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A Comparison of Rheological Models and Experimental Data of Metallocene Linear Low Density Polyethylene Solutions as a Function of Temperature and Concentration

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ABSTRACT

Knowledge of rheological properties of polymer and their variation with temperature and concentration have been globally important for processing and fabrication of polymers in order to make useful products. Basheer et al. [1] investigated, experimentally, the changes in rheological properties of metallocene linear low density polyethylene (mLLDPE) solutions by using a rotational rheometer model AR-G2 with parallel plate geometry. Their work covered the temperature range from $293\,K$ to $323\,K$ and concentration from 1000 ppm to $4500\,ppm$. In this paper, we reconsider Basheer work to describe the rheological behavior of mLLDPE solutions and its dependence on concentration and temperature.

Until now, several models have been built to describe the complex behavior of polymer fluids with varying degrees of success. In this article, Oldroyd 4-constant, Giesekus and Power law models were tested for investigating the viscosity of mLLDPE solutions as a function of shear rate. Results showed that Giesekus and power law models provide the best prediction of viscosity for a wide range of shear rates at constant temperature and concentration. Therefore, Giesekus and power law models were suitable for all mLLDPE solutions while Oldroyd 4-constant model doesn't.

A new proposed correlation for the viscosity of mLLDPE solutions as a function of shear rate, temperature and concentration has been suggested. The effect of temperature and concentration can be adequately described by an Arrhenius-type and exponential function respectively. The proposed correlation form was found to fit the experimental data adequately.

Indexing terms/Keywords

Rheology, Apparent viscosity, Viscoelastic models, Metallocene linear low density polyethylene.

Academic Discipline And Sub-Disciplines

Physics, Fluid Mechanics

SUBJECT CLASSIFICATION

Rheology of Polymers

TYPE (METHOD/APPROACH)

Theoretical Study.

INTRODUCTION

Knowledge of rheological properties of polymeric fluids and their variation with temperature and concentration have been globally important industrialization of polymer technology for quality [2] correlation with sensory evaluation, designing of transport system, equipment design (heat exchanger and evaporator), deciding pump capacity and power requirement for mixing [3-6]. In industrial operations a product is submitted to a range of shear rates and it is important to know how the viscosity will change with temperature at these shear rates to adequately design the equipment for these operations [7-9].

For many simple fluids the study of rheology involves measurements of viscosity. For such fluids the viscosity depends primarily upon the temperature and concentration. However, the rheology of polymers is much more complex because polymeric fluids show non-ideal and complex shear viscosity behavior [10, 11]. Most polymer melts and solutions are non-Newtonian, their viscosity depend on factors other than the velocity gradients. Except for rare cases, the viscosity of polymer melts and solutions decreases as the shear rate increases. This could be explained by the alignment of the polymer molecules under the application of the shear. The molecular alignment will allow easier flow of the molecules, which reduces the viscosity at higher shear rates.

One of the important researches on the rheology of polymer solutions was conducted by Diego et al. [12]. The power law model was employed to correlate experimental data of shear stress. They found that the model describes the experimental data adequately. They also found that all of the polymer solutions are generally non-Newtonian pseudo plastic fluids as the apparent viscosity decreases with increasing shear rate, therefore, they exhibit a shear thinning behavior.

There are many models dealing with the rheological characterization of polymer solutions and melts. Most of them are in the form of shear stress versus shear rate. On the contrary a few models describe the effects of temperature and concentration on fluid properties, despite their importance in many engineering design problems [13-15].

Basheer et al. [1] investigated experimentally the changes in rheological properties of mLLDPE solutions by using a rotational rheometer model AR-G2 with parallel plate geometry over shear rates $3.583 \le \dot{\gamma} \le 545\ \mathrm{s}^{-1}$. Their work covered the temperature range from $293\,\mathrm{K}$ to $323\,\mathrm{K}$ and concentration range from 1000 part per million (ppm) to $4500\,\mathrm{ppm}$. Cyclohexane was chosen as a solvent to prepare the dilute solutions of mLLDPE based on the scientific references. In this paper, we reconsider Basheer's work with the following objectives:

- Collect experimental data and carry out its analysis based on three simple viscoelastic fluid models; namely, Oldroyd 4-constant, Giesekus and Power law models.
- Investigate theoretically the effects of temperature and concentration on the rheological properties of mLLDPE solutions.
- Picked up the best rheological model which describes the experimental results.

FLOW MODELS

Newtonian Model

Modeling in fluid mechanics and rheology is based on conservative laws of mass, momentum and energy.

$$\nabla \cdot \underline{V} = 0\,, \tag{1}$$

$$\rho \frac{D\underline{V}}{Dt} = -\nabla p + \nabla \cdot \underline{\underline{\tau}}\,, \tag{2}$$

$$\rho C_p \frac{DT}{Dt} = k_p \nabla^2 T\,, \tag{3}$$

where $\underline{V}$, ρ, ∇p, $\underline{\underline{\tau}}$, C_p, T and k_p are respectively, the velocity field, the density, the pressure gradient, the extra stress tensor, the specific heat capacity, temperature and the thermal conductivity. In equations 2 and 3 the substantial time derivative, $\frac{D}{Dt}$, is the time derivative in a frame that translates with the material particle. It is related to the time derivative, $\frac{\partial}{\partial t}$, in a fixed frame by:

$$\frac{D}{Dt} = \frac{\partial}{\partial t} + \underline{V} \cdot \nabla\,, \tag{4}$$

to close the system of equations we require an equation relating the extra stress tensor $\underline{\underline{\tau}}$ to the kinematic variables. The simplest constitutive model for incompressible viscous fluids is based on the assumption that the extra stress tensor is proportional to the symmetric part of the velocity gradient,

$$\underline{\underline{\tau}} = 2\eta_o\, \underline{\underline{d}}\,, \tag{5}$$

where η_o is a (constant) viscosity and $\underline{\underline{d}} = \frac{1}{2}\left(\underline{\underline{L}} + \underline{\underline{L}}^T\right)$ is the rate of deformation tensor with $\underline{\underline{L}} = \nabla \underline{V}$. Substituting of equation 5 in the momentum equation, equation 2, leads to the well known Navier-Stokes system. As already discussed, this set of equations is commonly used to describe Newtonian fluids but polymers exhibits relevant non-Newtonian characteristics and more complex constitutive models should be used.

Non-Newtonian Model

Materials generate stress effects that are not represented by the Newtonian constitutive equation, equation 5, are called non-Newtonian fluids. For these fluids, the Newtonian constitutive equation does not make accurate predictions. There are many representations for viscoelastic models [16, 17]. The most common models are Oldroyd 4-constant model, Giesekus model and Power law model.

Oldroyd 4-Constant Model

This model represents a special case from Oldroyd 8-constant model. The differential form of this model can be written as:

$$\underline{\underline{\tau}} + 2\lambda_1 \overset{\nabla}{\underline{\underline{\tau}}} + \lambda_5 (\mathrm{tr}\, \underline{\underline{\tau}})\, \underline{\underline{d}} = 2\eta_o \left[\underline{\underline{d}} + \lambda_2 \overset{\nabla}{\underline{\underline{d}}} \right]. \tag{6}$$

In this model, the corresponding apparent viscosity, $\eta(\dot{\gamma})$, as a function of shear rate, $\dot{\gamma}$, is given as:

$$\eta(\dot{\gamma}) = \eta_o \frac{1 + \lambda_2 \lambda_5 \, \dot{\gamma}^2}{1 + \lambda_1 \lambda_5 \, \dot{\gamma}^2} \quad , \tag{7}$$

Giesekus Model

Giesekus proposed a constitutive model based on a concept of configuration-dependent molecular mobility. In this model, the viscoelastic components of the extra stress tensor, $\underline{\underline{\tau}}$, is represented with the following parameters η_p, λ_1, λ_2 and α [18, 19, 20].

$$\underline{\underline{\tau}}_p + \lambda_1 \overset{\nabla}{\underline{\underline{\tau}}}_p + \alpha \frac{\lambda_1}{\eta_p} (\underline{\underline{\tau}}_p \cdot \underline{\underline{\tau}}_p) = 2\eta_p \, \underline{\underline{d}}. \tag{8}$$

The parameter α is the dimensionless Giesekus-model mobility factor which controls the extensional viscosity and the ratio of second normal stress difference to the first one. For $\alpha = 0$, the model becomes isotropic upper convected Maxwell model, while for $\alpha = 1$ the model is isotropic drag, and for $\alpha > 0$ the model represents shear thinning behavior.

The inclusion of ($\underline{\underline{\tau}}_p \cdot \underline{\underline{\tau}}_p$) term in equation 8 gives a viscosity function that are much more realistic than any other model. The apparent viscosity in this model is given by:

$$\eta(\dot{\gamma}) = \eta_o \left\{ \frac{\lambda_2}{\lambda_1} + \left(1 - \frac{\lambda_2}{\lambda_1}\right) \frac{(1-f)^2}{1 + (1-2\alpha)\, f} \right\}, \tag{9}$$

where

$$f = \frac{1-\chi}{1 + (1-2\alpha)\,\chi}, \tag{10}$$

and

$$\chi^2 = \frac{\sqrt{1 + 16\alpha (1-\alpha)(\lambda_1 \dot{\gamma})^2} - 1}{8\alpha (1-\alpha)(\lambda_1 \dot{\gamma})^2}. \tag{11}$$

Power Law Model

Several non-Newtonian models have been used to describe the behavior of the polymeric fluids. The simplest of these is the power-law model

$$\underline{\underline{\tau}} = m \dot{\gamma}^n, \tag{12}$$

where m and n are the fluid consistency index and the flow behavior respectively. A number of proposed empirical equations describing the effect of shear rate on fluid viscosity may be found in Bouldin et al. [21]. When $n = 1$, equation 12 reduces to the Newtonian fluid, and m becomes the viscosity of the fluid. At other values of n, the apparent viscosity, defined as:

$$\eta = m \dot{\gamma}^{n-1}, \tag{13}$$

is a function of shear rate. Taking logarithms of both sides of equation 12:

$$\log \underline{\underline{\tau}} = n \log \dot{\gamma} + \log m, \tag{14}$$

the parameters m and n are determined from a plot of $\log \underline{\underline{\tau}}$ versus $\log \dot{\gamma}$, and the resulting straight line's intercept is $\log m$ and the slope is n.

