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Rheological behavior of gum cordia, effect of temperature and concentration over its shear rate profile and non-Newtonian behavior

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Abstract— Gum cordia is extracted from the fruits of flowering plant Cordia myxa. The gum has been reported to be used as controlled drug delivery agent, edible films and coatings. The gum contains an anionic polysaccharide having good adhering property. In addition it contains ample foaming ability and great capacity to hold both oil and water. Due to these properties it wields massive industrial applications in chemical and food industries as a binding agent. Even after having vast industrial applications, the rheological behavior of this important gum is still unexplored. This study revealed the rheological behavior, flow parameters and apparent viscosity of gum cordia as a function of temperature (25-75ºC), concentration (10-40 kg/m^3) and shear rate ($0.5s^{-1}$ to $50s^{-1}$). Flow behavior of gum cordia was found to be best explained by power law model as compared to Newtonian & Bingham models. Mathematical models were developed to predict the values of consistency index (K), flow behavior index (n) and apparent viscosity as a function of concentration and temperature. Increase in concentration of gum from 10 to 40kg/m^3 resulted in increase in K from 0.237 to 13.6 and decrease in n from 0.499 to 0.332.

Index Terms— Bingham model, Fluid Mechanics, Food ingredient, Gum cordia, Newtonian model, Polysaccharide, Rheology.

1 Introduction

Gum Cordia is a natural anionic polysaccharide obtained from the fruits of Cordia myxa [1].The fruit is grown in the Indo-Pak subcontinent, where ripe fruit is consumed as such and unripe fruit is harvested for making pickles [1],[2]. Polysaccharides are long chain biopolymers with ability to dissolve in water to make a stabilized viscous hydrocolloid. Among plant polysaccharides, gum cordia have gained much interest due to ease of extraction and is used frequently in many industries. They are employed to develop or control the texture of food products because of their ability to retard flow. Other main characteristic of gum cordia includes gel modification, emulsions preservation and suspensions stability. They are also extensively used in chemical, pharmaceutical and food industries. They can influence the absorption and works as an excellent additive due to their low toxicity, biodegradable, availability and low cost. Nutraceutical properties like antioxidant and anti-inflammatory activity have also been reported in many parts of the tree [3]. In traditional medicine, it is used for treating chronic fever and spleen disease.

The mucilage of the fruit contains a gum which is used as an adhesive material for cardboard and paper [2],[4].The gum has been proposed to be used in sustained delivery of the drug [3],[4] and has been successfully utilized as edible coatings. Our group has previously reported its effectiveness, as edible coating, for increasing the shelf life of peanuts [5],[6] and pine nuts [2],[7]. Recently, fabrication and characterization of standalone edible film from gum cordia has also been reported by our group [8]. The knowledge of the rheological properties of the food matrix is indispensable so as to have the successful operation of processing equipment e.g. evaporators and heat exchangers [9] and to have the desired texture of the final product [10].

Among many factors, the rheological properties are affected by type of polysaccharide, temperature and concentration. Recently, the rheological properties of the gum from leaf of Cordia myxa were reported [11].

2 Materials and methods

Extraction of gum cordia and its solution preparation is described elsewhere [2].Rheological properties were measured at controlled temperatures using a rotational viscometer (RVT model; Brookfield Engineering Labs. Inc., Stoughton, MA, USA), as described by Maskan [11]. Different spindles at various rotational speeds (rpm) were used to apply varying shear rates. The equipment manufacturer guidelines were followed regarding sample's presentation to the apparatus (Brookfield digital viscometer manual). Since the instrument did not give direct readout in terms of shear rate, the experimental data were converted into shear stress and shear rate by Mitschka method [12] . Flow data were obtained over the concentrations 10-40kg/m^3 maintained at temperature of 25, 35, 45, 55, 65 and 75ºC. All samples were tested in triplicate.

2.1 Data analysis

The rheological behavior of the gum solution was modeled by using power law, Bingham and Newtonian models.

