0.37	934.8	0.18	932.9	0.01	931.4	0.02	930.2	0.32	929.1	0.17	928.3	0.08	927.5	0.16
283.15	935.2	0.25	930.6	0.38	927.8	0.09	925.9	0.01	924.4	0.01	923.2	0.30	922.2	0.22	921.3	0.09	920.5	0.18
288.15	931.7	0.33	927.1	0.43	924.4	0.13	922.5	0.01	921.0	0.00	919.7	0.35	918.7	0.27	917.8	0.07	917.0	0.19
293.15	928.3	0.33	923.6	0.46	920.9	0.10	919.0	0.01	917.5	0.00	916.3	0.31	915.2	0.27	914.3	0.07	913.6	0.19
298.15	924.8	0.32	920.2	0.44	917.4	0.10	915.5	0.01	914.0	0.00	912.8	0.37	911.8	0.27	910.9	0.07	910.1	0.19
303.15	921.4	0.32	916.7	0.45	914.0	0.11	912.0	0.04	910.5	0.00	909.3	0.38	908.3	0.27	907.4	0.07	906.6	0.20
308.15	917.9	0.31	913.2	0.46	910.5	0.16	908.6	0.04	907.1	0.01	905.8	0.36	904.8	0.26	903.9	0.08	903.1	0.20
313.15	914.5	0.31	909.8	0.46	907.0	0.11	905.1	0.02	903.6	0.01	902.4	0.40	901.3	0.26	900.4	0.08	899.6	0.20
318.15	911.0	0.31	906.3	0.47	903.6	0.39	901.6	0.02	900.1	0.01	898.9	0.12	897.8	0.26	896.9	0.08	896.1	0.21
323.15	907.5	0.30	902.8	0.47	900.1	0.17	898.2	0.03	896.6	0.02	895.4	0.29	894.4	0.26	893.5	0.09	892.7	0.21
333.15	900.6	0.30	895.9	0.48	893.2	0.01	891.2	0.03	889.7	0.02	888.5	0.40	887.4	0.26	886.5	0.09	885.7	0.21
343.15	893.7	0.29	889.0	0.49	886.2	0.06	884.3	0.04	882.7	0.00	881.5	0.42	880.4	0.26	879.5	0.10	878.7	0.22
353.15	886.7	0.29	882.0	0.50	879.3	0.03	877.3	0.05	875.8	0.03	874.5	0.41	873.5	0.25	872.6	0.10	871.8	0.23
363.15	879.8	0.28	875.1	0.51	872.3	0.03	870.3	0.05	868.8	0.04	867.6	0.51	866.5	0.25	865.6	0.11	864.8	0.23
373.15	872.9	0.28	868.1	0.52	865.4	0.03	863.4	0.06	861.9	0.04	860.6	0.51	859.6	0.25	858.7	0.11	857.8	0.24
393.15	859.0	0.31	854.2	0.59	851.4	0.07	849.5	0.11	847.9	0.09	846.7	0.52	845.6	0.21	844.7	0.13	843.9	0.30
413.15	845.0	0.29	840.3	0.57	837.5	0.07	835.5	0.06	834.0	0.12	832.8	0.63	831.7	0.18	830.8	0.10	830.0	0.30
423.15	838.1	0.27	833.3	0.57	830.5	0.06	828.6	0.08	827.0	0.07	825.8	0.63	824.7	0.24	823.8	0.14	823.0	0.27
473.15	803.1	0.27	798.4	0.61	795.7	0.08	793.7	0.10	792.2	0.09	790.9	0.51	789.9	0.23	789.0	0.16	788.2	0.30
523.15	768.1	0.29	763.4	0.63	760.7	0.09	758.8	4.88	757.3	0.10	756.1	0.52	755.0	0.22	754.1	0.19	753.3	0.34