EFFECTS OF TEMPERATURE AND CONCENTRATION

The temperature of the polymer solution has a profound effect on its viscosity. Most well known expression describing the effect of temperature on the viscosity was proposed by Eyring [22] as:

$$\eta = k\,\mathrm{Exp}\left(\frac{E}{T}\right), \tag{15}$$

where k is constant and $E = \Delta H / R$, ΔH the "flow activation energy", R is Boltzmann gas constant, T is the temperature in Kelvin. Amin and Maddox [23] have reviewed most common correlation used for the prediction of liquid viscosity as function of temperature.

Al-Zahrani alone [24] and with collaboration with Al-Fariss [25] studied the rheological behavior of some dilute aqueous polymer solutions. The modified correlation proposed in his study has been found to fit the experimental data adequately for crude oils as well as the polymer tested. The proposed relationship is:

$$\eta(\dot{\gamma}) = a\,\mathrm{Exp}(bC)\,\dot{\gamma}^{c}\,\mathrm{Exp}\left(\frac{d}{T}\right), \tag{16}$$

where a, b, c and d are constants and C is the polymer concentration in ppm. They proposed this correlation to predict the viscosity of polymer solution as a function of temperatures, shear rate and concentrations.

MATERIALS AND EXPERIMENTAL WORK

The materials and preparation of mLLDPE solutions experimental procedure, were employed by Basheer et al. [1]. The rheological measurements were carried out using a rotational rheometer model AR-G2 with parallel plate geometry ($40\,\mathrm{mm}$ diameter) to measure the viscosity versus shear rate of polymer solutions. The rheometer is equipped with a plate temperature unit that gives a very good temperature control over an extended time.

Basheer used four different concentrations which are ranged from $1000\,\mathrm{ppm}$ to $4500\,\mathrm{ppm}$ (with $500\,\mathrm{ppm}$ increments). The temperature were varied from $293\,\mathrm{K}$ to $323\,\mathrm{K}$ (with $10\,\mathrm{K}$ increments). In all experimental range, the shear rate was varied randomly between $3.5\,\mathrm{s}^{-1}$ and $545\,\mathrm{s}^{-1}$. In this paper, we reconsider Basheer data to predict, theoretically, the dependence of the apparent viscosity $\eta(\dot{\gamma})$ on the concentration and temperature of the mLLDPE solutions.

RESULTS AND DISCUSSION

Apparent Viscosity

The rheological behavior of the mLLDPE solutions at concentrations of 4000 and 4500 ppm and with different temperatures (293, 303, 313 and 323 K), is represented in figures 1 and 2 respectively. It is observed that the apparent viscosity decreased with increasing shear rates for mLLDPE solutions that can be explained by the structural breakdown of the blend due to the hydrodynamic forces generated and the increased alignment of the constituent molecules. In figures 1 and 2, it is also observed that apparent viscosity decreases at a faster rate for lower temperature than at high temperature which suggested the shear thinning nature of mLLDPE solutions.

For large values of shear rates, the apparent viscosity is expected to decrease until they reach a constant value, which compatible with Newtonian viscosity. When shear force was applied, the molecules would under go rearrangement among themselves in a direction parallel to that shear force. This would results the molecules in the fluid to flow easily due to the reduced molecule-molecule interaction and less restriction in flow. Thus the viscosity of the fluid decreases. These results are in agreement with similar studies in solution of different polymers [7, 16, 17].

Figures 3, 4 show the effect of mLLDPE concentration on the viscosity at temperatures of 293 and 303 K respectively. It can be clearly shown that the viscosity increases with increasing concentrations. This rise in the viscosity is due to the increasing in the intermolecular interactions of the mLLDPE molecules. The given figures clearly indicate that the rate of decrease in the low shear region is much greater than in the intermediate shear region. At a relatively high shear rates, the dependency on concentration is negligible, whereas at a very low shear rate there is a very strong concentration effect on viscosity.

Comparison Between Computational Results and Experimental Data

Comparing the experimental data with the computational results based on Oldroyd 4-constant, Giesekus and Power law models will be achieved. Parameters for the apparent viscosity, $\eta(\dot{\gamma})$, for the three models are determined by fitting calculated values with experimental data. The results are shown in figures 5 to 10 and values of parameters for each model are listed in tables 1, 2 and 3; respectively. These figures illustrate the fitting of the three models to the apparent viscosity as a function of shear rate and at different values of temperature and concentration. The points represent experimental data, and the solid lines represent the prediction for the different models. From the figures we can see that, as the temperature increases the viscosity decreases. The viscosity at high shear rates simulate a linear relationship (the shear rate has less effect on viscosity, Newtonian fluid) while non-Newtonian behavior is shown at low shear rates.

The variation of viscosity with shear rate for different concentrations are illustrated in figures. 6, 8 and 10. Again viscosity varies exponentially with shear rate and sensitivity is maximum at lower levels of shear rate. For the same shear rate, it is seen that viscosity increases as the concentration increase too.

As can be seen, using the fitting parameters given in tables 1, 2 and 3 the Giesekus model found accurately predicts viscosity as a function of shear rate. Therefore, Giesekus model can be used to extrapolate the viscosity data for different shear rates. The success of this model is attributed to the deformation rate dependence of its viscosity and time constants. For the power law model, the correlation parameter given in Basheer et al. work [1] are incorrect. The corrected data are given in table 3.

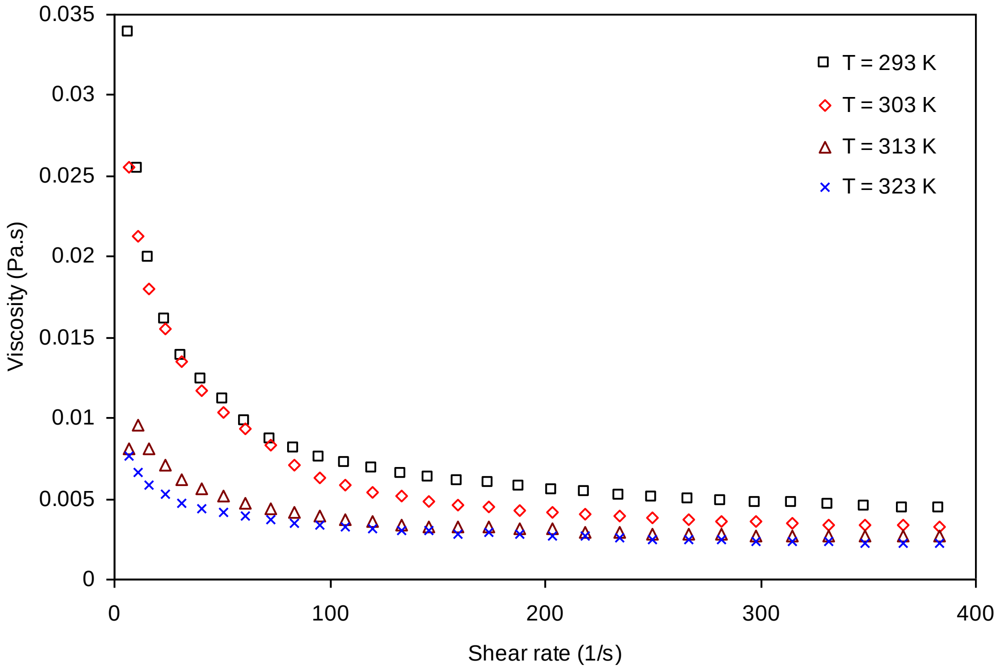

Fig 1: Effect of temperature on the rheological behavior of mLLDPE solutions at $C = 4000\,\text{ppm}$.