Power law model: $\sigma = K(\gamma)^n$ (1)

Bingham model: $\sigma = \mu_0(\gamma)+\tau_0$ (2)

Newtonian model: $\sigma = \mu(\gamma)$ (3)

Where σ is shear stress (mPa), γ is shear rate (s^{-1}), K ($Pa.s^n$) and n are the parameters of power law model, μ_0 (mPa. s) and τ_0 (Pa) are the parameters of Bingham model and μ (mPa. s) is the Newtonian model parameter. The model parameters were determined by nonlinear regression using SPSS® software (Statistical Package for the Social Sciences).

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3 Result and Discussion

3.1 Rheological behavior of gum solution

Food rheology is the function of the characteristic component present in food [13]. The fluid food with dissolved low molecular weight compounds exhibits the Newtonian behavior. For Newtonian fluid, the viscosity remains constant as a function of shear rate i.e. shear rate is directly proportional to shear stress. However, for non- Newtonian fluids, the viscosity changes as a function of shear rate. The dissolved polymer, even at low concentration, substantially alters the flow characteristic of fluid food from Newtonian to non- Newtonian. The literature suggests that the most of the food exhibits non-Newtonian and shear-thinning behavior, i.e. the viscosity decreases with increasing shear rate [13]. The power law and Bingham models are commonly used to characterize the shear-thinning and shear-thickening behavior. The consistency index (K) and flow behavior index (n) are the parameters used to characterize the power law model [14]. For Newtonian fluids (n=1), the consistency index (K) corresponds to the viscosity of the fluid. The value of n<1 signifies the shear-thinning behavior while value of n>1implies the shear-thickening behavior. The constants of Newtonian, power law and Bingham models as a function of temperature and concentration are reported in Table 1,2 and 3 respectively

Table 1

C	*T*	Newtonian Model	
kgm^{-3}	°C	μ (mPa. s)	R^2
10	25	0.429	0.996
	35	0.443	0.991
	45	0.459	0.991
	55	0.471	0.99
	65	0.488	0.989
	75	0.499	0.989
15	25	0.4	0.991
	35	0.423	0.989
	45	0.441	0.997
	55	0.455	0.991
	65	0.465	0.991
	75	0.487	0.982
20	25	0.374	0.987
	35	0.393	0.996
	45	0.415	0.992
	55	0.44	0.99
	65	0.459	0.998
	75	0.473	0.938
25	25	0.357	0.916
	35	0.379	0.939
	45	0.413	0.946
	55	0.433	0.938
	65	0.459	0.945
	75	0.474	0.969
30	25	0.341	0.936
	35	0.369	0.966
	45	0.393	0.979
	55	0.421	0.966
	65	0.452	0.978
	75	0.471	0.975
35	25	0.341	0.973
	35	0.369	0.965
	45	0.396	0.976
	55	0.427	0.959
	65	0.441	0.949
	75	0.462	0.947
40	25	0.331	0.942
	35	0.358	0.945
	45	0.388	0.996
	55	0.42	0.946
	65	0.437	0.949
	75	0.448	0.959

Table 2

C	*T*	Power Law Model			
kgm^{-3}	°C	*n*	*K* (Pa. s^n)	n_{ave}	R^2
10	25	0.437	2.151	0.467	0.998
	35	0.45	1.213		0.992
	45	0.46	0.69		0.996
	55	0.471	0.305		0.992
	65	0.488	0.229		0.995
	75	0.499	0.237		0.997
15	25	0.405	3.656	0.445	0.998
	35	0.413	2.387		0.998
	45	0.439	1.49		0.998
	55	0.458	0.914		0.993
	65	0.469	0.601		0.999
	75	0.486	0.351		0.986
20	25	0.367	5.691	0.424	0.998
	35	0.384	3.787		0.996
	45	0.414	2.508		0.999
	55	0.44	1.581		0.997
	65	0.463	1.095		0.998
	75	0.478	0.657		0.954
25	25	0.356	7.378	0.419	0.996
	35	0.382	5.102		0.999
	45	0.41	3.659		0.997
	55	0.431	2.437		0.999
	65	0.461	1.749		0.997
	75	0.474	1.088		0.998
30	25	0.34	9.295	0.408	0.996
	35	0.373	6.561		0.997
	45	0.4	4.65		0.999
	55	0.424	3.388		0.997
	65	0.452	2.31		0.997
	75	0.463	1.791		0.996
35	25	0.344	11.279	0.405	0.994
	35	0.372	8.261		0.996
	45	0.394	6.178		0.997
	55	0.422	4.509		0.999
	65	0.441	3.267		0.997
	75	0.462	2.397		0.998
40	25	0.332	13.6	0.398	0.994
	35	0.359	10.18		0.995

