JASEM ISSN 1119-8362 Full-text Available Online at www.bioline.org.br/ja J. Appl. Sci. Environ. Mgt. *September*, 2006 Vol. 10 (3) 67 - 74

Effect of strong electrolytes on edible oils part II: vViscosity of maize oil in 1,4-dioxane in the presence of HCl, NaOH and NaCl at different temperatures

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ABSTRACT: The effects of strong electrolytes like HCl, NaOH and NaCl on the viscosity of maize oil at various temperatures (298 – 323 K) with the difference of 5 K using 1,4 dioxane as solvent were determined. The viscosity of oil was found to be increased with the increasing concentration of oil and decreases with the rise of temperature. The addition of electrolytes decreases the viscosity of oil although very little which shows that the electrolytes increase the distance between oil molecules and cause the enhancement of rate of flow and the increment of temperature drops the rate of flow of the solutions. Furthermore the concentration of electrolytes increases the viscosity of oil solutions. It is due to the presence of unsaturated ingredients present in the oil and thermal effect. The electrolytes behave as structure breaker. The effect of temperature was also determined in terms of fluidity parameters, energy of activation, latent heat of vaporization, molar volume of oil and free energy change of activation for viscous flow. @JASEM

The effect of strong electrolytes on the flowing parameters of sunflower oil has been reported earlier (Khan et al 2005). The viscosity of sunflower oil was found to be increased with increasing concentrations of the oil and decreased with rising of temperature. It is also observed that the viscosity of oil in 1,4 dioxane gets increased with increment in concentration of electrolyte. But on comparison with electrolytes, the viscosity obtained in the absence of electrolyte reveals that the addition of strong electrolyte reduces the flowing rates of oil. The results are also dealt with Jones-Dole coefficients (A and B). The positive values of B-coefficient get increased with the rise of temperature. This leads to the conclusion that ion-solvent interactions increase and electrolytes such as HCl, NaOH and NaCl behave as structure breaker. Maize is also an important source of edible oil, which is highly consumable oil used in our food preparations and behaves in same way as sunflower does. Palmitic acid, oleic acid and linoleic acid are important ingredients of maize oil. Higher iodine value and the presence of mono, di and triglycerides of unsaturated fatty acids have made the use of maize oil very significant. The theory that poly unsaturated fatty acids of oils are essential to human metabolism and they tend to combat the onset of the thromboses has made oil with high contents of these acids highly recommended as substituents for butter and in diet generally. Some of the better refined quality of maize oil reach a high degree of excellence as regards of tastes, smell and keeping properties of food unchanged find a use of baking. This paper deals with the effect of HCl, NaOH and NaCl on the viscosity of maize oil in 1, 4 dioxane.

EXPERIMENTAL

Material: Oil of maize was extracted from its dried and dehulled seeds and measured its density at different temperatures as shown in Table 1.

Table 1: Densities of maize oil at different temperatures

Temperature (K)	Density (g/cm^3)
298	0.960
303	0.958
308	0.950
313	0.949
318	0.942
323	0.937

The electrolytes like HCl, NaOH, NaCl, Oxalic acid of E.Merck and 1,4 dioxane (BDH) were used without further purification.

Procedure: All experiments were carried out according to the procedure given elsewhere (Khan et al 2005). The viscosity was measured at different temperatures ranging 298-323K with Ostwald viscometer type Techniconominal constant 0.1(Cs/S) capillary ASTMD 445. The average of the measurements of each solution was recorded to ensure the reproducibility of the observations.

