Arrhenius-Type Relationship of Viscosity as a Function of Temperature for Mustard and Cotton Seed Oils

Ike, E.

Department of Physics, Modibbo Adama University of Technology,
P. M. B. 2076, Yola, Adamawa State, Nigeria

Abstract

The knowledge and estimate of transport behaviours of fluids are very influential in heat and mass flow. In this study, an equation correlating the Arrhenius energy (Ea), the pre-exponential factor (A), the Arrhenius temperature (T) and the Arrhenius activation temperature (T*) is applied so as to buttress the depth of discussion. The results obtained from the viscosity experiments of Mustard and Cotton seed oils at different temperature ranges offers very good results statistically. The Activation energy E_a, Entropic (pre-exponential) factor A, Arrhenius temperature T_A and the Arrhenius activation temperature for the mustard oil were observed to be 374.37381 J/mole, 12.39260595 cP, -17.89797783 ℃, 45.051 ℃ respectively while Activation energy E_a, Entropic (pre-exponential) factor A, Arrhenius temperature T_A and the Arrhenius activation temperature for the cotton seed oil are respectively 451.90611 J/mole, 8.210386507 cP, -25.8292961 ℃, 54.381 ℃. The coefficients of regressions (R^2) for the graph of the natural log of viscosity versus reciprocal of temperature (Figures 2 and 4) for the mustard oil and cotton seed oil are 0.9996 and 0.9996 respectively. Since the correlation coefficient is the measure of how well a collection of data points can be modeled by a line, we can hence conclude that the natural log of the viscosity of both seed oil samples versus the inverse of their respective temperatures have a very good fit.

Keywords: Viscosity, Arrhenius Parameters, model, Correlation, Statistics, temperature, Vegetable oil.

Introduction

The term oil refers to any nonpolar chemical substance that is a viscous liquid at ambient temperatures. Depending on their chemical composition, a distinction may be drawn between fatty, essential mineral and silicone oils. Fatty oils include liquid, semisolid and solid products of vegetable and animal origin. They are also known as sweet oils. Mustard oil is of vegetable origin and is obtained from seeds of the black and white (Synapsis alba) mustard plants. In the crude state, black mustard oil (Brassica nigra, light color) has a spicy odor and a pungent taste. When refined it is neutral in odor and taste. White mustard oil (yellow color) has a bitingly pungent taste due to the allyl mustard oil it contains. On the other hand, though the cottonseed oil is also of vegetable origin and its obtained from cottonseeds, as a crude oil, it is dark and cloudy brown. However, it is yellowish, high-grade edible oil when refined. Cottonseed oil exhibits only a slight tendency to evaporate because of its viscous nature. Its commercial significance is in decline, as producers are increasingly dispatching fatty acids as semif-inished products. (Deutscher Transport-Versicherungs-Verband, 1999-1994).

The viscosity of fluids is determined both by collision among particles and by the force fields which determines interactions among molecules. The theoretical description of viscosity is therefore quite complex (Ben Haj-Kacem R., Ouerfelli N., J. Herraez, M. Guettari, H. Hamda, and M. Dallel, 2014). Hence, several models have been proposed in literature based on Eyring theory and other empirical relations that are not always applicable to a large number of mixtures (Noureddine Ouerfelli, Moncef Bouaziz & Herráez J.V., 2013; Herraez J. V., Belda R., O. Diez O, and Herraez M., 2008; Lee L. S. and Lee Y.S, 2001; Qunfang L. and Yu-Chun H., 1999;