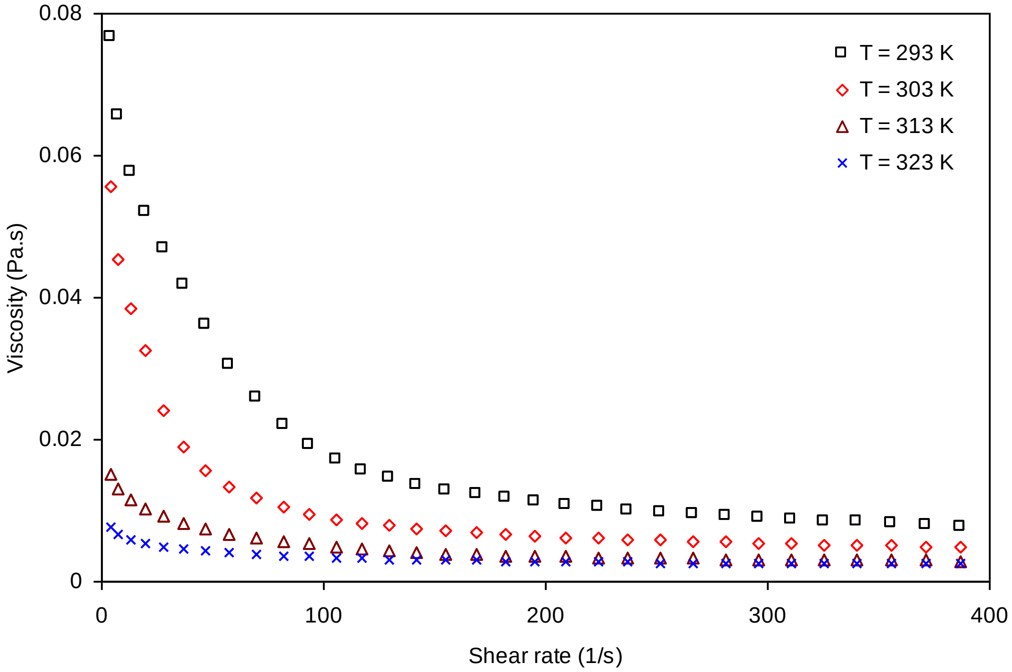

Fig 2: Effect of temperature on the rheological behavior of mLLDPE solutions at $C = 4500\,\text{ppm}$.

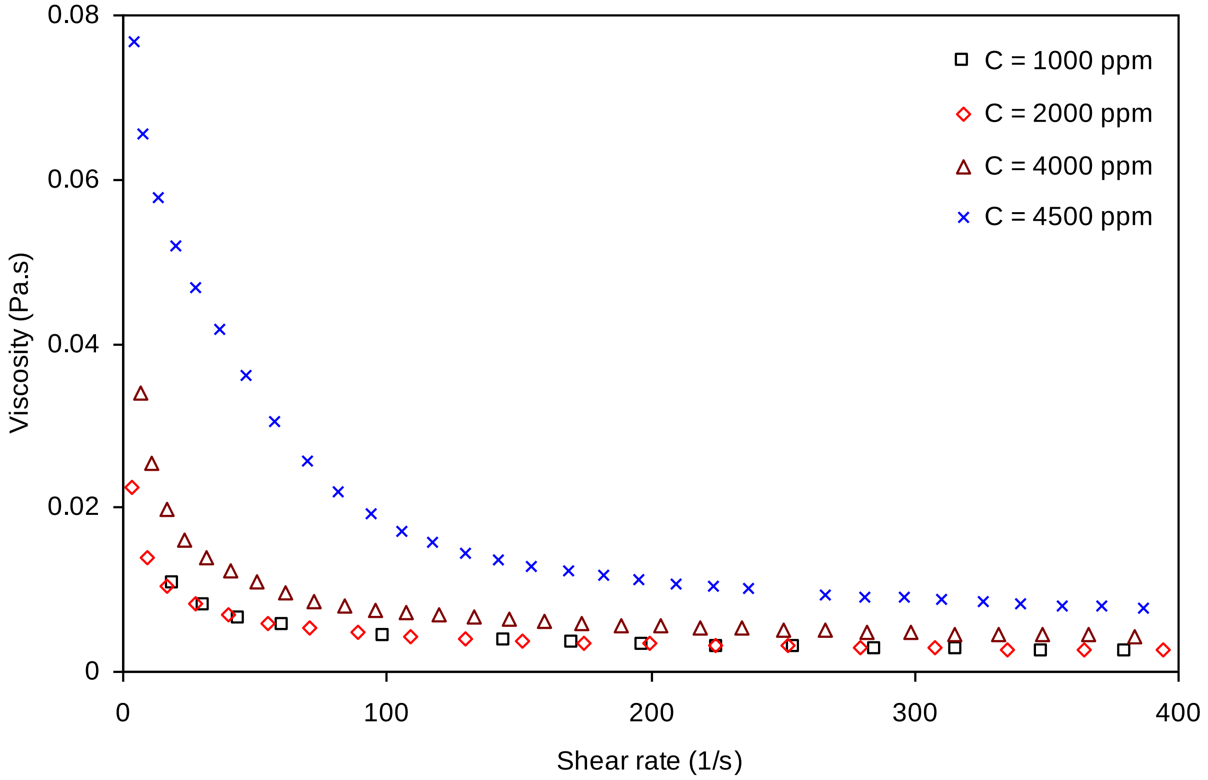

Fig 3: Effect of concentration on the rheological behavior of mLLDPE solutions at $T = 293K$.

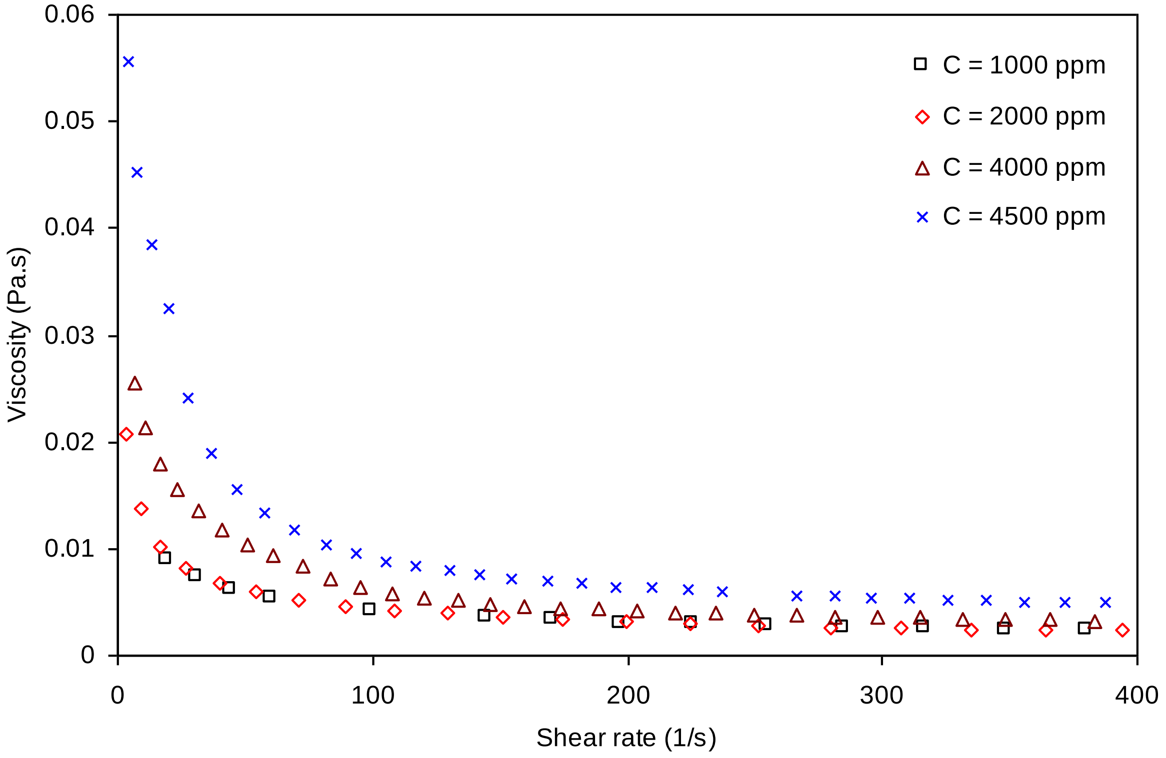

Fig 4: Effect of concentration on the rheological behavior of mLLDPE solutions at $T = 303K$.

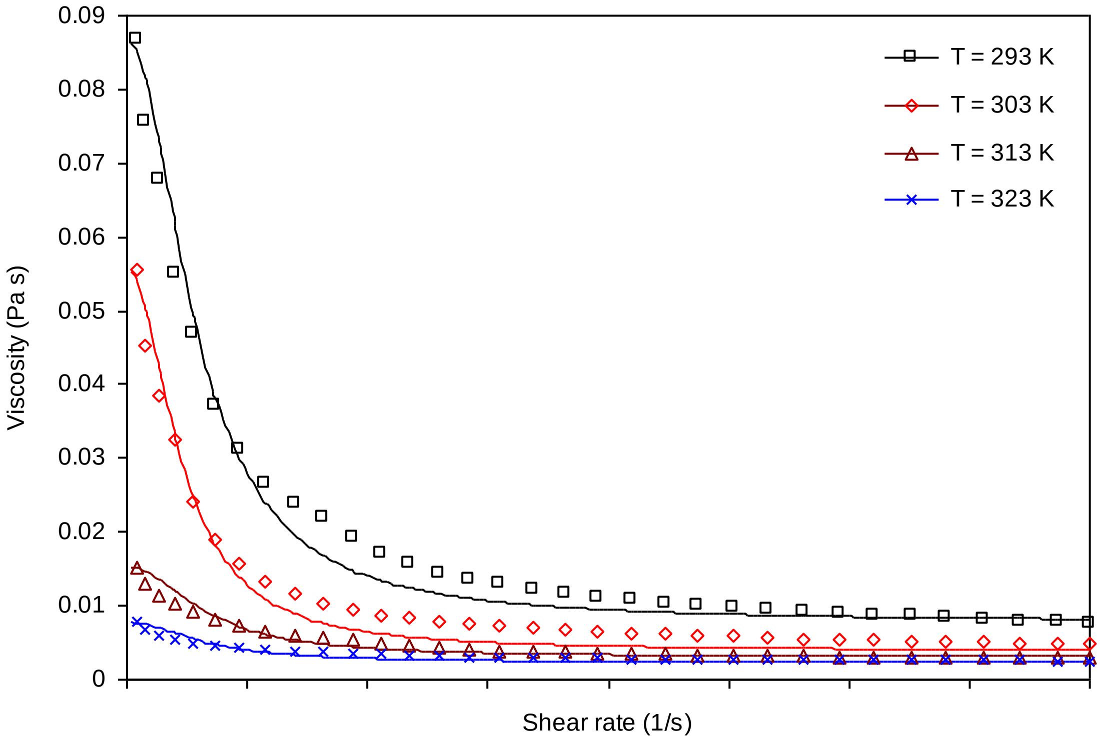

Fig 5: Viscosity-shear rate curves at $C = 4500\,\text{ppm}$ **where dots represent the experimental data and solid lines represent Oldroyd 4-constant model fit.**

