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Theoretical and experimental study of honey's viscosity in three Southern States of Nigeria: Application of Vogel-Tamman-Fulcher (VTF) and power law (PL) models

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Abstract--In this study, we employed Power-law (PL) and Vogel, Tamman and Fulcher (VTF) models, to evaluate viscosity as honey's property of rheological origin. Five samples of local market honey and three samples of farm honey collected from some states in Southern Nigeria were evaluated. Using the linearization of these models, all necessary constants in the resulting equations were obtained. The assessment of these models for their suitability in this study were carried out on the basis of regression analysis, with the aid of resulting correlation coefficients (R). The results obtained showed that the values of correlation coefficient of these models were suitable for the study of viscosity using the samples of honey as a major rheological property owing to their good correlation coefficients. The obtained results qualitatively agree with those discussed in the literature.

Keywords--honey, viscosity, glass transition temperature, rheological properties, VTF, PL.

Introduction

Honey is a sweet substance of nature with an aromatic origin and is usually produced by bees by using the sugar obtained from flowering plants. It can also be produced by using the excretory materials of insects which feed on plants as the raw material. This is then collected and converted by the bees for the benefit of man [1]. There are different species of honey from bees found all over the world. However, the major variety in Nigeria is the *Apis mellifera* [2]. Though there are a number of sources of honey, the two major sources are those which are obtained from the nectars of flowers and those which result when plants living parts are secreted [3]. While the former is referred to as blossom or nectar or floral honey, the latter is known as honeydew honey. In a situation where a mixture of the two arises, it is proper to call it compound honey. Floral honey specie may be broadly divided into two sub-groups namely, monofloral honey specie and multifloral honey specie. While monofloral honey is the type of honey which is mainly produced from the flowers of a single plant, multifloral honey refers to the honey produced from different plants [3].

Honey's viscosity is an important index used to access the sensory quality of the product and other aspects related to its operations, like the heating, mixing, filtering, transportation and bottling [4]; [5]; [6]; [7]; [8]. The behaviour of rheological attributes of honey can be described by a number of mathematical models all of which focus generally on the description of viscosity as a function of temperature [9]. This viscosity-temperature connection has been described by different equations employed in several studies. The models include those of Arrhenius, William, Landel and Ferry (WLF), Vogel, Tamman and Fulcher, (VTF). The power-law (PL) model is yet another useful model for the study of the rheological properties of honey [10].

Among the different models, the simplest equation to describe temperature dependence of viscosity is the Arrhenius equation [11]. However, there are some variation in the result from experiment and a given model for some foods. In most studies, the importance of the VTF and WLF equations are emphasized because of the inclusion of glass temperature of transition which is a reference temperature obtainable from experiments. However, evaluating the viscosity of the sample at the glass temperature of transition poses a major problem to the use of these equations [11].

The Arrhenius model takes the form given by equation (1):

$$\mu = \mu_0 \cdot \exp \left[-\frac{E_a}{R.T} \right] \quad (1)$$

where μ is viscosity (Pa.s), μ_0 is material constant (Pa.s) which defines the viscosity of the sample just before temperature influences it, T is absolute temperature (K), E_a is an exponential constant which is used to indicate the stability of the honey sample [12] and it is known as the flow activation energy per mole (Jmol^{-1}), and R is the universal gas constant the unit of measurement of which is $\text{Jmol}^{-1}\text{K}^{-1}$ [7]; [13]; [14].

The material constant, μ_0 and the flow activation energy, E_a can be calculated by solving equation (1)

$$\ln\mu = \ln\mu_0 + \ln e \left[-\frac{E_a}{R.T} \right] \quad (2)$$

$$\ln\mu = \ln\mu_0 - \left[-\frac{E_a}{R.T} \right] \ln e \quad (3)$$

$$\ln\mu = \left[-\frac{E_a}{R} \right] \frac{1}{T} + \ln\mu_0 \quad (4)$$

Equation (4) can be compared with the general equation of a straight line which takes the form

$$y = mx + c \quad (5)$$

With this comparison, it can be seen that plotting the graph of $\frac{1}{T}$ on the horizontal axis and $\ln\mu$ on the vertical axis, the antilog of the intercept will give μ_0 and the slope of the straight line will give the value of $-E_a/R$. Since R is a constant whose value is $8.314 \text{ m}^2 \text{ kg K}^{-1} \text{ mol}^{-1}$, E_a can be readily obtained. This dependence is said to describe honey viscosity with a relatively small error less than 5 % [9].