CD – calculated data, ε - absolute relative error

Table 10. Coefficients for equation no. 10 and 11

Types of oil	No. oil	Equation form	Equation 10 and 11 coefficients			
			a_1	a_2	a_3	R^2
Olive	1	Linear	-5769.75	130.7488	-	1.0000
		Quadratic	-8125.92	145.1321	-0.0218	0.9999
Peanut	2	Linear	-6272.97	132.1976	-	1.0000
		Quadratic	-8399.41	145.1786	-0.01968	0.9999
Sesame seed	3	Linear	-6541.90	132.5994	-	1.0000
		Quadratic	-9190.47	148.7677	-0.02809	0.9999
Almond	4	Linear	-6568.07	132.6778	-	1.0000
		Quadratic	-9603.38	151.207	-0.02809	0.9999
Cotton seed	5	Linear	-6865.68	133.2528	-	1.0000
		Quadratic	-9650.00	150.2499	-0.02577	0.9999
Corn Germ	6	Linear	-572.70613	111.9087	-	0.9977
		Quadratic	-14151.62	194.8021	-0.12566	0.9984
Sunflower seed	7	Linear	-7567.14	135.1274	-	1.0000
		Quadratic	-10558.09	153.3859	-0.02768	0.9999
Soybean	8	Linear	-7983.71	135.8686	-	0.9999
		Quadratic	-12735.06	164.8735	-0.04397	0.9999
Grape seed	9	Linear	1616.28	103.0172	-	0.9942
		Quadratic	-31775.79	306.8613	-0.30902	0.9990
Flax seed	10	Linear	-8712.73	137.6372	-	0.9999
		Quadratic	-15256.20	177.5822	-0.06055	0.9999

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:

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

or,

$$E_a = 2.303(\log \mu_0 - \log \mu) \cdot T \cdot R \qquad (9)$$

The "activation energy" values for the employed mathematical model calculated with equation 8 are presented in Table 8.

By plotting the results in Microsoft Excel™ 2010 vs. the hierarchy number presented in Table 10 linear and quadratic fits were generated aiming regression coefficients higher than 0.99.

$$Ea = a_1 + a_2 No.oil \qquad (10)$$

$$Ea = a_1 + a_2 No.oil + a_3 No.oil^2 \qquad (11)$$

Table 11. Coefficients for equation no. 12

Coefficients	**Equation 12 coefficients**		
	b_1	b_2	b_3
Equation 10 linear form			
a_1	- 5692.2127115371	201.8399767361	-9.9651083023
a_2	130.6751204116	0.4627603814	0.0231271749
Equation 11 quadratic form			
a_1	8474.0103824656	152.9493975868	-81.5090633257
a_2	147.6567872653	-1.7030746902	0.4598719541
a_3	0.0262570346	0.0031491884	-0.0006400863

Table 13. Coefficients for linear equation for Corn Germ and Grape seed oils

Coefficients	Equation coefficients	
	b_1	b_2
Equation 10 linear form		
a_1	-4950.6812066483	729.6625128295
a_2	129.6917928126	-2.963845767
Equation 11 quadratic form		
a_1	21096.7101893943	-5874.7221955377
a_2	-29.3162212206	37.3530550858
a_3	0.241047859	-0.0611183197

Table 14. Coefficients for equation no. 15

Type of oil	Number of hierarchy, *No. oil*	Equation 15 coefficients			
		a_1	a_2	a_3	R^2
Olive	1	-132.03834	9017.1334	17.38052	0.9999
Peanut	2	-133.79189	9055.1531	17.66107	0.9999
Sesame seed	3	-131.07408	8891.4359	17.26973	0.9999
Almond	4	-131.07408	8891.4359	17.26973	0.9999
Cotton seed	5	-130.3914	8822.401	17.18138	0.9999
Corn Germ	6	-72.451994	6633.858	8.292642	0.9998
Sunflower seed	7	-129.3447	8699.931	17.05969	0.9999
Soybean	8	-119.46252	8176.0589	15.61938	0.9999
Grape seed	9	26.465529	2074.489	-6.42073	0.9999
Flax seed	10	-110.42295	7658.5937	14.32277	0.9999

In order to correlate a_1, a_2 and a_3 coefficients with temperature, several equations were generated and by comparing the regression coefficient value and maintaining a level of simplicity in structure a quadratic model presented the best fit (Table 11).