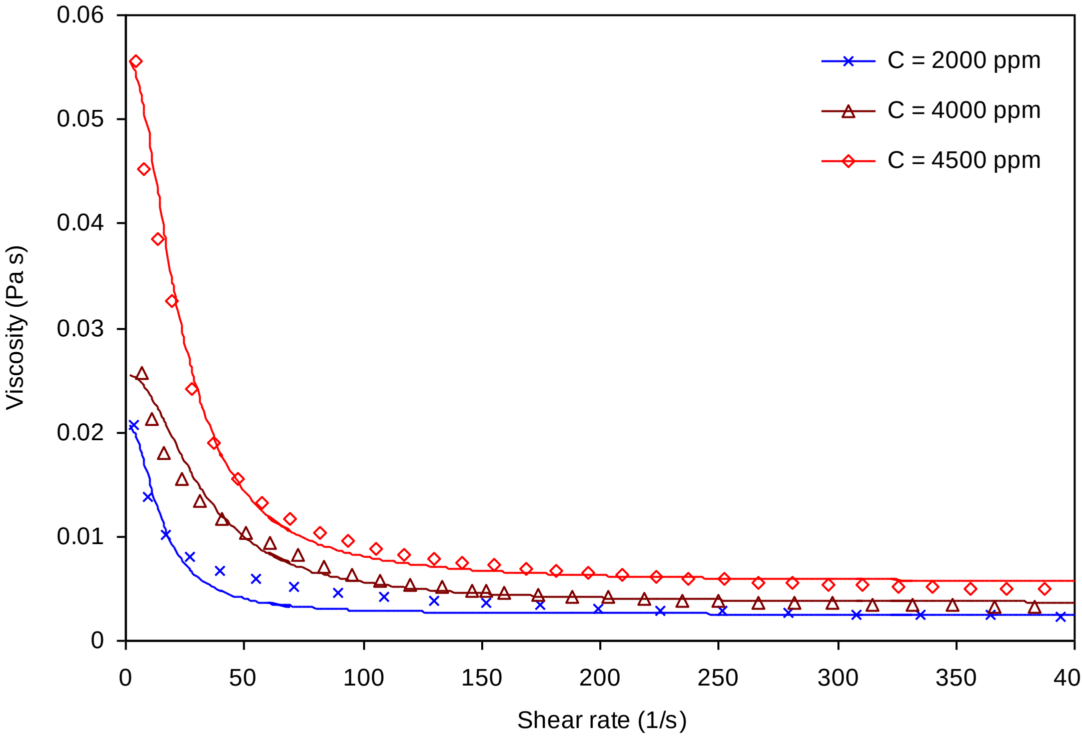

Fig 6: Viscosity-shear rate curves at $T = 303\,\text{K}$ **where dots represent the experimental data and solid lines represent Oldroyd 4-constant model fit.**

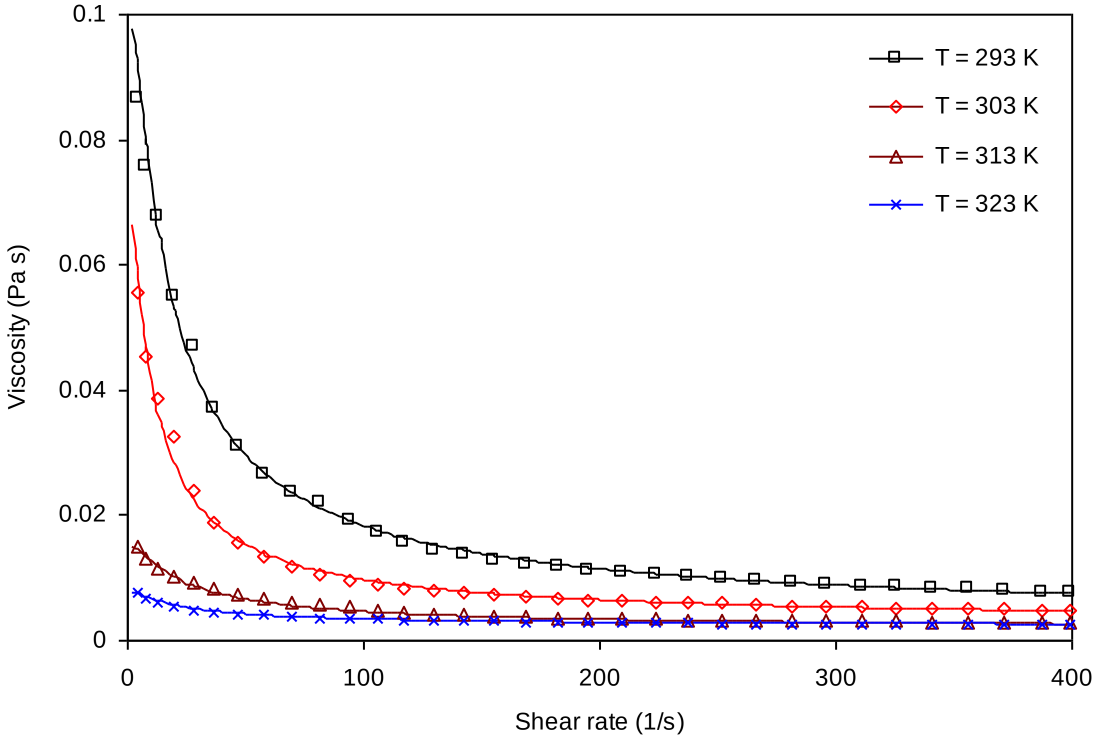

Fig 7: Viscosity-shear rate curves at $C = 4500\,\text{ppm}$ **where dots represent the experimental data and solid lines represent Giesekus model fit.**

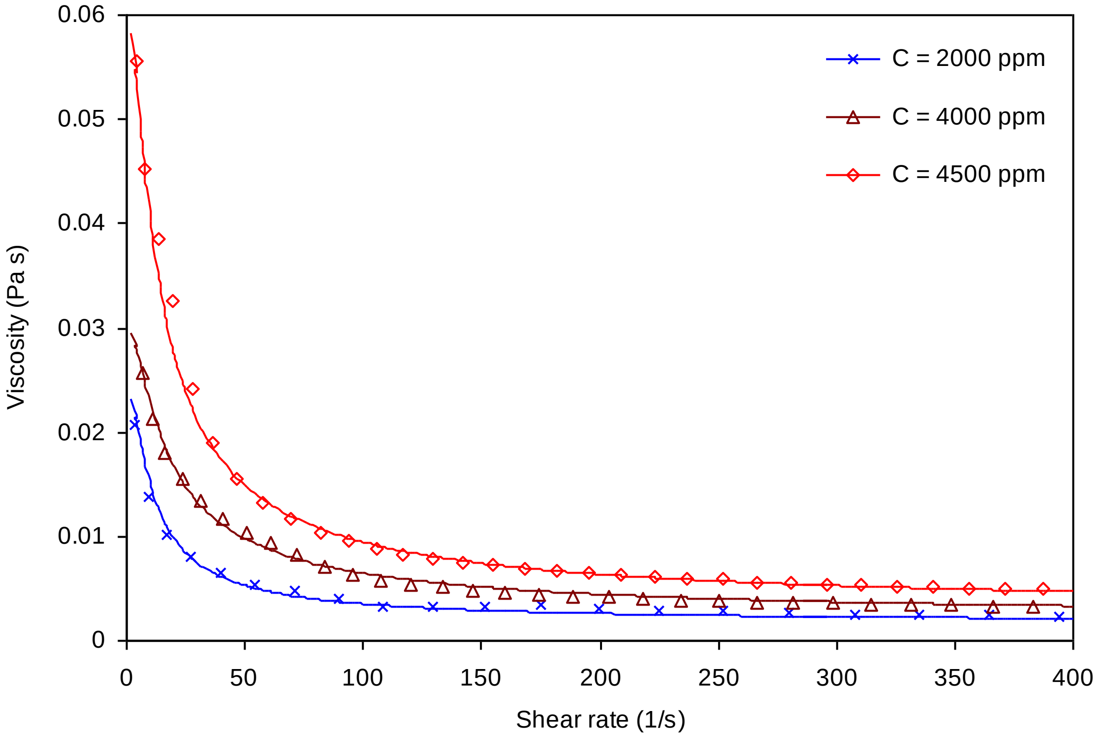

Fig 8: Viscosity-shear rate curves at $T = 303\,\text{K}$ **where dots represent the experimental data and solid lines represent Giesekus model fit.**