45	0.391	7.548	0.996
55	0.422	5.629	0.997
65	0.437	4.223	0.998
75	0.448	3.76	0.999

Table 3

C	T	Bingham Model		
kgm^{-3}	°C	μ_0 (mPa. s)	τ_0 (Pa)	R^2
10	25	0.158	2.833	0.879
	35	0.133`	1.351	0.928
	45	0.076	0.915	0.953
	55	0.026	0.623	0.95
	65	0.017	0.699	0.973
	75	0.018	0.668	0.974
15	25	0.217	4.832	0.861
	35	0.222	2.546	0.902
	45	0.109	1.982	0.896
	55	0.071	1.412	0.921
	65	0.048	1.066	0.941
	75	0.038	0.761	0.962
20	25	0.266	7.234	0.834
	35	0.199	4.801	0.839
	45	0.156	3.213	0.856
	55	0.125	2.056	0.898
	65	0.088	1.612	0.91
	75	0.068	1.193	0.928
25	25	0.724	7.07	0.895
	35	0.249	6.533	0.847
	45	0.333	3.837	0.89
	55	0.172	3.151	0.865
	65	0.147	2.215	0.89
	75	0.115	1.769	0.931
30	25	0.541	10.388	0.866
	35	0.457	7.354	0.874
	45	0.256	6.303	0.872
	55	0.228	4.327	0.884
	65	0.186	2.805	0.901
	75	0.152	2.349	0.89
35	25	0.865	11.597	0.89
	35	0.589	9.133	0.883
	45	0.344	7.88	0.858
	55	0.302	5.649	0.879
	65	0.264	4.143	0.886
	75	0.224	3.057	0.896
40	25	1.136	13.078	0.844
	35	0.858	10.334	0.891
	45	0.607	8.254	0.894
	55	0.375	7.187	0.877
	65	0.417	5.028	0.934
	75	0.495	2.957	0.93

The rheological behavior of the gum cordia was found to fit best into the power law model based on its high coefficient of determination (R2).Power law model generally explains the behavior of most of the food hydrocolloids [15]. The gum exhibited the shear-thinning behavior at all concentrations and temperatures as the value of n is less than 1. Fig. 1 shows the representative rheogram for the gum solution at 20kg/m³.The rheograms plotted for other concentrations also exhibited the similar pattern (data not shown).

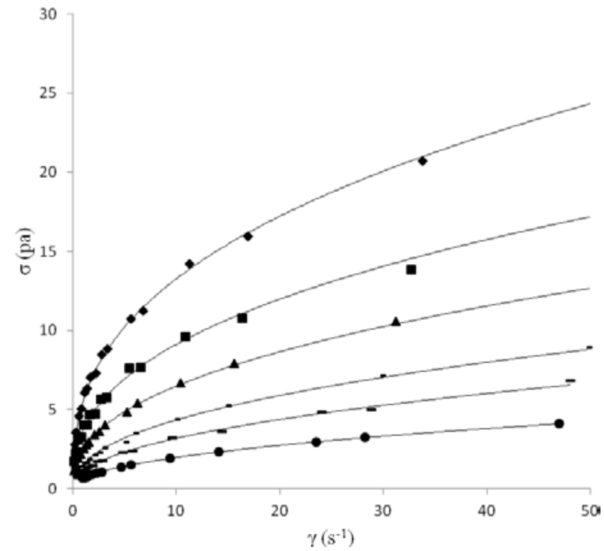

Figure 1 Rheogram of Gum Cordia solution (20kgm^{-3}) at different temperature (°C)
(♦25 ■35 ▲45 -55 −65 ●75)

3.2 Effect of concentration and temperature on flow parameters

The consistency index (K) increased with concentration. It showed the inverse relation with temperature (Table 1,2 & 3). On the contrary, the n showed the opposite trend. Increase in consistency index (K) was found to be more profound at higher concentrations (Fig. 2).