Macías-Salinas R., García-Sánchez F. and G. Eliosa-Jiménez, 2003; Belgacem, F. B. M., 2001. Vegetable oils are very relevant for their nutritional purposes and for several industrial products spanning across fuels, skin care products, flow measurement, heat exchange, sterilization, freezing, and numerous operations, high pressure lubricants and alkyd resins for paint (Rewolinski, C., 1985; Agbo, N. G., Chatigre, K.O., Simard, R.E.; 1992; Kamel, B.S., Kakuda, Y., 1992; Noureddini, H., Teoh, B.C., Clements, L.D., 1992; Ibemesi, 1993; Foidl, N., Foidl, G., Sanchez, M., Mittelbach, M., Hackel, S. 1996; Eromosele C.O. and Paschal N. H., 2002). They are used in frying, cooking, salad dressing, shortening of pasty etc (Bakker N, Van't P, and Zoc L, 1997). They mainly consist of lipids with some other minor components including antioxidants, colorants, flavors and emulsifiers (Fasina O, Hallman C, and Clementsa, C., 2006). Some of these compounds occur naturally and some are added during the manufacturing process. The presence of hydrocarbons or mineral oils such as n-alkenes in vegetable oils has also been reported (Johnson S, and Saikia N. (2009). Vegetable oils act as carriers of fat-soluble vitamins (A, D, E, and K) and play important sensory and functional roles in food products. They provide the most concentrated source of energy, supply essential fatty acids linoleic and linolenic acids which are precursors for important hormones, the prostaglandins and responsible for growth, provide satiety and increases the palatability of food (Atasie V, Akinhanmi T, and Ojiodu C. (2009).

Previous studies (Noureddini et al., 1992; Abramovic H. and Klofutar C., 1998; Santos J. C. O, Santos I. M. G., and Souza A. G., 2005; Fasina O. O., and Z. Colley Z., 2008; Bhatia, V.K., Chaudhry, A., Sivasankaran, G.A., Bishi, R.P.S., Kashyap, M., 1990; Kapseu, C., Kayem, G.J., Balesdent, D., Schuffenecker, L., 1991; Kashulines, P., Rizvil, S.S.H., Harriott, P., Zollweq, J.A.,1991; Liew, K.Y., Seng, C.E., Lau, E.K ., 1991; Dandik, L., Sahin, E., Karaosmanglu, F., Isigigur, A., Aksoy, H.A., 1992; Noureddini et al., 1992) had based on the viscosities of different oils, organic compound, water and their derivatives as a function of temperature, storage age, and intensity of light, pressure, molecular weight and density of liquid. Also it has been shown that some plants have seeds

whose oils may be of immense values (Eromosele, I.C. and Eromosele, C.O., 1993; Eromosele, I.C., Eromosele, C.O., Akintoye, A.O., Komolafe, T.T., 1994; Eromosele, C.O., Okieimen, F.E.., 1998). It has also been well established that temperature has a strong influence on the viscosity of fluid products with viscosity generally decreasing with increase in temperature (Rao, M.A., 1999). Several researchers have reported the viscosity of vegetable oils at room temperature (Timms, R.E., 1985; Krishna, A. G. G., 1993; Rao, 1999). At higher temperature, oil must have sufficient viscosity to carry loads. Hence heavier oils are used at higher temperature. Similarly, light oils are used at low ambient temperature. Studies have also been carried out on the effect of temperature on the viscosity of some vegetable oils at temperatures less than 110 °C

(Coupland, J.N.; McClements, D.J.; 1999; Toro-Vazquez, J. F.; Infante-Guerrero, R., 1993; Rao, 1999) and at temperatures between 150–180°C (Miller, K. S.; Singh, R. P.; Farkas, B. E., 1994). Viscosity is the property of a fluid that determines its resistance to flow. It is an indicator of flow ability of lubricating oil and through its film strength, viscosity values are used in evaluating load carrying capacity, in denoting the effect of temperature changes and for determining the presence of contaminants in used oil during service. Absolute viscosity values are required in all ball bearing design calculations and in the technical design of other lubrication engineering.