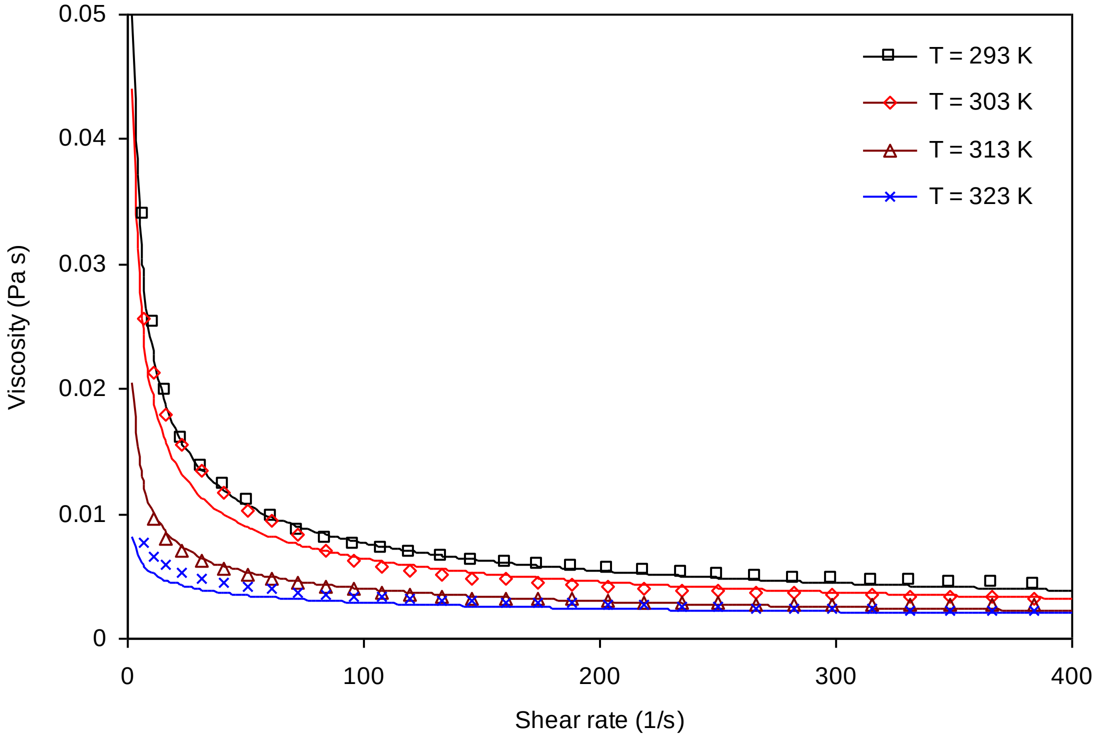

Fig 9: Viscosity-shear rate curves at $C = 4000\,\text{ppm}$ **where dots represent the experimental data and solid lines represent Power law model fit.**

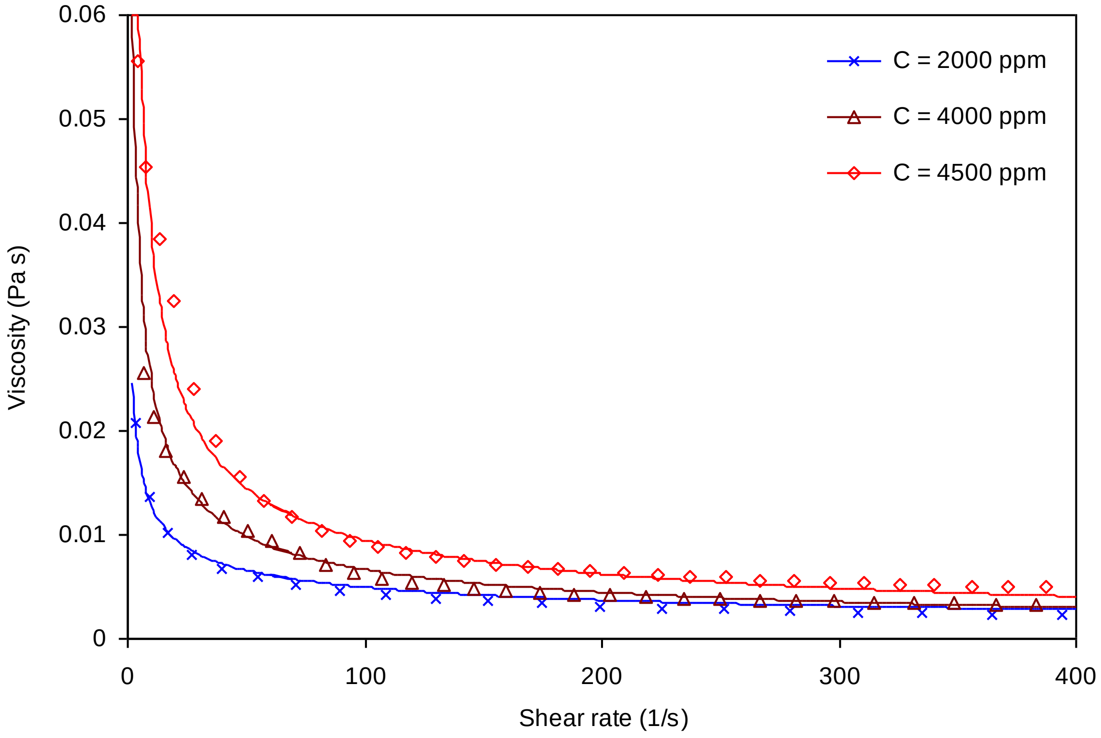

Fig 10: Viscosity-shear rate curves at $T = 303\,\text{K}$ **where dots represent the experimental data and solid lines represent Power law model fit.**

Table 1. Parameters of Oldroyd 4-constant model for mLLDPE solutions.

Concentration (ppm)	Temperature (K)	Model parameters			
		η_o (Pa.s)	λ_1 (s)	λ_2 / λ_1	λ_5 / λ_1
4000	293	0.035	0.04	0.12	1.14
	303	0.027	0.04	0.160	1.14
	313	0.013	0.04	0.18	1.14
	323	0.008	0.04	0.20	1.14
4500	293	0.0867	0.04	0.090	1.30
	303	0.0556	0.04	0.070	1.20
	313	0.0153	0.04	0.20	1
	323	0.0078	0.04	0.300	1

Table 2. Parameters of Giesekus model for mLLDPE solutions.

Concentration (ppm)	Temperature (K)	Model parameters			
		η_o (Pa.s)	λ_1 (s)	λ_2 / λ_1	α
1000	293	0.0145	0.08	0.120	0.180
	303	0.0120	0.10	0.130	0.070
	313	0.0100	0.10	0.130	0.035
	323	0.0083	0.10	0.170	0.035
2000	293	0.0250	0.24	0.075	0.100
	303	0.0230	0.30	0.080	0.060
	313	0.0200	0.40	0.090	0.030
	323	0.0170	0.70	0.100	0.013
4000	293	0.0400	0.26	0.080	0.070
	303	0.0276	0.22	0.060	0.030
	313	0.0110	0.20	0.190	0.030
	323	0.0082	0.35	0.250	0.015
4500	293	0.1000	0.26	0.030	0.030
	303	0.0700	0.26	0.040	0.080
	313	0.0153	0.26	0.120	0.015
	323	0.0078	0.26	0.300	0.015

Table 3. Parameters of Power law model for mLLDPE solutions.

Concentration (ppm)	Temperature (K)	Model parameters			
		Correct data		Basheer et al. data	
		m(Pa.s^n)	n (-)	m(Pa.s^n)	n (-)
1000	303	0.018	0.70		
2000		0.035	0.55		
4000		0.065	0.51		
5400		0.140	0.41		
4000	293	0.0756	0.5044	0.45	0.5044
	303	0.065	0.51	0.42	0.5264
	313	0.025	0.59	0.26	0.5795
	323	0.011	0.74	0.20	0.5671

Concentration Effects

To further illustrate the dependence on concentration that viscosity exhibits, figures 6, 8 and 10 show the viscosity as a function of mLLDPE concentration, at constant temperature, $T = 303\,K$. The figures indicate that, the mLLDPE solutions exhibit a large dependence on concentration. The effects of concentration on apparent viscosity at different shear rates and at $T = 303\,K$ are given in figue 11. It is observed that mLLDPE solutions at higher concentration will have higher viscosity.