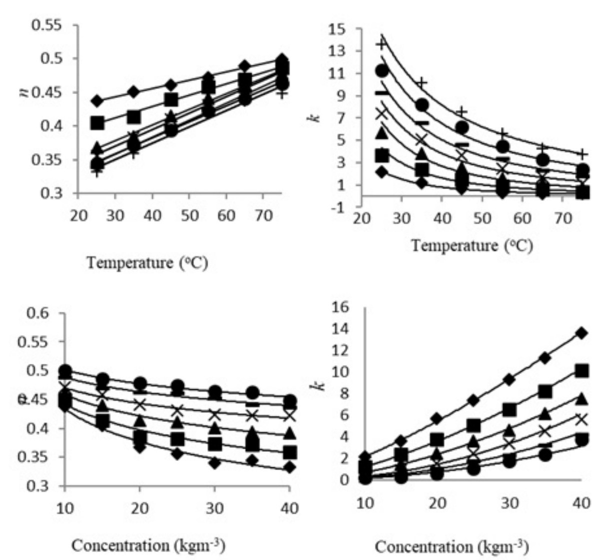

Figure 2 Variation of *K* and *n* with concentration (kgm^{-3}) (♦10 ■15 ▲20 ×25 −30 ●35 +40) and Temperature °C(♦25 ■35 ▲45 ×55 −65 ●75).

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This could be attributed to increase in particle-particle interaction [14, 16]. Overall, the values of K were found to be varied from 0.237 to 13.6. The extent of shear-thinning behavior is assessed by flow behavior index (n) [17]. Smaller value of n indicates greater deviation from Newtonian behavior. The value was found to be dependent on temperature and concentration, ranging from 0.332 to 0.499. At a given temperature, flow behavior index (n) and concentration were found to be inversely related. Thus, non-Newtonian behavior was distinct at higher concentration. At all concentrations, non-Newtonian behavior amplified with temperature (Fig. 2).Similar behavior has been reported for xanthan gum [18], carboxyl methyl cellulose (CMC) [14] , pectin, starch and carrageenan [19]. Among common hydrocolloids - xanthan gum, starch, carrageenan and pectin- gum cordia exhibited the second highest shear-thinning behavior after xanthan gum. For hydrocolloids, the consistency index (K) increases with concentration. At a given temperature, gum cordia exhibited intermediate consistency index (K) among carrageenan, xanthan gum, starch and pectin. Presence of solids increases the viscosity of solution, mainly due to molecular movement and interfacial film formation [17]. To analyze the effect of concentration on shear-thinning behavior, the average of flow behavior index was calculated (nave). It showed the power-type relationship with concentration (Fig. 3). Turian approach was adopted to study the effect of temperature on K and n [16].