Replacing the calculated activation energy value in equation 6 and using the water dynamic viscosity at the same temperature, two mathematical models were proposed with good accuracy regarding regression coefficient and relative error. For equation 13, R^2 registered a value of 0.9948 and ε% (absolute value of relative error) of 8.15% and R^2 of 0.9983 and ε% of 2.27% were determined for equation 14 (Table 12).

In order to create a better fit and to reduce de relative error value for Corn Germ and Grape seed oil, a linear equation in place of a quadratic fit could be employed for equation 12 coefficients (Table 13). For Corn Germ the average of ε% decreases from 24.26 % at 4.51 % and for Grape seed oil from 47.35% to 6.94 % in linear form but quadratic version the differences are unnoticeable.

Another approach in finding a mathematical model formed by simpler equations with high accuracy was by plotting the dynamic viscosity vs. temperature, T [K] in CurveExpert® software.

$$Coefficient = b_1 + b_2 T + b_3 T^2 \quad (12)$$

Combining the equations 10 or 11 with 12, the proposed mathematical forms for "activation energy" are:

$$Ea = (b_{1a1} + b_{2a1}T + b_{3a1}T^2) + (b_{1a2} + b_{2a2}T + b_{3a2}T^2) \cdot No.oil \quad (13)$$

$$Ea = (b_{1a1} + b_{2a1}T + b_{3a1}T^2) + (b_{1a2} + b_{2a2}T + b_{3a2}T^2) \cdot No.oil + (b_{1a3} + b_{2a3}T + b_{3a3}T^2) \cdot No.oil^2 \quad (14)$$

Table 12. Oil dynamic viscosity absolute value of relative errors for calculated data versus tabular data (Equation 14)

Temp., T[K]	Dynamic viscosity, $\mu \cdot 10^3$ [Pa·s]																			
	Oil type																			
	Olive		Peanut		Sesame seed		Almond		Cotton seed		Corn germ		Sunflower seed		Soya bean		Grape seed		Flaxseed	
	CD*	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %
288.15	97.19	2.52	93.89	2.91	90.54	0.38	87.15	3.27	83.74	1.82	86.91	2.12	76.91	3.62	73.50	0.83	68.84	2.44	66.78	1.49
293.15	76.40	2.18	73.91	2.62	71.40	0.14	68.88	3.26	66.36	1.26	66.94	0.82	61.32	3.12	58.83	1.78	53.84	1.39	53.91	2.30
298.15	60.95	2.48	59.04	2.41	57.14	0.24	55.23	2.93	53.34	2.31	52.26	1.07	49.58	2.96	47.73	1.35	42.49	0.22	44.10	0.91
303.15	49.20	3.16	47.72	3.40	46.26	0.73	44.81	3.64	43.37	2.10	41.22	0.29	40.54	3.93	39.15	0.65	33.75	1.01	36.44	0.66
308.15	40.10	3.37	38.95	4.06	37.82	2.03	36.70	4.92	35.60	2.73	32.79	0.62	33.45	4.15	32.40	0.30	26.94	2.74	30.36	0.12
313.15	33.91	2.83	32.98	2.71	32.07	0.86	31.18	2.26	30.31	1.28	27.04	2.05	28.62	2.34	27.80	0.35	22.20	1.77	26.21	2.00
318.15	28.04	2.99	27.30	4.20	26.59	1.52	25.89	4.09	25.22	2.26	21.76	2.84	23.92	3.94	23.29	0.03	17.80	1.10	22.09	0.43
323.15	24.00	2.04	23.40	3.30	22.82	1.62	22.26	3.62	21.72	1.71	18.12	2.56	20.69	2.87	20.20	0.02	14.72	0.55	19.26	1.36
333.15	17.74	1.99	17.34	3.66	16.96	0.82	16.59	3.52	16.24	1.55	12.64	2.80	15.59	2.57	15.28	1.21	10.02	0.23	14.71	1.43
343.15	13.58	1.62	13.30	3.59	13.05	1.17	12.80	3.03	12.57	1.04	9.09	0.23	12.14	2.89	11.94	1.20	6.96	0.14	11.58	2.47
353.15	10.60	1.88	10.41	4.50	10.23	1.61	10.07	3.22	9.91	1.90	6.64	9.47	9.62	3.77	9.49	0.99	4.86	2.56	9.26	1.76
363.15	8.47	2.66	8.34	4.16	8.22	2.20	8.10	3.57	7.99	2.54	4.96	5.48	7.80	3.74	7.71	0.13	3.43	6.60	7.55	0.73
373.15	6.93	2.33	6.84	4.95	6.76	2.07	6.68	3.24	6.60	1.48	3.78	7.14	6.46	3.52	6.40	0.05	2.45	5.61	6.29	1.51