RESULTS AND DISCUSSION

Viscosities of oil solutions in the absence and presence of strong electrolytes using 1,4 dioxane as solvent were measured at different temperatures ranging 298-323 K with the help of Ostwald type viscometer. Table 1 includes the viscosities of oil solutions (5-25% v/v) in 1,4 dioxane at different temperatures ranging 298-323 K with a difference of 5 K. These results show that viscosity of maize oil solution increases with increment of concentration

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whereas decreases with rise in temperature. A representative plot showing the effect of concentration of oil and temperature on viscosities of maize oil solution is shown in Fig.1. It is a general trend, which was also observed in the viscosities of sunflower solution 1,4 dioxane as solvent (Khan et al 2005). The results of the effect of different concentration of HCl ($1.5 x 10^{-3}$mol.dm^{-3}) on the viscosity of 5% maize oil solution in 1,4 dioxane at different temperatures were summarized in Table 1. These observations reveal that the viscosity of oil solutions increased with the increasing concentration of electrolyte. On comparison with the viscosities of oil solutions obtained in the absence of HCl show that addition of HCl reduces the viscosities of oil solution though not in high value. It means that the molecules of electrolyte increase the distance between oil molecules, which causes the enhancement of rate of flow. Furthermore viscosity of oil solution decreases with rise in temperature even in the presence of electrolyte. Similarly NaOH (Table 1) and NaCl (Table 1) behave in the same way. But it is noted that HCl is more efficient reducing electrolyte than NaOH and NaCl. It was very necessary to see the results in the light of the ingredients present in the oil. Maize oil is the mixture of both saturated and unsaturated in gradients. The saturated fatty acids are palmitic acid 8-10% [$CH_3(CH_2)_{14}COOH$] and stearic acid 2-4% [$CH_3(CH_2)_6COOH$] whereas unsaturated fatty acids are present as oleic acid 30-50% and linoleic acid 34-56%

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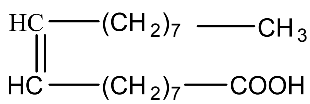
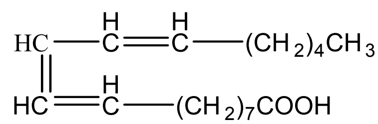


(Oleic acid)



(Linoleic acid)

Table 2: Viscosities of oil solutions in 1,4 –dioxane at different temperatures (K).

Concentration of oil (%)	Viscosity (cp) at different temperature (K)					
	298	303	308	313	318	323
5	1.335	1.206	1.147	1.079	1.001	0.920
10	1.570	1.372	1.300	1.243	1.131	1.046
15	1.774	1.581	1.514	1.420	1.274	1.200
20	2.119	1.882	1.742	1.656	1.473	1.367
25	2.460	2.162	2.046	1.903	1.718	1.598
[HCl]10^3 (mol.dm^{-3})	Viscosity of 5% maize oil solution in presence of electrolyte					
1.0	1.300	1.200	1.140	1.055	0.987	0.906
2.0	1.358	1.288	1.157	1.071	1.003	0.921
3.0	1.426	1.338	1.189	1.103	1.020	0.937
4.0	1.444	1.356	1.222	1.120	1.036	0.953
5.0	1.461	1.390	1.239	1.137	1.067	0.983
[NaOH]10^3 (mol.dm^{-3})						
1.0	1.305	1.200	1.141	1.072	0.952	0.863
2.0	1.401	1.279	1.158	1.099	0.968	0.889
3.0	1.418	1.312	1.175	1.115	0.904	0.904
4.0	1.436	1.329	1.207	1.148	1.031	0.920
5.0	1.470	1.364	1.254	1.164	1.042	0.950
[NaCl]10^3 (mol.dm^{-3})						
1.0	1.310	1.250	1.142	1.076	0.988	0.902
2.0	1.464	1.318	1.200	1.132	1.028	0.942
3.0	1.482	1.336	1.217	1.149	1.095	0.962
4.0	1.516	1.370	1.234	1.150	1.060	0.978
5.0	1.534	1.405	1.282	1.167	1.092	0.940

It also contains free acid as oleic acid 1-4% and traces of other acids are also present (Williams et al 1966). The increase in viscosity at various temperatures in the absence and presence of electrolyte is due to the unsaturation of ingredients present in the oil. Due to thermal effect probably radicals are formed which activate the ingredients of oil resulting the aggregation of the molecules of these acid and hence enhance the viscosity. It may be observed that continuos heating of oil results in the formation of sticky and viscous mass. It is certainly due to the polymerization of unsaturated species. The saturated fatty acids behave as chain transfer agent. All these reasons make the solution viscous. Beside this, the results are also observed in the light of change of pH values. The value of pH in 1,4 dioxane is obtained as 5.15-5.2 and for oil concentration 5% (v/v) in 1,4 dioxane, the pH reduced to 2.8-2.85.