In relation to temperature most of the time, honey viscosity is described by the WLF equation and employs the use of such parameters as glass temperature of transition and glass state viscosity [9].

According to the WLF model,

$$\ln \left[\frac{\mu}{\mu_g} \right] = \frac{-C_1(T-T_g)}{C_2+(T-T_g)} \quad (6)$$

Where μ is viscosity (Pa.s); μ_g is material constant, which is the viscosity corresponding to the glass temperature of transition, (T_g); T is absolute temperature (K).

Several definitions for the glass temperature of transition have been reported [15]. Among the various definitions, one common definition which can be adopted for T_g is the one reported by Debenedetti and his colleague, Stillinger. According to this definition, the glass temperature of transition of any fluid sample is the temperature at which its viscosity increases until it reaches 10^{12} Pa.s [16]. When the state of a food substance upon cooling changes from rubbery to glassy, glass transition in the food occurs and the temperature range at which these changes occur is known as the glass temperature of transition [10]. The information on the glass temperature of transition (T_g) is needed in quality assurance as well as stability and safety of various food products. Based on the obtained value of glass temperature of transition, it is possible to investigate the rheological attribute of honey and analyze its adulteration attempts [5]; [17].

Glass transition in honey is an important predictor which reveals the tendency of the honey sample to granulate or crystalize and it is very useful in the honey

industry. However, honey granulation is a very important topic of consideration especially during the cold season of the year amongst honey processors [18]; [19]. Also, this glass temperature of transition of honey samples is a very important parameter in the production of granulated honey, which is a source of instant energy for sports men and women for example. Equation (6) can be further transformed into the form of a straight line as follows:

$$\begin{aligned} \frac{1}{\ln\left(\frac{\mu}{\mu_g}\right)} &= \frac{1}{\frac{-C_1(T-T_g)}{C_2+(T-T_g)}} \\ \frac{1}{\ln\left(\frac{\mu}{\mu_g}\right)} &= \frac{C_2+(T-T_g)}{-C_1(T-T_g)} \\ \frac{1}{\ln\left(\frac{\mu}{\mu_g}\right)} &= \left(-\frac{C_2}{C_1}\right)\left(\frac{1}{T-T_g}\right) + \left(\frac{-1}{C_1}\right) \end{aligned} \quad (7)$$

Upon plotting the graph of $\frac{1}{\ln\left(\frac{\mu}{\mu_g}\right)}$ on the vertical axis and $\left(\frac{1}{T-T_g}\right)$ on the horizontal axis, the slope (gradient) of the graph will give $\left(-\frac{C_2}{C_1}\right)$ and the vertical intercept will represent $\left(\frac{-1}{C_1}\right)$. Thus, the values of the constants C_1 and C_2 can be obtained.

Another mathematical relation which is being used to describe honey rheological properties as it relates to temperature including the glass temperature of transition is given by the VTF equation that takes the form of equation (8), where A and B are constants, T is temperature and the glass temperature of transition is T_g is [9]; [19]:

$$\mu = A \cdot \exp\left[\frac{B}{(T-T_g)}\right] \quad (8)$$

Equation (8) may be solved to obtain

$$\ln\mu = \ln A + \ln \exp\left[\frac{B}{(T-T_g)}\right] \quad (9)$$

$$\ln\mu = \left[\frac{B}{(T-T_g)}\right] + \ln A$$

$$\ln\mu = B \left[\frac{1}{(T-T_g)}\right] + \ln A \quad (10)$$

Thus, plotting the graph of $\ln\mu$ on the vertical axis and $\left[\frac{1}{(T-T_g)}\right]$ on the horizontal axis, the values of the constants A and B in the VTF equation can be readily obtained. The PL model is yet another useful model to study the rheological properties of honey. Just like the WLF and VTF models, this model also makes use of such parameter as the glass temperature of transition, T_g . According to the Power-law model,