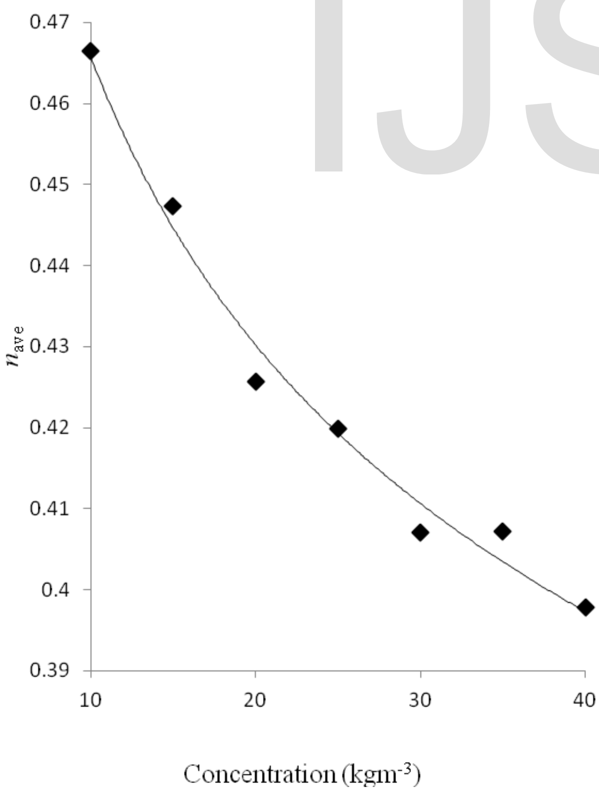

Figure 3 Relationship between average flow behavior index (n_{ave}) and concentration (kgm^{-3})

Turian approach was adopted to study the effect of temperature on K and n [16].

$\log K = \log K_o - a_1 T$ (4)

$n = n_o + a_2 T$ (5)

The concentration of the solution has significant effect on the flow parameters of the solution. The variation of K and n with concentration is given by power law and exponential relationships.

$K = K_1 C^{K_2}$ (6)

$n = n_1 C^{n_2}$ (7)

The exponential relationship is given by the following equation;

$K = K_3 \exp(K_4 C)$ (8)

$n = n_3 \exp(n_4 C)$ (9)

The variation of $\log K_o$ and a_1, in equation (4), with concentration can be related by the following equations;

$\log K_o = [(\log K_o)_1]\, C^{(\log K_o)_2}$ (10)

$a_1 = a_{11} C^{a_{12}}$ (11)

$\log K_o = (\log K_o)_3 \exp[(\log K_o)_3$ (12)

$a_1 = a_{13} \exp(a_{14} C)$ (13)

The effect of temperature and concentration on flow parameters can be combined into single equation for example, for the processes where simultaneous heat and mass transfer takes place. The following equations are obtained by using the equation (4), (10)-(13).

$\log K = (\log K_o)_1 C^{(\log K_o)_2} - \alpha_{11} C^{a_{12}} T$ (14)

$\log K = (\log K_o)_1 C^{(\log K_o)_2} - a_{11} \exp(a_{12} C) T$ (15)

$\log K = (\log K_o)_3 \exp[(\log K_o)_4 C] - a_{11} C^{a_{12}} T$ (16)

$\log K = (\log K_o)_3 \exp[(\log K_o)_4 C] - a_{11} \exp(a_{12} C) T$ (17)

Variation of flow index n_o and a_2, in equation (5), with respect to concentration is given by the following equations [16].

$n = aC^b$ (18)

$\beta_2 = \beta_{21}(C^{\beta_{22}})$ (19)

$n = a[\exp(b^*C)]$ (20)

$\beta_2 = \beta_{21} \exp(\beta_{22}^*C)$ (21)

The following equations are obtained by combining equation (5), (18)-(21).

$n = aC^b + \beta_{21} C^{\beta_{22}} T$ (22)

$n = aC^b + \beta_{21} \exp(\beta 22^*C) T$ (23)

$n = a\exp(b^*C) + \beta_{21} C^{\beta_{22}} T$ (24)

$n = a\exp(b^*C) + \beta_{21} \exp(\beta_{22}^*C) T$ (25)

(log Ko)1, (log Ko)2, (log Ko)3, (log Ko)4, α11, α12 , a, b, β21,β22are the constants in the equation.

The values of the constants, obtained by multiple regression analysis, are reported in Table 4.