Table 3: Ion- ion and Ion- solvent interactions of 5% maize oil with 1,4-dioxane in presence of strong electrolytes at different temperatures (K).

Temperature (K)	Ion-Ion and Ion-Solvent Interactions as A and B Coefficient of Jones-Dole equation		
	HCl	NaOH	NaCl
	A-Coefficient $(dm^3mol^{-1})^{1/2}$		
298	-2.828	-1.819	-1.688
303	-2.232	-2.846	-2.828
308	-2.762	-3.174	-3.146
313	-3.609	-3.008	-3.374
318	-3.385	-3.502	-3.748
323	-3.889	-4.491	-4.037
	B-Coefficient (dm^3mol)		
298	46.539	32.616	28.038
303	47.056	47.109	44.520
308	51.009	50.717	45.983
313	54.936	53.053	51.289
318	55.774	56.604	55.625
323	58.391	69.686	57.791

The change in pH reveals that the hydrogen ion concentration is increased by dissolving the oil in 1,4 dioxane (Khan et al 2005, Williams et al 1966, Adesun et al 1999). The similar results are also obtained by titrating the oil solutions versus standard NaOH. As more volume of NaOH consumed to titrate the oil solution as compared to 1,4 dioxane. The electrolyte reduces the viscosities and when a strong electrolyte of acidic nature (HCl) is added, it further decreases in concentration of monomers and decrease in intermolecular interaction. By adding strong electrolyte of alkaline nature such as NaOH, it is observed that with the increment in pH the viscosities are reduced as compared to the acidic medium as in alkaline medium, oil molecules form the sodium salt of stearic acid. As a result long chain association is decreasing due to intermolecular interaction (Shama 2004, Falken et al 1932, Jones et al 1929, Robson 1988, Saeed et al 2002). The same behavior was also observed in the viscosities of sunflower solution in 1,4 dioxane as solvent. The shape and structure of molecules are generally known to be more viscous than those with small and symmetrical molecules. Since only the hard symmetrical molecules have perfectly elastic collisions, the large and irregular molecules will have less elastic collision amongst themselves. As it is happening in the present case, Maize oil contains the ingredients of different molecular shape and molecular weight. Due to the formation of reactive species as a result of rising in temperature, more large chain of molecules is formed. Thus the collision between large molecules involves the loss of kinetic energy and as a consequence the intermolecular forces dominating the molecules tend to stick together which increases the viscosity of solutions. The intermolecular attractive forces do not permit a free flow of molecules in a liquid and strength of intermolecular forces will be measured the viscosity of a liquid. As we know the greater values of attractive molecular forces formed aggregated and bulky structure in solution resist the flow of liquid and increase the viscosity whereas the repulsive forces break this union of molecules in their simplest units and as a result decrease in viscosity takes place. The main aim of present study was to find out what happens when an electrolyte is added to dilute solution of maize oil using 1,4 dioxane as solvent. These two types of interactions take place in ion-ion interaction and ion-solvent interactions. These interactions may be explained in terms of Jones-Dole coefficients by using the following relation (Jones et al 1929):

$$\eta_{SP} = A(C)^{1/2} + B.C \quad (1)$$

Here η_{sp} is the specific viscosity, C is the concentration and A and B are Jones-Dole coefficients representing ion-ion and ion-solvent interactions, showing the behavior of electrolyte. B-coefficient provides information concerning the solvation of ions and their effects on the structure of the solvent (1,4 dioxane) in the vicinity of oil particles. On the other hand A-coefficient tells us about the complete or incomplete dissociation and ion association of electrolytes with maize oil in 1,4 dioxane systems (Salunkhe et al 1992).