Theoretical Framework

We have had a good number of equations for correlating the viscosity of liquids with temperature using experimental data. This can be shown as follows;

$$In \, \eta = A + \frac{B}{T+C} + a \log T + bT + cT^2 + \frac{D}{T^2} + \frac{E}{T^3} + \frac{F}{T^n} \qquad 1$$

In order to establish the relationship between viscosity and temperature, the following models can be extracted from equation 1;

$$In \, \eta = A + \frac{B}{T} \qquad 2$$

$$In \, \eta = A + \frac{E}{T^3} \qquad 3$$

$$In \, \eta = A + \frac{E}{T^n} \qquad 4$$

$$In \, \eta = A + \frac{B}{T} + \frac{D}{T^2} \qquad 5$$

$$In \, \eta = A + a \log T \frac{B}{T} \qquad 6$$

$$In \, \eta = A + \frac{B}{T} + bT \qquad 7$$

$$In \, \eta = A + \frac{B}{T+C} \qquad 8$$

De Guzman proposed an equation to represent dynamic viscosity of liquids as a function of temperature (De Guzman J., 1913; Andrade E. N. C, 1930), this is given by;

$$\eta = A e^{\frac{B}{T}} \qquad 9$$

Abramovic suggested a new formulation to represent the dynamic viscosity as a function of temperature;

$$Log \, \eta = \frac{A}{T} - B \qquad 10$$

$$\eta = A - B Log t \qquad 11$$

In addition, Abramovic adopted a multi-constant equation known as the Andrade equations, which are expressed as;

$$Ln \, \eta = A + \frac{B}{T} + \frac{C}{T^2} \qquad 12$$

$$Ln \, \eta = A + \frac{B}{T} + CT \qquad 13$$

Natarajan used the Antoine type equation which is given by;

$$Log \, \eta = \frac{B}{C-T} + A \qquad 14$$

Furthermore, Reid (Reid R. C., Prausnitz J. M., Poling B. E., 1987) used a polynomial equation to represent the dynamic viscosity of liquids as a function of temperature in the form;

$$Ln\,\eta = A + \frac{B}{T} + CT + DT^2 \qquad 15$$

Reid in his study determined the constants A, B, C and D for several substances [8]

Furthermore, there are multi-constant equations that models the properties of Arrhenius equations and its parameters, such fluids include melting salts, glasses and metals, ionic liquids, heavy and vegetable oils, fuels and biofuels, etc (Viswanath D. S., Ghosh T. K., Prasad G. H. L., Dutt N. V. K. and Rani K. Y., 2007). The Arrhenius type' equation is expressed as;

$$\eta = A e^{\frac{E_a}{R}} \qquad 16$$

In this study, Ea and A are temperature dependent parameters, hence the Arrhenius-type equation is generally replaced by VTF-type equation (Vogel H., 1921; Fulcher G. S., 1925 and Tammann G, W. Hesse, Z. Anorg, 1926) expressed as;

$$\eta = A e^{\frac{E_a}{R(T-T_0)}} \qquad 17$$

Where **η** is the dynamic viscosity measured in centi Poise, T is the temperature in Kelvin; and t is the temperature in degrees Celsius. A, B, C D, E, E_0, and T_0 are constants to be determined in each case. The magnitude of these constants for olive oil and other oils have been provided by Abramovic (Abramovic, 1998)

The Arrhenius exponential relations is used in expressing the relationship between viscosity and the reciprocal of absolute temperature of some physicochemical properties such as chemical rate, diffusion, electrical conductivity, gas kinetics and viscosity. The pre-exponential factor in particular is correlated to motion, rate, disorder, speed, and entropy whereas the other important parameter (Ea) denotes the activation energy or a gap where the studied phenomenon is expected to proceed through an intermediate "transition state".

Methodology
Materials: The vegetable oil is available in local supermarkets and specialty shops. All oil was stored at room temperature (at about 20 °C) and in a dark place before the analysis. The chemicals used in cleaning or removing traces of solvent are suitable organic solvent like CCl₄, ether, petroleum spirit or benzene. The apparatus used were Red Wood Viscometer, stop watch, Kohlrausch flask, thermometer, filter paper. Prior to the experiment, the oil was thoroughly filtered through a muslin cloth to remove solid particles that may clog the jet.