$$\mu = A(T - T_g)^B \quad (11)$$

where A and B are constants, T is temperature and T_g is the glass temperature of transition. When solved, equation (11) yields

$$\ln\mu = \ln A(T - T_g)^B$$

$$\ln\mu = B \ln(T - T_g) + \ln A \quad (12)$$

A plot of $\ln\mu$ against $\ln(T - T_g)$ can then be obtained and from such a plot, the value of A and that of B can be found. Generally, the suitability of any model for experimental data is usually assessed by various statistical approaches, using the coefficient of correlation (R) or determination (R^2) obtained from regression analysis [10].

Materials and Methods

Materials

Three farm and five market honey samples were used in this study. The three farm honey samples were purchased from Okunigho-Jesse in Delta State, Akure in Ondo State and Benin City in Edo State and were coded sample 1, 2 and 6 respectively. The market honey samples were sourced from Saki in Oyo State and labeled sample 3, Nsukka in Enugu State and labeled sample 4, Okirighwre in Delta State and labeled sample 5, Boru in Oyo State and labeled sample 7, and Amukpe in Delta State and labeled sample 8.

Determination of Rheological Properties

The VTF and PL models considered in this study require that the viscosity and glass temperature of transition of the honey samples be known. Therefore, the viscosity of the samples of honey was determined with a Digital Rotary Viscometer (Model NDJ-5S, M & A, Instruments Inc., Shanghai China), employing the method described by [20]; [21]. The glass temperature of transition of the honey samples was determined using the method described by [18].

Results and Discussion

Results

Table 1 Parameters for utilization of the VTF and PL Models for the Rheological Properties of Honey Sample 1

T (K)	T - T _g (K)	1/T - T _g × 10 ⁻³ (K ⁻¹)	ln(T - T _g)	lnμ
273.15	56.37	17.7	4.03	8.32
283.15	66.37	15.1	4.20	8.19
293.15	76.37	13.1	4.34	8.17
303.15	86.37	11.6	4.46	8.09
313.15	96.37	10.4	4.57	8.01
323.15	106.37	9.4	4.67	7.93
333.15	116.37	8.6	4.76	7.85

Table 2. Parameters for utilization of the VTF and Power-Law Models for the Rheological Properties of Honey Sample 2

T (K)	T - T_g (K)	1/T - T_g × 10⁻³ (K⁻¹)	ln(T - T_g)	lnμ
273.15	54.00	18.5	2.92	8.08
283.15	64.00	15.6	2.75	8.00
293.15	74.00	13.5	2.60	7.94
303.15	84.00	11.9	2.48	7.85
313.15	94.00	10.6	2.36	7.77
323.15	104.00	9.6	2.26	7.69
333.15	116.37	8.7	2.16	7.61

Table 3. Parameters for utilization of the VTF and Power-Law Models for the Rheological Properties of Honey Sample 3

T (K)	T - T_g (K)	1/T - T_g × 10⁻³ (K⁻¹)	ln(T - T_g)	lnμ
273.15	52.14	19.2	2.95	7.56
283.15	62.14	16.1	2.78	7.31
293.15	72.14	13.9	2.63	6.82
303.15	82.14	12.2	2.50	6.44
313.15	92.14	10.9	2.39	5.93
323.15	102.14	9.8	2.28	5.33
333.15	112.14	8.9	2.19	7.56

Table 4. Parameters for utilization of the VTF and Power-Law Models for the Rheological Properties of Honey Sample 4

T (K)	T - T_g (K)	1/T - T_g × 10⁻³ (K⁻¹)	ln(T - T_g)	lnμ
273.15	61.89	16.2	2.79	8.22
283.15	71.89	13.9	2.63	7.49
293.15	81.89	12.2	2.50	6.79
303.15	91.89	10.9	2.39	6.07
313.15	101.89	9.8	2.28	5.40
323.15	111.89	8.9	2.19	4.67
333.15	121.89	8.2	2.10	4.07