Table 4

Numeric values of theoretical .model's parameters using equations 14-17 and 22-25

Eqns	Constant	Values	Eqns	Cnsts	Values
14	$(\log K_o)_1$	0.374	22	A	0.717
	$(\log K_o)_2$	0.372		B	-0.264
	α_{11}	0.056		β_{21}	0.001
	α_{12}	-0.396		β_{22}	0.358
	R^2	0.991		R^2	0.98
15	$(\log K_o)_1$	0.459	23	A	0.634
	$(\log K_o)_2$	0.307		B	-0.223

	α_{11}	0.028		β_{21}	-0.002
	α_{12}	-0.022		β_{22}	0.01
	R^2	0.986		R^2	0.972
16	$(\log K_o)_3$	0.892	24	a	0.461
	$(\log K_o)_4$	0.011		b	-0.015
	α_{11}	0.082		β_{21}	0
	α_{12}	-0.523		β_{22}	0.583
	R^2	0.987		R^2	0.96
17	$(\log K_o)_3$	0.999	25	a	0.441
	$(\log K_o)_4$	0.007		b	-0.013
	α_{11}	0.034		β_{21}	0.001
	α_{12}	-0.032		β_{22}	0.019
	R^2	0.979		R^2	0.97

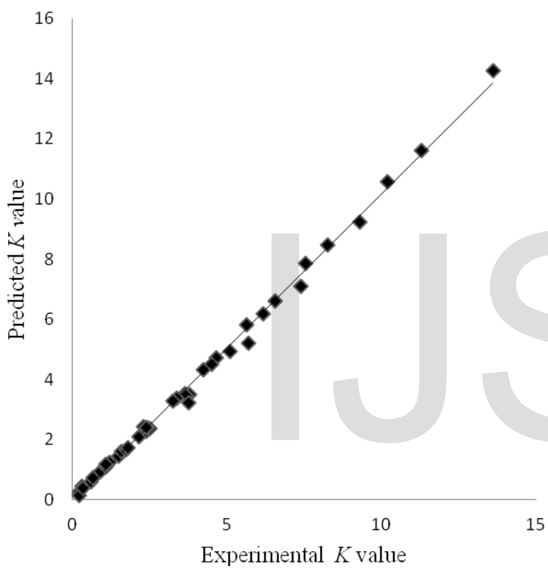

Figure 4 Comparison of experimental values of K with the predicted values, obtained using equation 14

Equations (14) and (22) were found to be the best fit as evident by their coefficient of determinations. Fig. 4 and 5 were obtained by the experimental values and the predicted values using equation (14) and equation (22) for K and n, respectively. The figures show the overall uniform distribution of values with the R2 being 0.991 and 0.980 for equation (14) and equation (22), respectively.

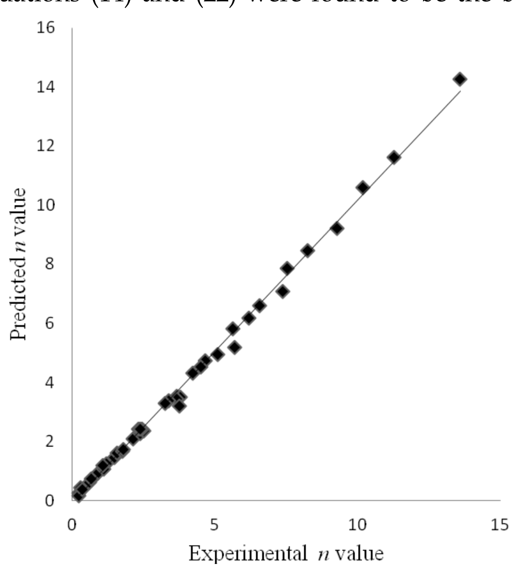

Figure 5 Comparison of experimental values of n with the predicted values obtained using equation 22

3.3 Effect of temperature and concentration on apparent viscosity

The viscosity of the fluid is due to the resistance in the motion of fluid layers, which depends on their inter-molecular forces of attraction, primarily governed by temperature and the concentration of molecules [14]. Since the viscosity of non-Newtonian fluids varies by shear rate, the term apparent viscosity is used. The apparent viscosity of gum cordia was analyzed, at specific shear rates, as a function of temperature and concentration. Increase in shear rate was found to be associated with decrease in viscosity at all temperatures and concentrations (Fig. 6). Decrease in viscosity was found to be more