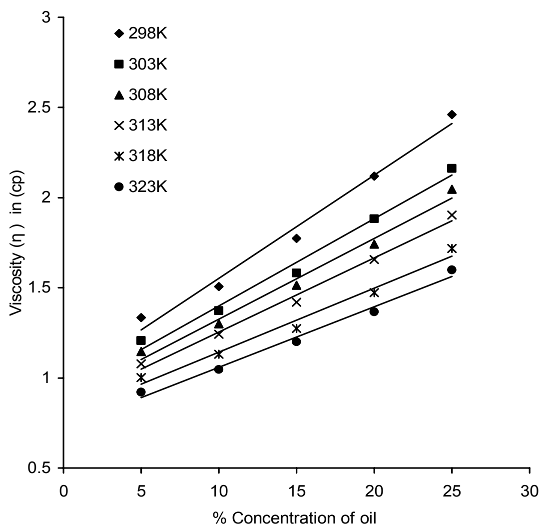


Fig 1 : Plot of viscosity (η) (cp) of Maize oil versus % concentration.

The value of A and B-coefficients of Jones-Dole equation may be determined graphically by plotting $\eta_{sp}/\sqrt{C}$ versus $\sqrt{C}$. The intercept and the slope of this plot will give respectively the values of A and B-coefficients, representative plot is shown in Fig. 2. The values of A and B-coefficients were determined graphically and shown in Table 2. These results indicate irregular variations in values of A-coefficients for various compositions of oil in 1,4 dioxane at various temperatures. In the concentration of HCl, the value of A-coefficient at 298 K was determined as –2.828 $(dm^3mol^{-1})^{1/2}$ whereas it drops to –2.232 $(dm^3mol^{-1})^{1/2}$ at 303 K. But it increases at 308 K as –2.762 $(dm^3mol^{-1})^{1/2}$. Similar observations were obtained in other electrolytes. This irregular behavior reveals an incomplete dissociation of ion association of electrolytes in 1,4 dioxane. The decrease in the values of A-coefficient with rise in temperature is due to the fact that A-coefficient depends upon the inflexible effect of the space lattice. The effect is greater at lower temperature. It is because the space lattice is less distributed by thermal variation. The increase in negative values may be probably due to the agitation at higher temperature (Robson 1988, Saeed et al 2002, Salunkhe at al 1992, Das et al 1997). Similar observations were also obtained in sunflower oil in 1,4 dioxane (Khan et al 2005).

Table 4: Fluidity parameters a, k, α_1, β_1, α_2, and β_2 of 5%(v/v) maize oil solution in 1,4-dioxane at different temperatures.

Temperature (K)	Fluidity Parameters					
	k	a	α_1	β_1	α_2	β_2
298	0.683	0.376	0.372	-0.122	0.439	-0.158
303	0.642	0.370	0.354	-0.135	0.404	-0.167
308	0.618	0.350	0.343	-0.140	0.392	-0.173
313	0.592	0.344	0.332	-0.146	0.379	-0.180
318	0.562	0.325	0.314	-0.157	0.360	-0.192
323	0.516	0.330	0.290	-0.155	0.333	-0.189

The B-coefficient of Jones-Dole equation explains the ion-solvent interactions. Temperature and solvent both affect the values of B-coefficient. The values are positive in all three electrolytes and increase with rise in temperature. The value of B-coefficient is determined as 46.539 (dm^3mol^{-1}) at 298 K whereas at 323 K, it becomes 58.39 (dm^3mol^{-1}). Similar behavior is also observed in NaOH and NaCl (Table 2). The increase of B-coefficient with rise of temperature reveals that electrolytes behave as structure breaker in 1,4 dioxane. The increase of B-coefficient temperature further shows that decrease in viscosity may be due to structure of 1,4 dioxane. Distortion of solvent structure is small which results the positive values of B-coefficient. Similar results are also obtained for the systems of sodium citrate solutions in acidic aqueous methanol (Berry et al 1980) and sunflower in 1,4 dioxane systems with strong electrolytes (Khan et al 2005). The effect of temperature was also determined in terms of six fluidity parameters, which are obtained from their respective plots using following relations of volume and viscosities (Adesun et al 1999).