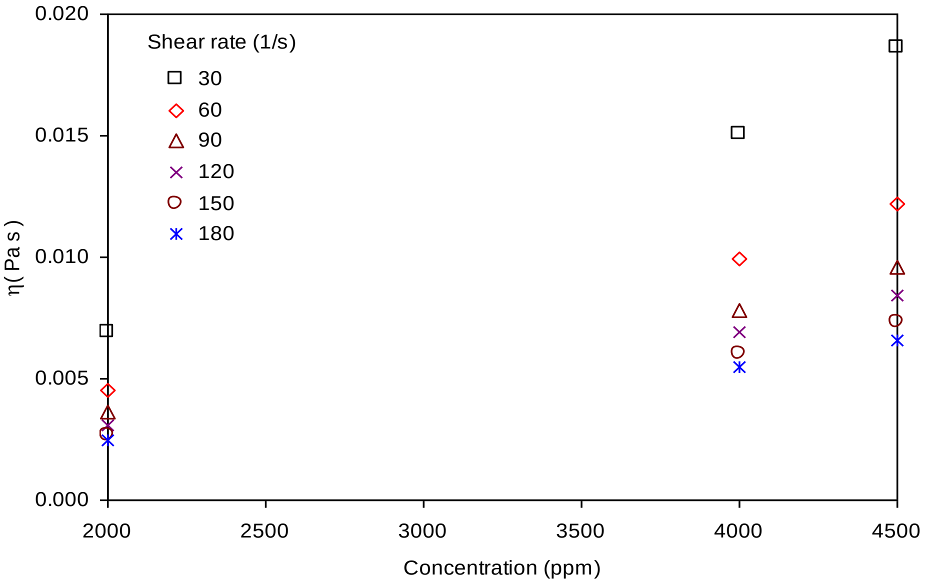

Fig 11: Experimental data for the apparent viscosity as a function of concentration at $T = 303\,K$ **and at different shear rates.**

The relationship between concentration and apparent viscosity is expressed by the following expression:

$$\eta = a_1 \mathrm{Exp}(b_1 C)\,, \tag{17}$$

which linearised as:

$$\ln\eta = b_1 C + \ln a_1 . \tag{18}$$

According to the last equation, plot of $\ln\eta$ and concentration should be a straight line, figure 12, where b_1 and a_1 are obtained from the slope and the intersection respectively. Values of the parameters evaluated from figure 12 are given as $a_1 = 0.02214\,\dot{\gamma}^{-0.578}$ and $b_1 = 0.00039$. Therefore, the data have a trend line equation:

$$\eta = 0.02214\dot{\gamma}^{-0.578}\,\mathrm{Exp}(0.00039\,C)\,. \tag{19}$$

Equation 19 represents a new correlation for the effect of concentration on the apparent viscosity of mLLDPE solutions. Figure 13 shows that, the proposed correlation is in a good agreement wit the experimental data

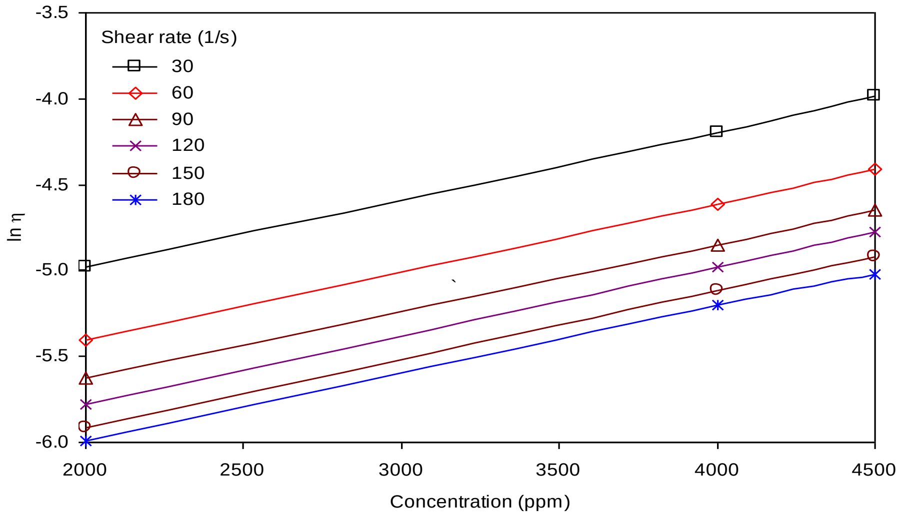

Fig 12: Effects of concentration on the apparent viscosity at $T = 303\,K$ **and at different shear rates.**

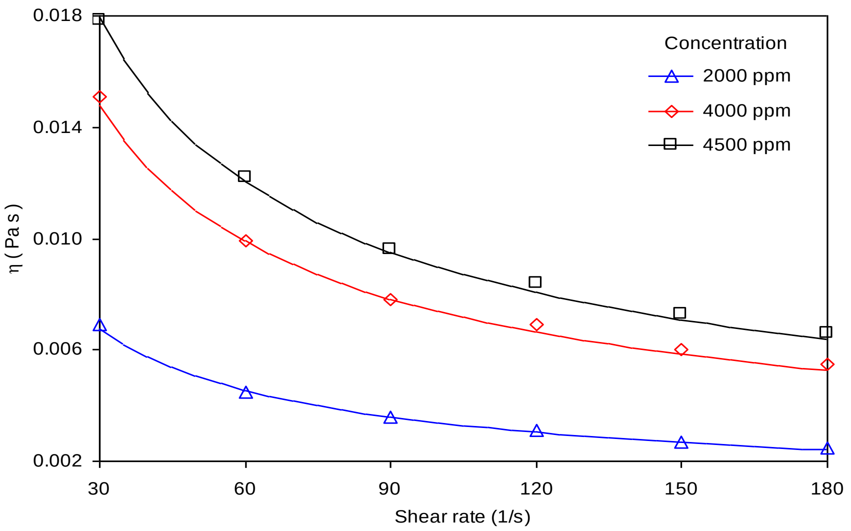

Fig 13: Effects of concentration on apparent viscosity, where solid lines represent the proposed correlations fit from equation 19 and marks represents the experimental data.

Temperature Effects

Temperature has an important role on both rheological characteristics and melt processing of any polymeric fluid [26]. The applicability of the Arrhenius model to describe the effect of temperature on the apparent viscosity, equation 15, at a constant concentration, $C = 4500\,\text{ppm}$, was investigated. As expected at constant shear rate and constant concentration, the temperatures have a direct effect on the apparent viscosity.

As given in figure 14, the experimental data shows that the apparent viscosity decrease with increasing temperature. With the help of figure 15, the parameters obtained from Arrhenius model by curve fitting is given in table 4. The applicability of the Arrhenius model to the apparent viscosity versus temperature data at different shear rates are shown in figure 16. Therefore, the dependence of the apparent viscosity on the temperature take the form:

$$\eta = 13586 \times 10^{-10}\, \dot{\gamma}^{-0.494}\, \text{Exp}\left(\frac{6106.11}{T}\right). \qquad (20)$$

Figure 16 shows that, the proposed correlation for the apparent viscosity as a function of the temperature is in a good agreement wit the experimental data.

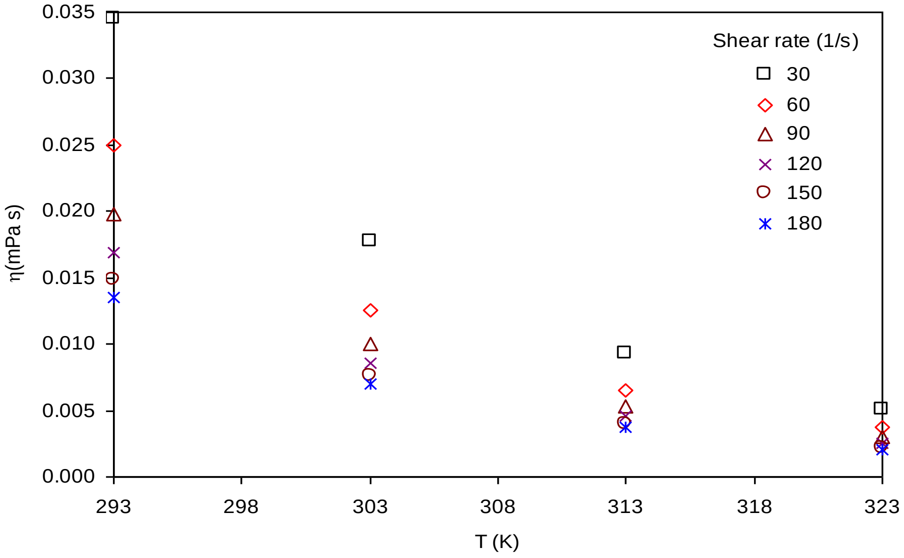

Fig 14: Experimental data for the apparent viscosity as a function of temperature at $C = 4500\,\text{ppm}$ and at different shear rates.

Table 4. Optimum values for the Arrhenius model for apparent viscosity.

$k\,(Pa \cdot s^{n})$	$E\,(K)$	$\Delta H = ER\ \ (J/mole)$
1.539×10^{-11}	6106.11	50741.774

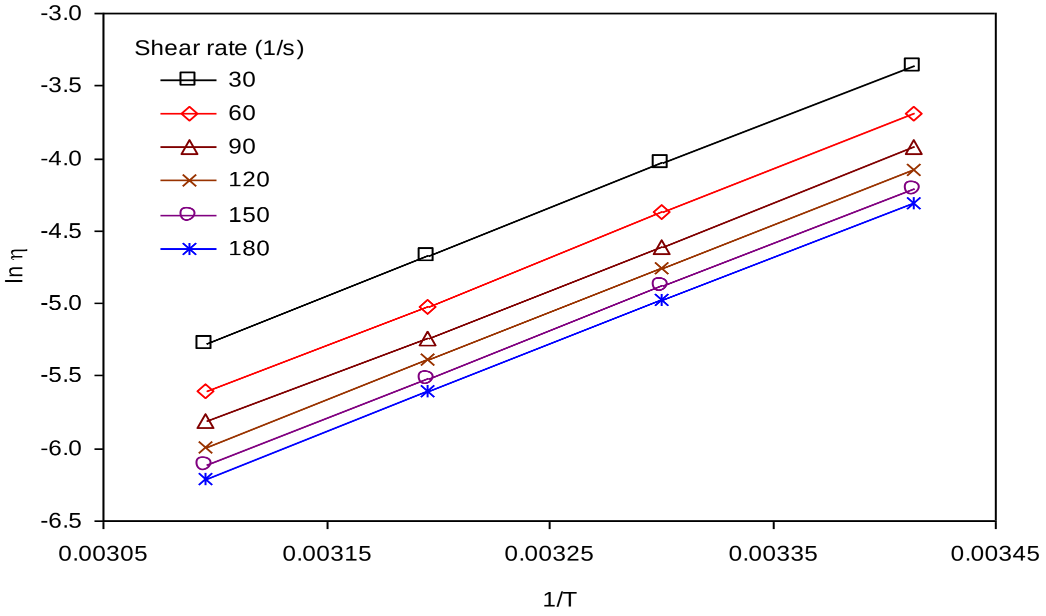

Fig 15: Effects of temperature on apparent viscosity at $C = 4500\,ppm$.