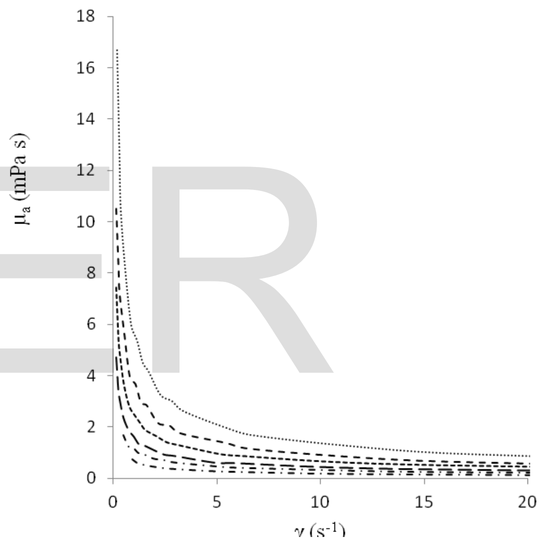

Figure 6 Apparent viscosity of gum cordia solution (20kgm^{-3}) at different temperature (°C)
(25 35 45 55 65 75)

profound at lower shear rates (Fig. 6). This could be attributed to the breakdown of the entangled agglomerates [16, 20, 21] at high shear rates, allowing intermolecular forces to contribute less resistance to flow. On the other hand, apparent viscosity was found to be decreased at elevated temperature. The effect was observed at all concentrations (Fig. 6 & 7).

This could be the result of decreased interactions between the polymer chains due to gain in the kinetic energy of molecules, which increases the distance between them [14]. At elevated temperature, the rise in dehydration of the polymer increases its flexibility, leading to more free motion [16]. The apparent viscosity of xanthan gum, carrageenan, starch, pectin [19], CMC, and guar gum also showed the similar behavior as a function of shear rate and temperature [17]. The Arrhenius equation is used to model the change in viscosity with temperature [16].

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μa= μoexp(Ea/RT) (26)

Where μais the apparent viscosity (mPa.s);μo, the viscosity at infinite temperature (mPa.s); Eais the activation energy(Jmol-1); R, the molar gas constant (Jmol-1K-1);T is the temperature (K).

The change in viscosity with concentration is given by the power-law and exponential relationship. This relation is also meaningful for viscosity at infinite temperature (μo).

μo=ω(C)λ (27)

μo= ω1exp(λ1C) (28)

Where ω, ω1, λ, λ1are the constants.

By combining equation (26)-(28), the single equation, showing the effect of concentration and temperature, is obtained.

μa = ω(C)λexp(Ea/RT) (29)

μa = ω1exp(λ1C)exp(Ea/RT) (30)

These equations were used to determine the apparent viscosity at given temperature and concentration. Table 5 and 6 shows the values of the constants of equations 29-30.

Table 5.

Constant values for equation 29

shear rate (s-1)	ω [mPa. s(kgm-3)-λ]	λ	E_a($Jmol^{-1}$) (Jmol-1)	R^2
0.5	0.000003	1.451	25287.387	0.9508
1	0.000001	1.4616	26244.03	0.9912
2	0.000004	1.4148	23433.507	0.9713
3	0.000002	1.4133	23945.441	0.9854
4	0.000003	1.3993	22663.556	0.9807
5	0.000003	1.3885	22862.95	0.9789
6	0.000003	1.3902	22084.435	0.9827
7	0.000003	1.3708	22141.343	0.9714
10	0.000004	1.3504	21365.773	0.9589
12	0.000002	1.3881	21953.073	0.9845
20	0.000005	1.3032	19810.722	0.9085
30	0.000002	1.3728	21024.516	0.9699
40	0.000008	1.2398	18146.296	0.7893
50	0.000003	1.3419	19203.27	0.9777

Table 6.