$$\log\eta = \log k + a\log V_o \qquad (2)$$

$$\eta/V_o = \alpha_1 + \beta_1\eta \qquad (3)$$

$$1/V_o = \alpha_2/\eta + \beta_2 \qquad (4)$$

Here k, a, α_1, α_2, β_1 and β_2 are empirical constants, which are structural parameters. The constant 'k' and 'a' were determined respectively from the intercept and slope of the plot of log η versus log V_o. The values of α_1, β_1 and α_2, β_2 were obtained respectively from the plots of η / V_o versus η and $1/V_o$ versus 1/η. The values of these parameters were summarized in Table 3. These results show that the values of 'k' and 'a' decrease with rise of temperature. At 298 K the value of 'k' and 'a' were 0.683 and 0.376

respectively. But when temperature rises, they begin to decrease. The values of 'k' at temperatures 303,308,313,318 and 323 K, are obtained as 0.642, 0.618, 0.592, 0.562 and 0.516 respectively. Similarly the values of 'a' were obtained as 0.370, 0.350, 0.344, 0.325 and 0.330 with the difference of 5 K. The results clearly reveal that 'k' and 'a' are structural parameters. Similarly 'α_1' also decrease with increase in temperature. The values of 'α_1' were obtained as 0.372, 0.354, 0.343, 0.332, 0.314 and 0.290 at temperatures 303,308,313,318 and 323 K respectively, whereas 'β_1' values negatively increase with rise in temperature. At 298 K, it was obtained as –0.122 whereas at 323 K, the value increased to –0.155. Both 'α_1' and 'β_1' are temperature dependent. In the same way the value of 'α_2' decreased (0.439, 0.404, 0.392, 0.379, 0.360 and 0.333) with rise in temperature with a difference of 5 K. On the other hand the values of 'β_2' show negatively increase. These values further ensure that 'α_2' and 'β_2' are structural parameters. In brief the fluidity parameters as discussed above are related to structure

Table 5: Energy of activation (ΔE_v), Latent heat of vaporization (ΔL_v) and Molar volume (V_m) of maize oil (5% v/v) in 1,4-dioxane.

Electrolyte	Energy of Activation	Latent heat of vaporization	Molar volume of oil
$[HCl]10^3$ ($mol.dm^{-3}$)	ΔE_v ($kJ.mol^{-1}$)	ΔL_v ($kJ.mol^{-1}$)	V_m ($10\ e^8$) (dl)
1.0	12.54	31.35	4.76
2.0	12.62	31.56	4.54
3.0	13.58	32.42	6.61
4.0	12.96	33.95	6.14
5.0	13.43	33.59	5.01
$[NaOH]10^3$			
1.0	13.75	34.39	10.58
2.0	13.80	34.51	8.96
3.0	14.08	35.21	8.13
4.0	14.28	35.76	7.02
5.0	14.67	36.67	6.97
$[NaCl]10^3$			
1.0	13.36	33.41	6.75
2.0	13.48	33.70	6.29
3.0	13.62	34.07	5.92
4.0	13.65	34.13	6.66
5.0	13.70	34.26	6.37
Oil concentration % (v/v)		Without Electrolyte	
5	11.13	27.83	2.74
10	11.19	27.99	2.35
15	12.10	30.25	2.97
20	13.24	33.12	4.43
25	13.52	33.82	3.42