Table 5. Parameters for utilization of the VTF and Power-Law Models for the Rheological Properties of Honey Sample 5

T (K)	T - T_g (K)	1/T - T_g × 10⁻³ (K⁻¹)	ln(T - T_g)	lnμ
273.15	52.52	19.0	2.94	7.69
283.15	62.52	16.0	2.77	7.18
293.15	72.52	13.8	2.62	6.58
303.15	82.52	12.1	2.49	6.13
313.15	92.52	10.8	2.38	5.53
323.15	102.52	9.8	2.28	4.94
333.15	112.52	8.9	2.19	4.49

Table 6. Parameters for utilization of the VTF and Power-Law Models for the Rheological Properties of Honey Sample 6

T (K)	T - T_g (K)	1/T - T_g × 10⁻³ (K⁻¹)	ln(T - T_g)	lnμ
273.15	61.87	18.9	2.94	7.77
283.15	71.87	15.9	2.76	7.11
293.15	81.87	13.7	2.62	6.55
303.15	91.87	12.1	2.49	5.91
313.15	101.87	10.8	2.38	5.61
323.15	111.87	9.7	2.26	5.05
333.15	121.87	8.9	2.19	4.64

Table 7. Parameters for utilization of the VTF and Power-Law Models for the Rheological Properties of Honey Sample 7

T (K)	T - T_g (K)	1/T - T_g × 10⁻³ (K⁻¹)	ln(T - T_g)	lnμ
273.15	52.92	18.9	2.94	7.77
283.15	62.95	15.9	2.77	7.11
293.15	72.95	13.7	2.61	6.55
303.15	82.95	12.1	2.47	5.91
313.15	92.95	10.8	2.38	5.61
323.15	102.95	9.7	2.27	5.05
333.15	112.95	8.9	2.19	4.64

Table 8. Parameters for utilization of the VTF and Power-Law Models for the Rheological Properties of Honey Sample 8

T (K)	T - T_g (K)	1/T - T_g × 10⁻³ (K⁻¹)	ln(T - T_g)	lnμ
273.15	52.96	18.9	2.94	7.46
283.15	62.96	15.9	2.75	7.04
293.15	72.96	13.7	2.62	6.50
303.15	82.96	12.0	2.48	5.90
313.15	92.96	10.8	2.38	5.48
323.15	102.96	9.7	2.27	4.93
333.15	112.96	8.9	2.18	4.53

Table 9. Values of the constants and R² for with respect to the VTF and Power-Law Models

Honey Sample Code	(VTF Model)			(PL Model)		
	A	B	R²	A	B	R²
1	7.486	0.048	0.943	10.805	0.638	0.971
2	7.261	0.047	0.921	6.294	0.640	0.938
3	5.056	0.126	0.277	2.796	1.570	0.236
4	0.266	0.511	0.966	8.586	6.093	0.990
5	1.947	0.319	0.953	4.784	4.355	0.987
6	6.669	0.059	0.963	4.124	0.703	0.968
7	2.086	0.311	0.980	4.313	4.141	0.996
8	2.121	0.299	0.955	4.034	4.019	0.988

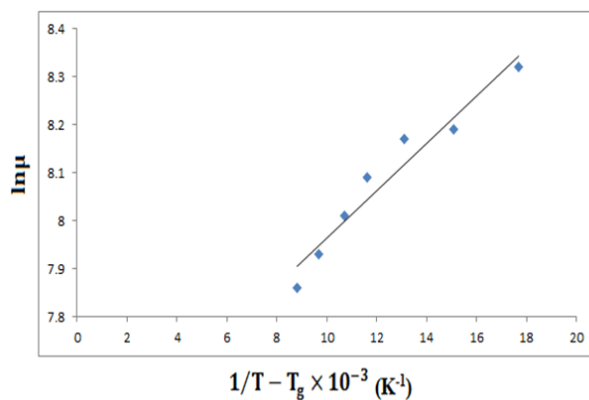


Figure 1. Graphical Description of the Rheological Properties of Honey Sample 1 using the VTF Model