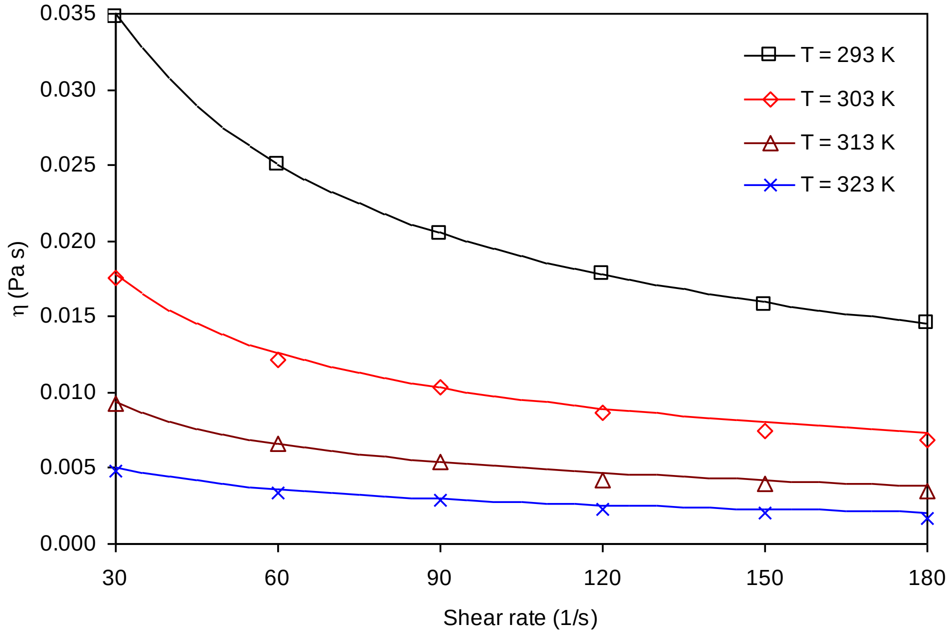

Fig 16: Effects of temperature on apparent viscosity, where solid lines represent the proposed correlations fit from equation 20 and marks represents the experimental data.

Suggested Correlation

For practical engineering applications, it is useful to get a simple equation describing the combined effect of concentration, temperature and shear rate on mLLDPE solutions rheology. As we have seen, many theoretical models have been commonly used by researchers. In the present paper, three theoretical models (Oldroyd 4-constant, Giesekus and Power law models) have been used to compare the measured data with the predicted viscosity as function of shear rate. The aforementioned models are the most powerful for predicting the rheological behavior of viscoelastic fluids in most dimensions.

In the present paper, by curve fitting on the experimental data, a new model to predict the apparent viscosity of mLLDPE solutions in terms of shear rate, temperature and concentration has been proposed as the following:

$$\eta(\dot{\gamma},C,T)=a_2\mathrm{Exp}\left[b_2C+\frac{d_2}{T}\right]\bar{\eta}(\dot{\gamma})\,, \tag{21}$$

where a_2, b_2 and d_2, are the proposed model parameters and $\bar{\eta}(\dot{\gamma})$ is taken from Giesekus model, equation 9 as:

$$\bar{\eta}(\dot{\gamma})=\frac{\lambda_2}{\lambda_1}+\left(1-\frac{\lambda_2}{\lambda_1}\right)\frac{(1-f)^2}{1+(1-2\alpha)f}\,, \tag{22}$$

Giesekus parameters λ_1, λ_2 and α as functions of mLLDPE concentration and temperature are given in table 5. Also, the optimum values of the proposed model a_2, b_2 and d_2 are given in table 6. These parameters are calculated as a function of mLLDPE concentration and temperature. Therefore, the final form of the correlation is as follows:

$$\eta(\dot{\gamma},C,T)=0.0075\mathrm{Exp}\left[C\left(-0.0039+\frac{1.324}{T}\right)\right]\bar{\eta}(\dot{\gamma})\,. \tag{23}$$

The combined effects of shear rate, concentration and temperature on mLLDPE viscosity can be seen from figures 17, 18 and 19 (in three dimensions form).

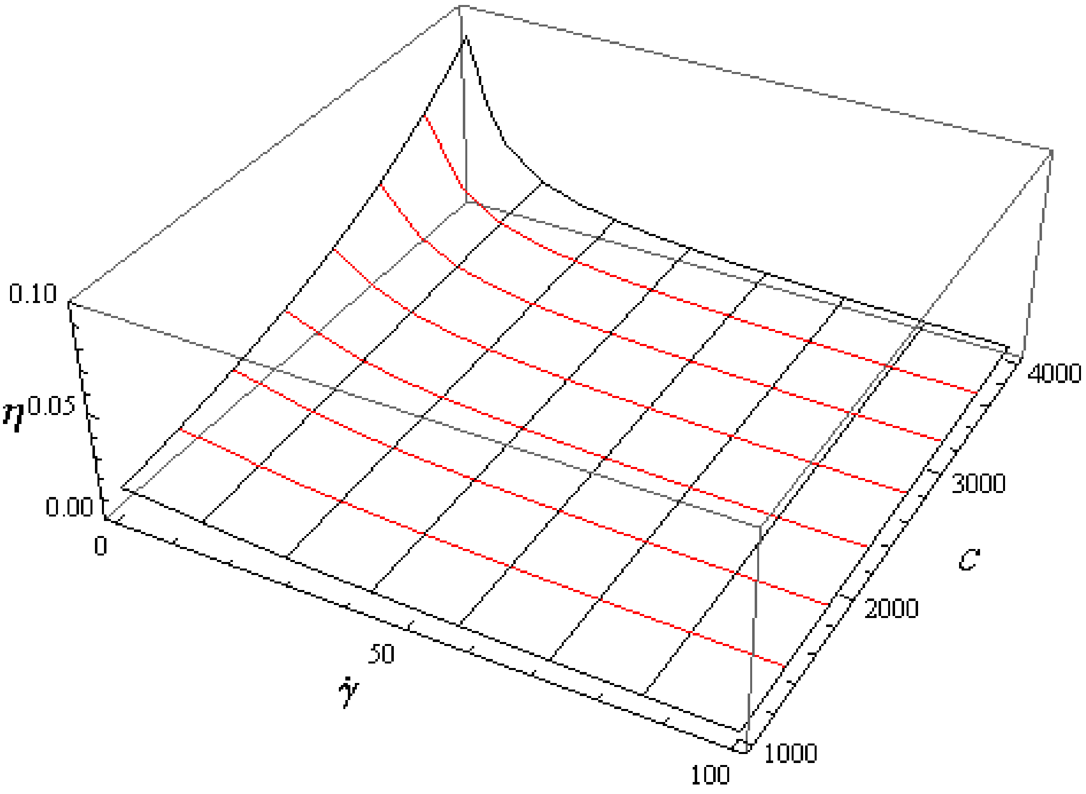

Fig 17: The combined effects of $\dot{\gamma}$ and C on η of mLLDPE solutions at $T = 293\,K$.

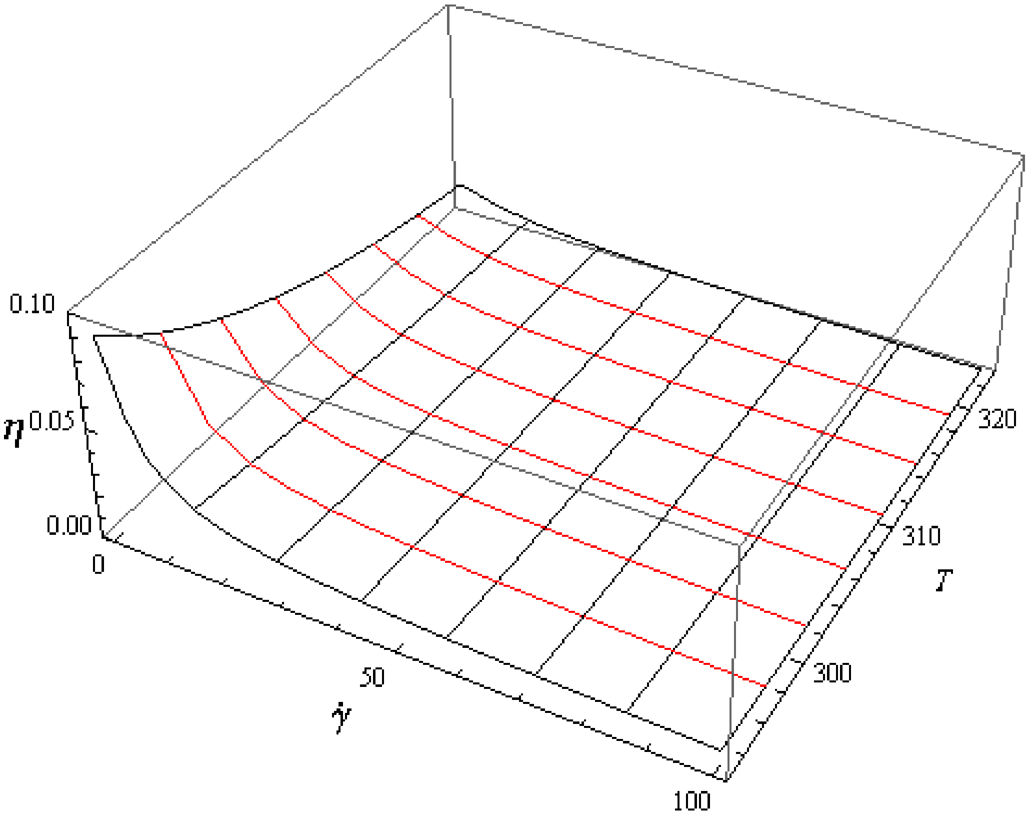

Fig 18: The combined effects of $\dot{\gamma}$ **and** T **on** η **of mLLDPE solutions at** $C = 4000\,\text{ppm}$**.**