CD – calculated data, ε - absolute relative error

The best fit to the graphical representations was established by the "vapour pressure" model (Equation 15).

$$\mu = \exp(a_1 + \frac{a_2}{T} + a_3 \ln(T)) \quad (15)$$

The a_1, a_2 and a_3 values are presented in Table 14 with regression coefficients R^2 greater than 0.99. Using the same methodology as in previous mathematical models, quadratic (Equation 15) connections were determined between a_1, a_2 and a_3 coefficients and the oil hierarchy number (Table 15).

The exceptions at this model are the coefficients of Corn Germ and Grape seed oil when a linear equation presented a better fit.

$$Coefficient = b_1 + b_2 No.oil + b_3 No.oil^2 \quad (16)$$

and for the coefficients for Corn Germ and Grape seed oils equation:

$$Coefficient = b_1 + b_2 No.oil \quad (17)$$

Combining the equations 15 with 16 and respectively 17, the final forms of proposed mathematical model (Equation 18 and 19) are:

$$\mu = \exp((b_{1a1} + b_{2a1} No.oil + b_{3a1} No.oil^2) + \frac{(b_{1a2} + b_{2a2} No.oil + b_{3a2} No.oil^2)}{T} + (b_{1a3} + b_{2a3} No.oil + b_{3a3} No.oil^2)\ln(T)) \quad (18)$$

and for Corn Germ and Grape seed oils:

$$\mu = \exp((b_{1a1} + b_{2a1} No.oil) + \frac{(b_{1a2} + b_{2a2} No.oil)}{T} + (b_{1a3} + b_{2a3} No.oil +)\ln(T)) \quad (19)$$

Table 15. Coefficients for equation no. 16

Coefficients	Equation 16 coefficients		
	b_1	b_2	b_3
a_1	- 129.6163324980	- 2.3601656199	0.4243930029
a_2	8909.0450533703	94.2012712067	21.7472700568
a_3	17.0228784540	0.3544234645	-0.0619242508
	Equation 16 coefficients		
a_1	-270.28704	32.9725076667	-
a_2	15752.5959	1519.7896666667	-
a_3	37.7193903	-4.9044581333	-

$$\ln \mu = a_1 + \frac{a_2}{T} + a_3 \cdot No.oil + \frac{a_4}{T^2} + a_5 \cdot No.oil^2 + a_6 \cdot \frac{No.oil}{T} \quad (20)$$

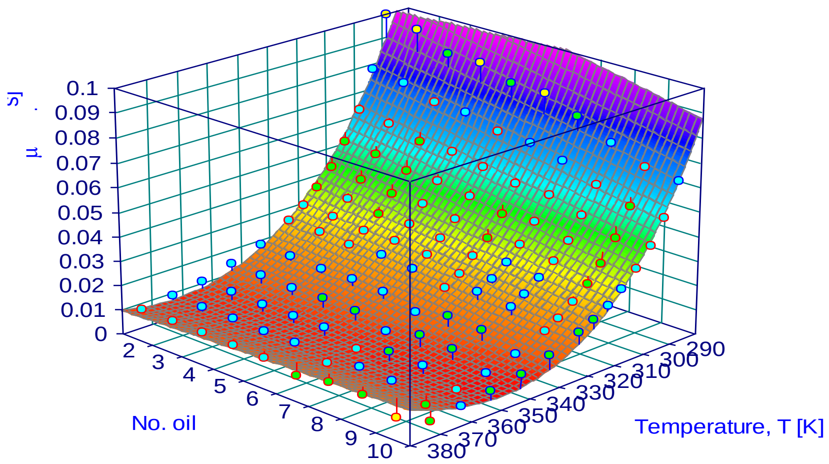

Figure 2. Oils dynamic viscosity values plotted in TableCurve 3D and fitted with polynomial equation type (Equation 20) with residuals