Methods: The viscometer was leveled with the help of leveling screws and the outer bath was filled with water for determining the viscosity at 90 °C and below. The jet was closed with a ball valve and then the test oil was poured into the cup, up to the tip of the indicator. A clean dry Kohlrausch flask was immediately placed below and directly in line with discharging jet while a clean thermometer, a stirrer was inserted into the cup and covered with a lid. The water bath was heated slowly with constant stirring and when the oil in the cup reached a certain temperature, the heating was stopped. The ball valve was lifted, the oil from the jet flows into the flask as the stop-watch was immediately engaged. The stop watch was disengaged when the lower meniscus of the oil reaches the 50 ml mark on the neck of receiving flask as we record the time taken. The same procedures were repeated to get more readings at different temperatures. Some empirical relations were found to describe the temperature dependence of the viscosity. The experimental data were fitted and the correlation constants of the best fits were estimated.

The dependence of the shear viscosity of a fluid with temperature has been shown to be in the general form of the Arrhenius-type for various classic solvents (equation 17). Though the equation was initially written by De Guzman (1913), but was further developed by Eyring H. (1936) and Eyring H. and Hirschfelder J. (1937) for numerous Newtonian classic solvents (Fasina and Colley, 2008), which can be expressed as:

$$In\ \eta = \left(\frac{E_a}{R}\right)\frac{1}{T} + In\ A \qquad\qquad 18$$

where R, E_a, and A are respectively the gas constant (8.31 JK^{-1}mol^{-1}), the Arrhenius activation energy, and the pre-exponential factor for the liquid system. There is no negative sign in front of E_a since equation 18 is derived from viscosity and not from the rate of the phenomenon as in the original Arrhenius equation.

The plot of the logarithm of viscosity versus the reciprocal of absolute temperature (1/T) for numerous liquid systems is practically linear and the Arrhenius parameters, which are the activation energy (E_a) and the pre-exponential factor (A), are thus independent of temperature over the temperature ranges (from 278.15 to 328.15) K approximately around the room temperature at constant atmospheric pressure. Using graphical method, the activation energy (Ea) is obtained by multiplying the slope of the graph by the gas constant R while the pre-exponential factor A is deduced via extrapolation by estimating the antilogarithm of the intercept.

In furtherance of the Arrhenius parameters estimation, is the third termed the Arrhenius temperature (T_a) deduced from the intercept with the abscissa of equation 18 related to the reciprocal of absolute temperature as:

$$T_A = \frac{-E_a}{R\ In\ A} = \frac{-E_a}{R} x \frac{1}{intercept} \qquad 19$$

Dividing the slope of equation 18 by the natural logarithm of the pre-exponential factor A, gives the Arrhenius temperature T_A.

Equation (19) simplifies the viscosity-temperature dependence following the (Dean J. A., 1987; Viswanath *et al.,* 2007; Eyring, 1936; Eyring and Hirschfelder, 1937) equation as;

$$In\ \eta = \left(\frac{E_a}{R}\right)\left(\frac{1}{T} - \frac{1}{T_A}\right) \qquad\qquad 20$$

For homogenous dimensions, we can calculate another parameter known as the Arrhenius activation temperature (T*) as:

$$T^* = \frac{E_a}{R} \qquad\qquad 21$$

Equation 18 can be derived from the thermodynamic standpoint where the flow is seen as a local transition of a molecule or a group of molecules between one state (position before flowing) to another (position after flow occurred) having to overcome an energy barrier. One of the most common models assumes that the flow is controlled by the presence of free volume enabling molecules to jump from one place to another. Such mechanism implying a relation between free volume and viscosity was first empirically propounded by Batschinski, A. J. (1913). For small molecules where forces resisting flow are mainly linked to interaction forces, there is a good correlation between activation energy (Ea) and the heat of vaporization at temperatures where there are large fractions of free volume (Vinogradov, G. V. and Malkin, Ya, A., 1980).

The focus of the present study is to investigate the physicochemical behaviours of two samples of vegetable oil and also the dependence of their viscosities on temperature. The relationship between the viscosities of mustard oil and cotton seed oil with temperature variations will be estimated by fitting the relevant equations.