The energy of activation (ΔE_v), latent heat of vaporization (ΔL_v) and molar volume of oil (V_m) were determined graphically. The values of energy of activation (ΔE_v) and molar volume of oil (V_m) were determined from the slope and intercept of the linear plot of log η versus 1/T respectively. A representative plot is shown in Fig. 3. The value of slope was used in determining the energy of activation (ΔE_v) by the following relations (Berry et al 1980, Atkins 1990):

$$\Delta E_V = Slope \times R \times 2.303 \qquad (5)$$

whereas the Latent heat of vaporization (L_v) was determined from the expression:

$$\Delta E_V = 0.4 L_V \qquad (6)$$

Similarly the molar volume of oil (V_m) was determined from the following relation:

$$\log V_m = \left[\log hN_A - C\right] \qquad (7)$$

where h, N_A and C respectively Planck's constant, Avogadros number and intercept of plot of log η versus 1/T respectively. Table 4 includes the values of energy parameters of maize oil in 1,4 dioxane without using any electrolyte. The values of energy of activation (ΔE_v) and latent heat of vaporization (ΔL_v) get increased with increase in oil concentration whereas change in molar volume of oil (V_m) is irregular with concentration. The numbers of molecules of oil are greater at higher concentration. The aggregation of molecules produces hindrance in the mobility of molecules. This makes difficult to provide vacant sites in the solvent matrix resulting high energy of activation (ΔE_v) and latent heat of vaporization (ΔL_v). The molar volumes of oil (V_m) generally decrease with increase in concentration of oil. Table 4 includes the observations obtained in the presence of electrolytes such as HCl, NaOH and NaCl. These results show that electrolytes influence the mobility of oil solutions. The concentrations of electrolyte generally increase the activation energy (ΔE_v) and latent heat of vaporization (ΔL_v). Increase or decrease of molar volume (V_m) is not regular with the concentration of electrolyte. Increase in molar volume of oil (V_m) in presence of electrolyte produce less vacant sites in the solvent and resulting an increase in activation energy as the concentration of electrolytes increases from $1.0x10^{-3}$ ($mol.dm^{-3}$) to $5.0x10^{-3}$ ($mol.dm^{-3}$).

Table 6: Free energy change of activation (ΔG*) ($kJ.mol^{-1}$) for maize oil solutions in 1,4-dioxane at various temperatures (K).

Electrolyte $[HCl]10^3$ ($mol.dm^{-3}$)	Free energy change of activation (ΔG*) ($kJ.mol^{-1}$) of 5% (v/v) maize oil solution in presence of strong electrolytes in 1,4-dioxane at various temperatures (K).					
	298	303	308	313	318	323
1.0	86.71	88.03	89.21	90.45	91.73	92.94
2.0	85.10	86.40	87.55	88.77	90.02	91.20
3.0	84.25	85.50	86.61	87.82	89.01	90.19
4.0	83.58	84.82	85.92	87.12	88.31	89.47
5.0	83.06	84.33	85.43	86.59	87.81	88.97
$[NaCl]10^3$						
1.0	86.64	87.87	89.06	90.37	91.48	62.65
2.0	85.10	86.30	87.47	88.76	89.89	91.02
3.0	84.18	85.40	86.52	87.79	88.91	90.04
4.0	83.52	84.73	85.88	87.15	85.25	89.34
5.0	83.04	84.25	85.31	86.62	87.72	88.84
$[NaOH]10^3$						
1.0	86.85	88.04	89.25	90.55	91.74	92.96
2.0	85.26	86.43	87.62	88.89	90.05	91.25
3.0	84.32	85.48	86.65	87.91	89.06	90.20
4.0	83.69	84.83	85.97	87.18	88.36	89.53
5.0	83.17	84.35	85.50	86.65	87.85	88.98
Oil Concentration (% v/v)	Without electrolyte					
5	65.34	66.18	67.16	68.10	68.99	69.86
10	65.63	66.50	67.48	68.47	69.34	70.21
15	66.04	66.86	67.86	68.81	69.62	70.57
20	66.47	67.29	68.22	69.22	70.00	70.92
25	66.84	67.64	68.63	69.57	70.41	71.33

The free energy change of activation (ΔG*) for viscous flow was also evaluated by the relation (Nightangle et al 1959):

$$\Delta G^* = RT \ln(\eta V / hN_A)$$
(8)

Here V is the volume of one mole of solution particles calculated by the following expression:

$$V = 1000/(n_1 + \nu n_2)$$
(9)

where 'ν' is the number of species into which the solute molecules dissociated and 'n_2' is the number of moles of solute per litre of solution. The number of moles of solvent 'n_1' per litre of solution is given by:

$$n_1 = (1000 d_o - n_2 M_2)/M_1$$
(10)

Here 'M_1' and 'M_2' are the molecular weight of the solvent and solute respectively, 'd_o' is the density of solvent. The results of free energy change of activation (ΔG*) are summarized in Table 5.