Table 16. Oil dynamic viscosity absolute value of relative errors for calculated data versus tabular data (Equations 18 and 19)

Temp., T[K]	Dynamic viscosity, $\mu \cdot 10^3$ [Pa·s]																			
	Oil type																			
	Olive		Peanut		Sesame seed		Almond		Cotton seed		Corn germ		Sunflower seed		Soya bean		Grape seed		Flaxseed	
	CD*	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %	CD	ε, %
288.15	98.50	1.20	95.50	1.24	92.20	2.22	88.70	1.55	85.10	0.23	85.00	0.12	77.40	3.01	73.40	0.69	67.30	0.15	65.40	0.61
293.15	78.00	0.13	75.70	0.26	73.20	2.38	70.60	0.84	67.90	1.04	66.20	0.30	62.10	1.90	59.10	2.25	53.30	0.38	53.10	0.76
298.15	62.50	0.00	60.80	0.50	58.90	3.33	56.90	0.00	54.80	0.37	52.10	0.77	50.50	1.17	48.20	2.34	42.50	0.24	43.70	0.00
303.15	50.70	0.20	49.40	0.00	47.90	2.79	46.40	0.22	44.80	1.13	41.50	0.97	41.50	1.66	39.70	2.06	34.00	0.29	36.30	0.28
308.15	41.70	0.48	40.60	0.00	39.40	2.07	38.20	1.04	37.00	1.09	33.30	0.91	34.40	1.43	33.10	1.85	27.40	1.08	30.40	0.00
313.15	34.60	0.86	33.70	0.59	32.80	3.14	31.90	0.00	30.90	0.65	27.00	2.17	28.90	1.37	27.90	0.72	22.20	1.77	25.80	0.39
318.15	29.00	0.35	28.30	0.70	27.60	2.22	26.80	0.74	26.10	1.16	22.10	1.34	24.50	1.61	23.70	1.72	18.10	0.56	22.00	0.00
323.15	24.50	0.00	24.00	0.83	23.40	0.86	22.80	1.30	22.20	0.45	18.20	2.15	20.90	1.88	20.30	0.50	14.80	0.00	19.00	0.00
333.15	18.00	0.55	17.70	1.67	17.30	1.17	16.90	1.74	16.50	0.00	12.60	3.08	15.70	1.88	15.30	1.32	10.00	0.00	14.40	0.69
343.15	13.70	0.72	13.50	2.17	13.20	0.00	13.00	1.52	12.70	0.00	9.00	1.21	12.10	3.20	11.80	0.00	6.90	1.00	11.30	0.00
353.15	10.80	0.00	10.60	2.75	10.40	0.00	10.20	1.92	10.10	0.00	6.60	8.73	9.70	3.00	9.50	1.06	4.80	1.27	9.00	1.10
363.15	8.70	0.00	8.50	2.30	8.40	0.00	8.30	1.19	8.20	0.00	5.00	6.38	7.90	2.47	7.70	0.00	3.40	5.59	7.40	1.33
373.15	7.10	0.00	7.10	1.39	7.00	1.45	6.90	0.00	6.80	1.49	3.80	6.63	6.60	1.49	6.50	1.56	2.50	3.85	6.20	0.00