Results and Discussion
Figures 1 to 4 indicate the obtained results from the research:

Table 1: Viscosity of Mustard oil

T (°C)	η (cP)	ln η	1/T (/°C)
90	20.35	3.01331	0.01111
85	20.97	3.04328	0.01176
80	21.69	3.07699	0.0125
75	22.54	3.11519	0.01333
70	23.54	3.15886	0.01429
65	24.76	3.20924	0.01538
60	26.26	3.26802	0.01667
55	28.15	3.33748	0.01818
50	30.60	3.42084	0.02
45	33.88	3.52272	0.02222
40	38.48	3.65008	0.025
35	45.32	3.81381	0.02857
30	56.38	4.03213	0.03333
25	73.54	4.29778	0.04
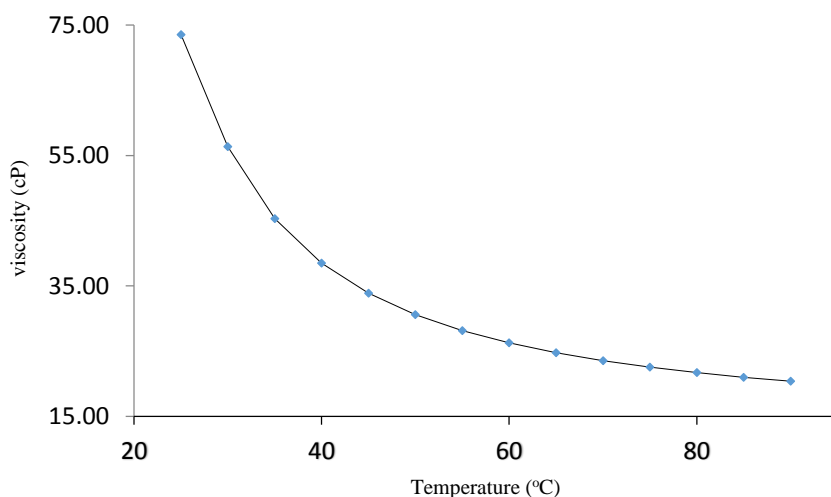
**Figure 1:** *Variation of Viscosity (Centipoise) of the Mustard oil with Temperature (°C)*

$$y = 45.051x + 2.5171$$
$$R^2 = 0.9996$$
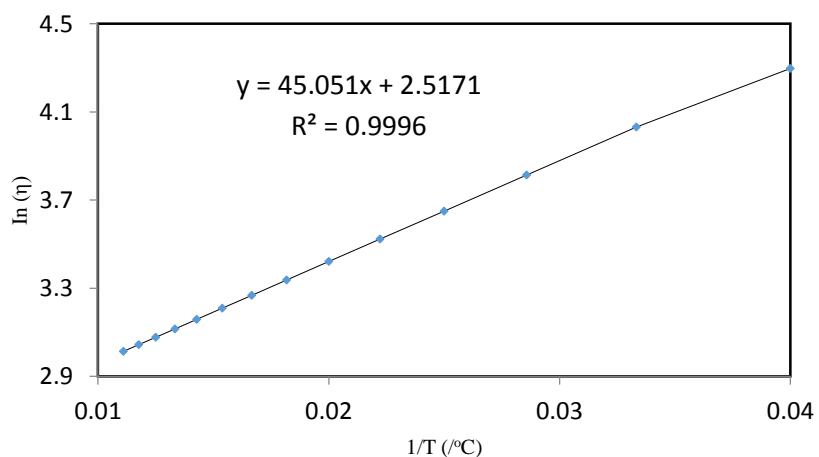
Figure 2: *Natural Log of Viscosity (In η) Versus Inverse of Temperature (°C⁻¹) for Mustard oil*