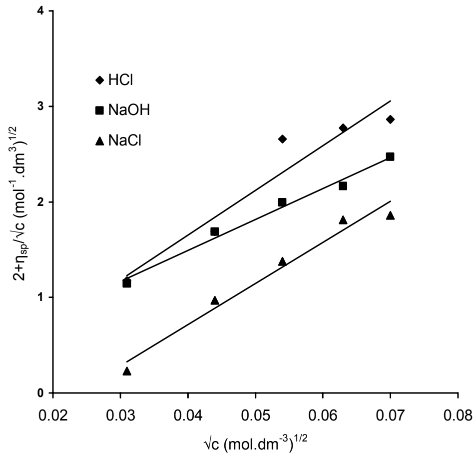


Fig 2 : Plot of $\eta_{sp}/\sqrt{c}$ versus $\sqrt{c}$ of Maize oil in electrolytes at 298K

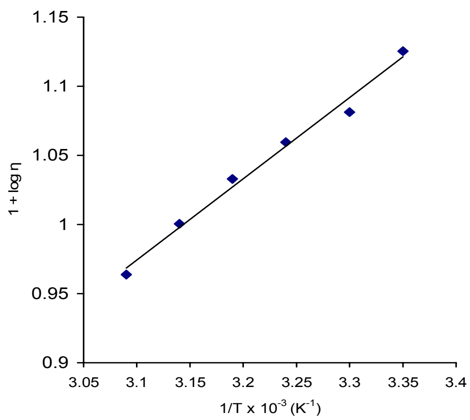


Fig 3: Plot of log η against 1/ T (K^{-1}).

It shows that the values of free energy change of activation (ΔG*) increase with the increase in concentration of maize oil at fix temperature. Further more rising of temperature enhance the values of free energy change of activation (ΔG*) which show that the association of oil molecules with solvent (1,4-dioxane) increases. At high temperature the intermolecular forces weaken between the oil molecules and this reduce molecular cohesion which would led to decrease in viscosity (Adesun et al 1999) and increase in free energy change of activation (ΔG*). The experiments are also accomplished in the presence of strong electrolytes such as HCl, NaOH and NaCl, which are summarized in Table 5. On comparing the results in the absence (Table 5) and presence of electrolyte (Table 5), the addition of electrolyte increases the value of free energy change of activation (ΔG*). In the presence of electrolyte the values of free energy change of activation (ΔG*) were increased with rising of temperature whereas increasing concentration of electrolyte, decreases the values of free energy change of activation (ΔG*). The values of free energy change of activation (ΔG*) control the rate of flow during the fluidity process. Actually the flow process was governed by the ability of molecules to move into the prepared holes and the readiness with which the holes were produced in the liquid. With the increasing concentration of electrolytes at fix concentrations of oil (Table 5) and temperatures, the values of free energy change of activation (ΔG*) are decreased. Increase in size of molecule makes the movement of molecule slow. Furthermore the high values of free energy change of activation (ΔG*) with the rise of temperature indicate that ion-solvent interaction increase with the rise in temperature.

In brief the unsaturation of fatty acid present in maize oil causes the changes of various parameters determined in the presence and absence of strong electrolytes. The behavior of electrolytes on the viscosity of maize oil solution was dealt with Jones Dole equation in terms of ion-ion and ion-solvent interactions and concluded that electrolytes like HCl, NaOH and NaCl behave as structure breaker.

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