Constant values for equation 30

shear rate (s-1)	ω_1 (mPa. s)	λ_1 (kgm-3)- 1	E_a (Jmol-1)	R^2
0.5	0.00009	0.0541	25315.981	0.9438
1	0.000043	0.05381	26208.205	0.9792
2	0.000083	0.05273	23451.076	0.9624
3	0.000057	0.05233	23880.91	0.9749
4	0.000075	0.0521	22673.923	0.9705
5	0.000065	0.05156	22774.223	0.9695
6	0.000074	0.05176	22093.716	0.9723
7	0.000072	0.05101	22031.095	0.9632
10	0.00008	0.05038	21226.5	0.9522
12	0.000053	0.05141	21946.52	0.9715
20	0.000104	0.04892	19590.67	0.9065
30	0.000045	0.05074	21010	0.9556
40	0.000144	0.04696	17800.247	0.7948
50	0.000066	0.04997	19205.982	0.9654

Results revealed that there was increase in activation energy for shear rates 0.5s-1 to 1s-1. This was followed by fall in activation energy with rise in shear rate up to 50s-1. The activation energy indicates the sensitivity of apparent viscosity to temperature change [20].The decline in activation energy at higher shear rates implies that the apparent viscosity was more sensitive to temperature at lower shear rate. This effect is more prevalent in CMC as compared to gum cordial [14]. Equation (29) correlates well the effect of temperature and concentration on the apparent viscosity. Fig. 8 shows the pooled data of apparent viscosity by using equation (29) and (30).The experimental apparent viscosity and predicted apparent viscosity were found to be in agreement i.e. R2>0.9.

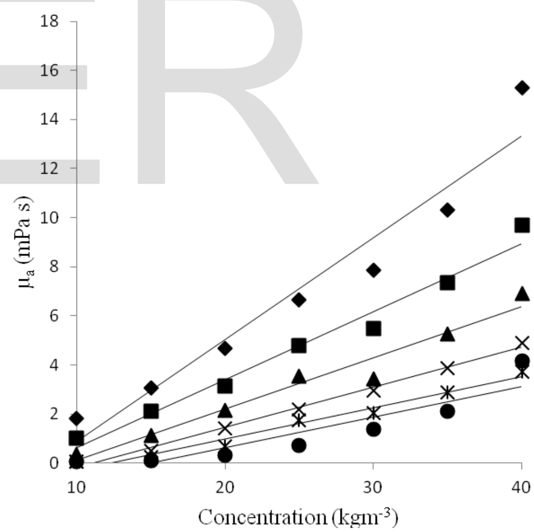

Figure 7 Apparent viscosity as function of Concentration (kgm-3) and Temperature (°C)

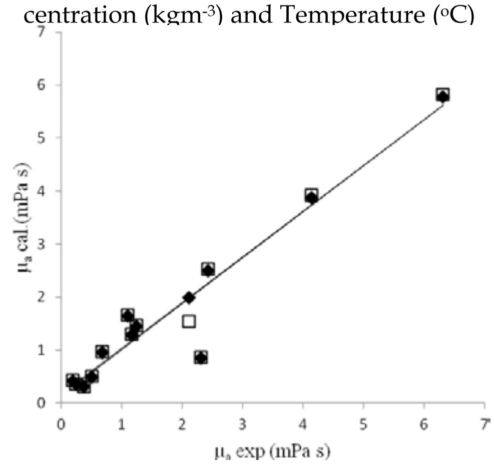

Figure 8 Comparison of predicted with experimental apparent viscosity for equation 29 and 30 (◆29 □30)

4 Conclusion

Gum cordia revealed non-Newtonian flow predominantly at high concentrations and low temperatures. Power law model explains flow characteristics of gum cordia. Flow behavior index (n) and consistency index (K) can be tailored by varying concentrations and temperatures of gum cordia solutions. The apparent viscosity of gum cordia has shown temperature susceptibility particularly at low shear rates. These findings would be helpful in designing mass and heat transfer systems for different industrial applications of gum cordia.

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