Table 2: Viscosity of Cotton Seed Oil

T (°C)	η (cP)	In η	1/T (/°C)
90	14.86	2.698878	0.011111
85	15.41	2.735171	0.011765
80	16.05	2.776	0.0125
75	16.82	2.822273	0.013333
70	17.73	2.875157	0.014286
65	18.84	2.936177	0.015385
60	20.23	3.007367	0.016667
55	22.01	3.0915	0.018182
50	24.35	3.19246	0.02
45	27.55	3.315856	0.022222
40	32.14	3.4701	0.025
35	39.19	3.668414	0.028571
30	51.05	3.932833	0.033333
25	71.02	4.26302	0.04
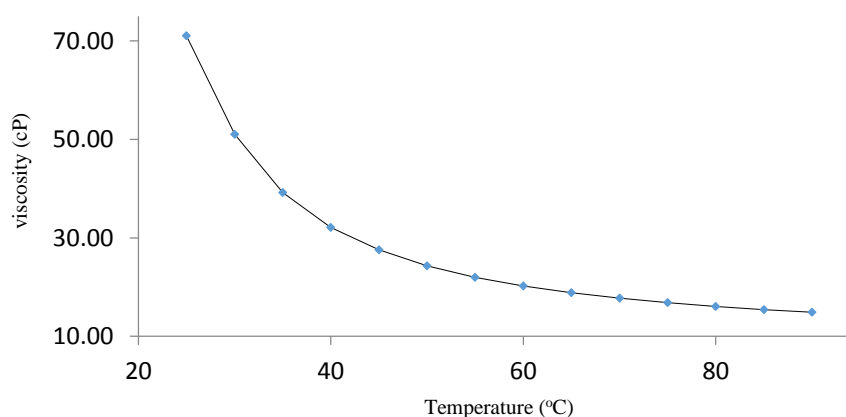
Figure 3: *Variation of Viscosity (Centipoise) of the Cotton Seed Oil with Temperature (°C)*
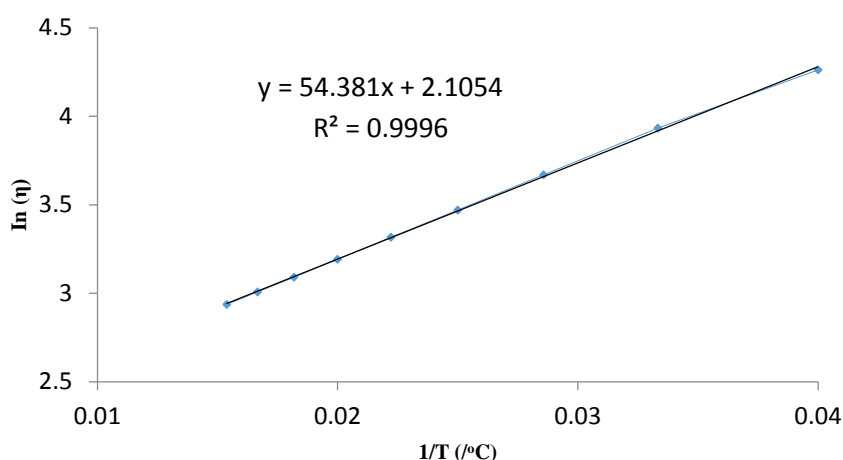
Figure 4: *Natural Log of Viscosity (In η) Versus Inverse of Temperature (ºC⁻¹) for Cotton Seed Oil*

Viscosity measurements were carried out on mustard oil and cotton seed oil in order to determine their viscosity as a function of temperature, from which other relevant parameters were estimated using equations 1, 2 and 4. The Activation energy E_a, Entropic (pre-exponential) factor A, Arrhenius temperature T_A and the Arrhenius activation temperature for the mustard oil are 374.37381 J/mole, 12.39260595 cP, -17.89797783 ºC, 45.051 ºC respectively. On the other hand, the results for the cotton seed oil shows 451.90611 J/mole, 8.210386507 cP, -25.8292961 ºC, 54.381 ºC respectively for the Activation energy E_a, Entropic (pre-exponential) factor A, Arrhenius temperature T_A and the Arrhenius activation temperature. The <u>activation energy</u>, E_a, is the minimum energy molecules must possess in order to react to form a product while the pre-exponential factor is related to the amount of times molecules will strike in the orientation necessary to cause a reaction. The Arrhenius type equation which is based on the collision theory states that particles must collide with proper orientation and with enough energy for the reaction to proceed. The coefficients of regressions (R^2) for the graph of the natural log of viscosity versus reciprocal of temperature (Figures 2 and 4) for the mustard oil and cotton seed oil are 0.9996 and 0.9996 respectively.