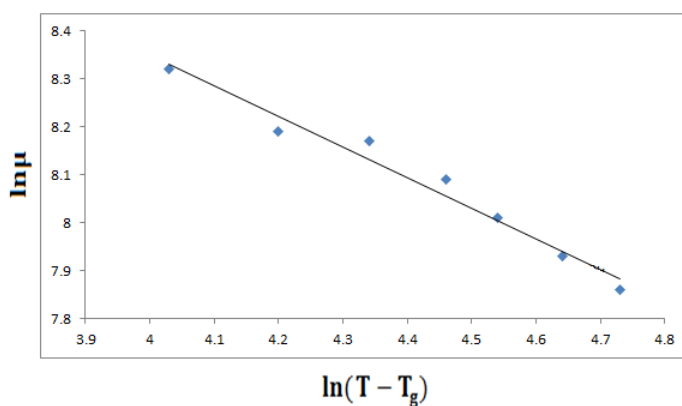


Figure 2. Graphical Description of the Rheological Properties of Honey Sample 1 using the Power-law Model

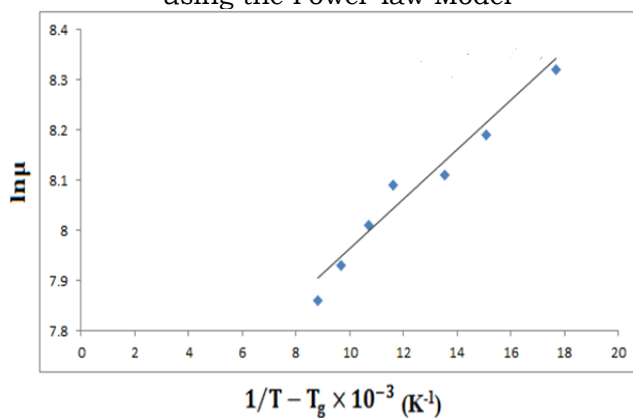


Figure 3. Graphical Description of the Rheological Properties of Honey Sample 2 using the VTF Model

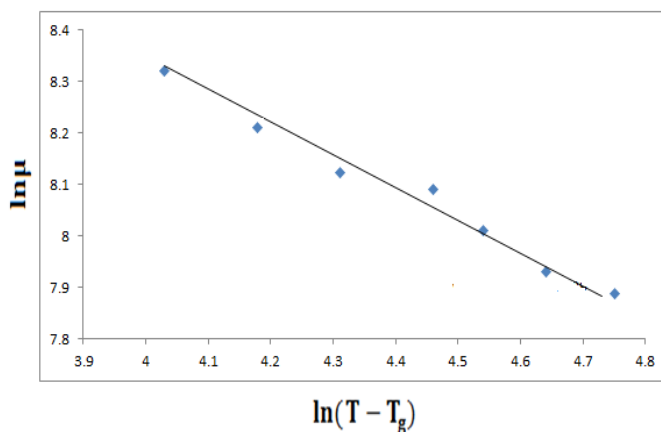


Figure 4. Graphical Description of the Rheological Properties of Honey Sample 2 using the Power-law Model

Discussion

The values of the various parameters used for both models to describe the rheological properties of the different honey samples in this research are shown in Tables 1-8, while the values of the constants in the equations describing the models are shown in Table 9, with their corresponding coefficient of determination, R^2 .