CD – calculated data, ε - absolute relative error

Table 17. The ANOVA test summary for oils dynamic viscosity

Summary	Dynamic viscosity, μ [Pa·s]										Total
	Olive	Peanuts	Sesame	Almond	Cotton	Corn germ	Sunflower	Soya bean	Grape seed	Flax	
Experimental data											
Count	13	13	13	13	13	13	13	13	13	13	130
Sum	0.4794	0.4683	0.4419	0.4414	0.4208	0.38285	0.4001	0.3708	0.30743	0.3441	4.05708
Average	0.03687	0.03602	0.03399	0.03395	0.03236	0.02945	0.03077	0.02852	0.02364	0.02646	0.03120
Variance	0.00083	0.00077	0.00067	0.00067	0.0006	0.00064	0.00051	0.00042	0.00042	0.00034	0.00056
Calculated data											
Count	13	13	13	13	13	13	13	13	13	13	130
Sum	0.4778	0.4654	0.4517	0.4376	0.4231	0.3824	0.3922	0.3762	0.3072	0.344	4.0576
Average	0.03675	0.0358	0.03474	0.03366	0.03254	0.02941	0.03016	0.02893	0.02363	0.02646	0.03121
Variance	0.00081	0.00076	0.00071	0.00065	0.00060	0.00064	0.00049	0.00044	0.00042	0.00034	0.00056

ANOVA

Source of Variation	*SS**	*df**	*MS**	*F*	*P-value**	*F crit*
Sample	1.04E-09	1	1.04E-09	1.76E-06	0.998943	3.880497
Columns	0.004199	9	0.000467	0.788549	0.627188	1.919026
Interaction	8.41E-06	9	9.34E-07	0.001579	1	1.919026
Within	0.141982	240	0.000592			

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

Table 18. Coefficients for equation no. 20

Coefficient	***Value***	**Coefficient**	***Value***
a_1	1.226571356	a_4	162120.869
a_2	-889.342671	a_5	-1.729e-05
a_3	0.010635199	a_6	-3.79101472

Using both "vapour pressure" model equations, the relative error (Table 16), considering its negative and positive values, between calculated and experimental data reaches a value of 0.083% or 1.218 in its absolute value and a regression coefficient R^2 of 0.9994.

The ANOVA analysis was used to compare the values of experimental and calculated dynamic viscosity data at 10 different types of oils in 13 temperatures variation. The results presented in Table 17 showed that the sample *P*-value is 0.998943 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 experimental and calculated data.

By plotting tabular data of dynamic viscosity for studied types of oils in TableCurve 3D® v.4 software (Figure 2) an equation for the response function was generated, chosen due to the accuracy and simplicity. The Equation 20 is a polynomial equation, Rank 51, Eqn. 303 in TableCurve 3D® v.4 library with a precision of R^2 = 0.9960, FitSdErr = 0.00329, Fstat. = 1321.4281. The coefficients values are presented in Table 18.

Combining the models developed for the calculation of dynamic viscosity and density of the same type of oils, the kinematic viscosity (ν) can be calculated using equation 21:

$$\nu = \frac{\mu}{\rho}\ [\mathrm{m^2 \cdot s^{-1}}] \qquad (21)$$

4. CONCLUSIONS

It is well known that both vegetable oils density and even more their viscosity have a profound impact on their use in technological processes or in products such as foods, cosmetics, biofuels etc.

Dependences between two thermodynamic properties (density and viscosity) of 10 vegetable oils (olive, peanuts, sesame, almond, cotton, corn germ, sunflower, soybean, grape seed and flax) and temperature and tabular hierarchy number oil were described by means of specific mathematical equations.

Four polynomial models with high accuracy (R^2 over 0.99) were established for density taking in consideration the unit measure for temperature (Kelvin or Celsius degrees), correction coefficients, for each type of oil density unique behaviour or complexity of equation structure.

In the case of viscosity various direct and combination of linear and/or quadratic relations, Arrhenius equation form and "vapour pressure" model conducted to a good correlation between experimental and calculated data. Results of this research show that the analysed vegetable oils thermodynamic properties are very well described by the developed mathematical model since the correlation coefficient, relative error and ANOVA test gave all appropriate values.

The proposed mathematical models can be loaded in the widespread PC 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 equipment and processes. For targeted temperature and oil type, more precise values of the studied thermophysical properties can be found easier using these models than using the existing experimental data in tabular form or graphic form.

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