The coefficient of correlation for both samples indicates very good fit. Most importantly, the aforementioned properties of our oil samples show a linear behaviour by obeying the Arrhenius viscosity model

(Equation 4) and there appears a remarkable statistical non-parametric correlation between the activation energy (E_a) and the entropic factor (A), so also the Arrhenius temperature (T_A) for both oils. Results used in plotting the graph in this work showed that the model which best fits the relationship between the Arrhenius parameters is a logarithmic type (Figures 2 and 4) correlating the activation energy (E_a) to the Arrhenius temperature (T_A) (Aymen M., Nesrine D, Hatem H., Fethi B. M. B., Yousry H. A. , Noureddine O. and Ahmed H. H., 2015).). The results also reaffirms that temperature affects the viscosity of vegetable oils at temperatures between 25–90 °C, with viscosity decreasing exponentially (Figure 1 and 3) and linearly with increase in temperature (Figures 2 and 4). Concerning the hydraulic calculations of fluid transportations and for energy, we have reduced the model using single variable without losing significant accuracy. The equation used in the present work allows for redefining the Arrhenius equation by using a single parameter (Ben Haj-Kacem *et al.,* 2012 and Kirkwood *et al.,* 1949) instead of two (Irving, 1977) using experimental viscosity data at several temperatures (Tables 1 and 2). We hereby re-emphasize that this model would be very useful in different fields of physical and chemical sciences (Ike and Ezike, 2018), especially in measuring an unknown parameter when the other quantity is available in engineering sciences (Das *et al.,* 2012) or evaluated by some relevant theories suggested in the

literature (Viswanath *et al.,* 2007). From the results, it can be concluded that viscosity increases exponentially when the activation energy decreases (Figures 1 and 3). Because viscosity with small activation energy does not require much energy to reach the transition state, it should proceed faster than a reaction with larger activation energy. Also, viscosity or chemical reactions occur more rapidly at higher temperatures. This is because thermal energy controls the motion at the molecular level. As the temperature rises, molecules move faster and collide more vigorously, greatly increasing the likelihood of bond cleavages and rearrangements.

Conclusion

From the viscosities data of two vegetable oils (mustard oil and cotton seed oil) over different temperature ranges, we have been able to determine values of the two viscosity Arrhenius parameters such as the activity energy (Ea) and the entropic factor. The overall results suggest some potential applications for the seed oils. From the viscosity-temperature profiles the vegetable oils are edible/table oil and can be utilized as lubricants, burning oil, in soap production, biodiesel, in margarine production (especially cotton seed oil) and generally as an oil base stock, although it may be necessary to examine their viscosities under extreme temperatures. It can be concluded from this study that temperature affects the viscosity of vegetable oils at temperatures between 25–90 °C, with viscosity decreasing exponentially (Figure 1 and 3) and linearly with increase in temperature (Figures 2 and 4). All the deductions obey the linear Arrhenius behavior. Here, we used statistical approach to analyze eventual correlation between the Arrhenius parameters for both oils. From the analysis of the graph plots, we found that there is significant statistical nonparametric correlation between the activity energy (Ea) and the entropic factor (A), as well as other related parameters; Arrhenius temperature (T) and Arrhenius activation temperature (T*). Also, the proposed equation allows us to redefine the Arrhenius equation using a single parameter instead of two through the use of experimental viscosity

values at several temperatures for two vegetable oils. We concluded that this model would be very important in several branches of engineering especially the chemical sciences and will allow the estimation of one non-available parameter when the second one is available, or otherwise estimated by some theories suggested in literatures (Viswanath *et al.,* 2007). It is our utmost expectation that the present study avails deeper insight into the investigation of other relevant fields such as organic liquids solvents (alcohols, ketones, amides etc.) and inspires theorists to merge some previous theoretical approaches. In addition, we hope that it will be useful in large domains of applied chemistry and engineering. Furthermore, further studies on the relationship between the Arrhenius parameters and properties of other liquid systems can prove the fitness of the method in predicting the properties of other non-treated fluid systems.

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