The application of the VTF model to honey sample 1 yields the values of 7.486 and 0.048 for A and B respectively while the coefficient of determination, R^2 is 0.943, which is about 94.3 %. With the PL model, these values are 10.805 and 0.638 for A and B respectively while the coefficient of determination, R^2 is 0.971 which is equal to 97.1 % as shown in Table 9. With the VTF model, the values of A and B are 7.261 and 0.047 with an R^2 value of 0.921, representing 92.1 %. The corresponding values of A, B and R^2 upon application of the PL model are 6.294, 0.640 and 0.938, respectively for honey sample 2 as described in Table 9. From Table 9, the values of A, B and R^2 on application of the VTF model are 5.056, 0.126 and 0.277 respectively while the corresponding values of A, B and R^2 when the PL is applied are respectively 2.796, 1.570 and 0.236 for honey sample 3. For honey sample 4, the values of A, B and R^2 on application of the VTF model are 0.266, 0.511 and 0.966 respectively, corresponding the values of 8.586, 6.093 and 0.990 for A, B and R^2 respectively when the PL model is applied as shown in Table 9. For honey sample 5, the values of A, B and R^2 on application of the VTF model are 1.947, 0.319 and 0.953 respectively while the corresponding values of A, B and R^2 when the PL is applied are respectively 4.784, 4.355 and 0.987. For honey sample 6, the respective values of A, B and R^2 on application of the VTF model are 6.669, 0.059 and 0.963 while the corresponding values of A, B and R^2 when the PL is applied are 4.124, 0.703 and 0.968, respectively. For honey sample 7, the values of A, B and R^2 when the VTF model is used are 2.086, 0.311 and 0.980 respectively while the corresponding values of A, B and R^2 when the PL model is applied are respectively 4.313, 4.141 and 0.996. With the VTF model, the values of A and B for honey sample 8 are 2.131 and 0.299 with an R^2 value of

0.955, representing 95.5 %. The corresponding values of A, B and R^2 when the PL model is applied are 4.034, 4.019 and 0.988, respectively.

Earlier, [10] had reported that the extent of suitability of any mathematical model for experimental data is a function of various statistical approaches and therefore, can be assessed by them, using the coefficient of correlation (R) or determination (R^2) obtained by performing some regression analysis. Relying upon this fact, the Power-law mathematical model can be considered to be relatively better than the VTF model for the assessment of the rheological properties of the honey samples in this study, considering the values of the coefficient of determination, R^2 obtained from these two models. However, either of these models could be used to describe the rheological properties of the honey samples in this study since both models give a good coefficient of correlation, R. For instance, since the value of R^2 for honey sample 1 according to the VTF model is 0.943, the value of R according to this model will be the square root of this value and this is equal to 0.97. Similarly, since the value of R^2 according to the PL model is 0.971, the value of R according to this model will be the square root of this value and this is equal to 0.99. Thus, the two models give a good coefficient of correlation [19]; [22]. However, some studies have shown that the equation which defines the power law model tends to infinity as T approaches T_g attain so that when both T and T_g are exactly the same in value, the equation becomes undefined. By implication, the physical meaning of the model in the predicting viscosity is defeated [23]. In this case, the VTF model is preferable to predict viscosity as a rheological property when cases of extrapolation arise.

However, plots of relevant parameters related to the applications of both VTF and PL models for the rheological properties of honey samples 1 and 2 in this study are shown in Figs. 1, 2, 3 and 4, and it is expected that similar behaviour is obtainable for the other honey samples. The plotted graphs are closely related with the results presented in the tables. For instance, in Figure 1 and Table 1, the relationship between viscosity and temperature is clearly demonstrated for honey sample 1. An increase in temperature gives rise to a decrease in velocity, resulting in the values of A and B presented in Table 9. Similar explanations hold for the other honey samples.

Conclusion

The study examined the application of Vogel-Tamman-Fulcher (VTF) and Power Law (PL) models for the description of honey rheological properties collected from some states in the six geopolitical zones of Nigeria. The suitability of any model for experimental data is usually assessed by various statistical approaches, using the coefficient of correlation (R) or determination (R^2) obtained from regression analysis. However, the study revealed that either of these models could be used to describe the rheological properties of the selected honey samples since both models give a good coefficient of correlation, R.

Availability of data and materials

Every data generated and materials in the course of this research are included in this article.

Competing interests

The authors declare that they have no competing interests.

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Authors' contributions

GK Agbajor carried out the design of the problem, did the literature review, carried out the methodology and did part of the writing. OOE Enaroseha did part of the writing, plotted the graphs, analyzed it and did the draft preparation. OD Osahon read the manuscript and made necessary corrections. The authors read and approved the final manuscript.

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