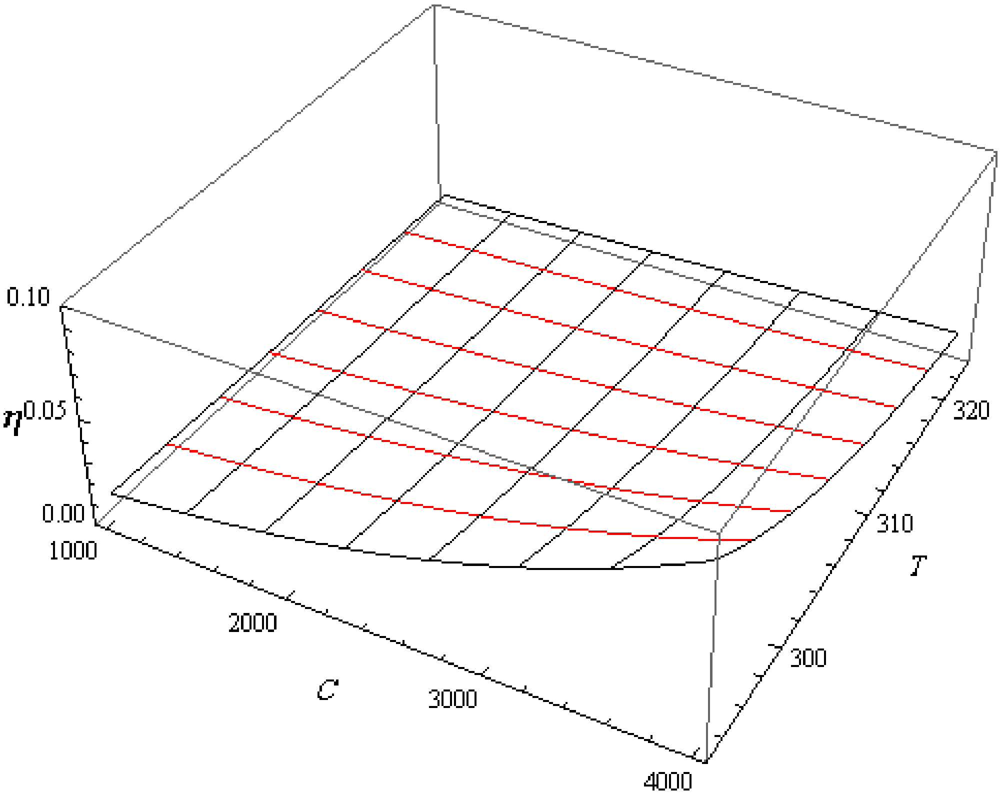

Fig 19: The combined effects of C **and** T **on** η **of mLLDPE solutions at** $\dot{\gamma} = 50\,\text{s}^{-1}$**.**

Table 5. Giesekus model parameters for mLLDPE solutions as functions of concentration and temperature.

λ_1 (s)	λ_2 / λ_1	α
0.5	$0.08 - 10^{-5} C$	$0.000695\ \mathrm{Exp}\left[C\left(-0.0043 + \frac{1.6357}{T}\right)\right]$

Table 6. Parameters in the proposed model, equation 21.

Concentration (ppm)	Temperature (K)	a_2 (Pa s)	b_2	d_2 (K)
1000	293	0.00028284	0.0006	964.15
	303	0.00031529	0.0005	964.15
	313	0.00034903	0.0003	964.15
	323	0.00038396	0.0002	964.15
2000	293	2.5213E-06	0.0006	2347.15
	303	3.2843E-06	0.0005	2347.15
	313	4.2065E-06	0.0003	2347.15
	323	5.3058E-06	0.0002	2347.15
4000	293	2.0034E-10	0.0006	5113.15
	303	3.5636E-10	0.0005	5113.15
	313	6.1099E-10	0.0003	5113.15
	323	1.0132E-09	0.0002	5113.15
4500	293	1.8915E-11	0.0006	5804.65
	303	3.6371E-11	0.0005	5804.65
	313	6.7075E-11	0.0003	5804.65
	323	1.1910E-10	0.0002	5804.65

Figure 20 shows a comparison between the results of the experimental data and the predicted model for the apparent viscosity of mLLDPE solutions. As can be seen, the results of the model are in agreement with those of the experimental data in all the studied temperatures and concentrations.

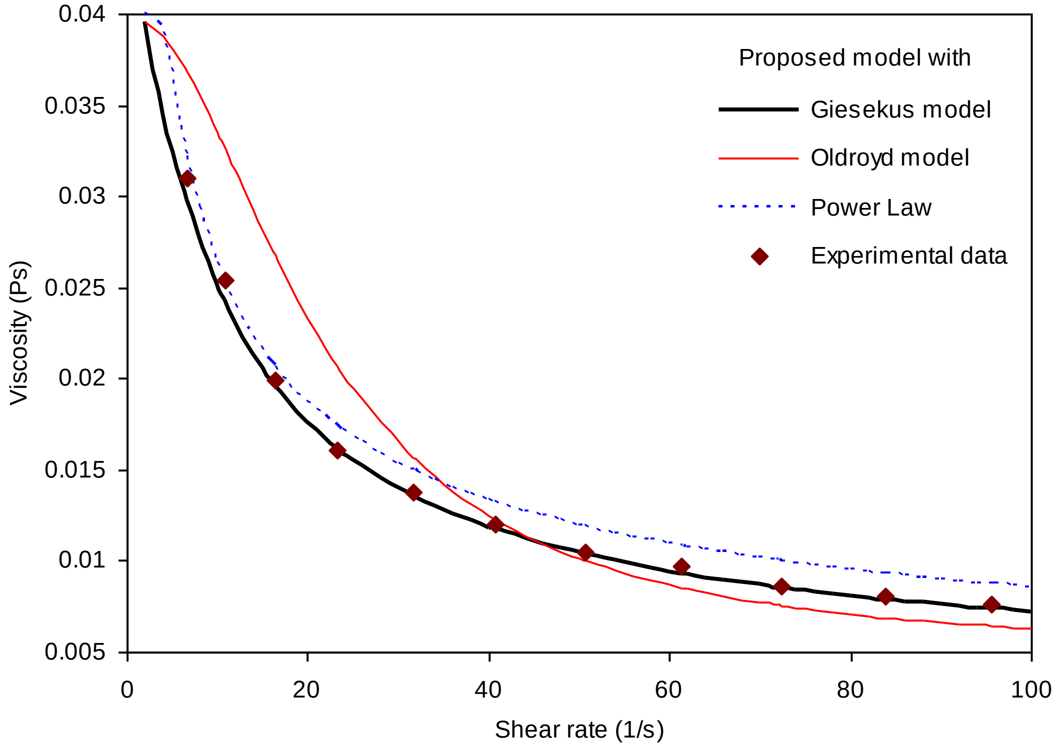

Fig 20: Viscosity versus shear rate for mLLDPE solutions at $C = 4000\,\text{ppm}$ **and** $T = 293\,\text{K}$**, where solid lines represent the proposed correlations fit from equation 23 by using the three models.**

CONCLUSION

The aim of this study was to determine the rheological behavior of mLLDPE solutions at different concentrations and temperatures by using rotational rheometer model AR-G2 with parallel plate geometry. The models that have been investigated are the Oldroyd 4-constant model, Giesekus model and power law model. Based on the experimental data, a new model to predict the apparent viscosity of mLLDPE solutions has been proposed. Hence, the following points can be concluded:

- Depending on the mLLDPE concentration and temperature, different flow behavior are observed. At high temperature and low concentration, the rheological behavior simulate a linear relationship (Newtonian fluid) which indicates that the shear rate has less effect on viscosity. At low temperature and high concentration, the shear rate has a large effect on the apparent viscosity of mLLDPE solutions which simulate the non-Newtonian fluids.
- For the same temperature and concentration the viscosity varies exponentially with the shear rate.
- The drop in viscosity is very sharp as shear rate increases slightly.
- The higher the temperature the lower the viscosity at the same shear rate.
- The results show that Giesekus model perform better than Oldroyd 4-constant and Power law models.

REFERENCES

[1] Basheer, A., Al-Fariss, T.F., Al-Sewailm, F.D. and Elleithy R. Rheology of metallocene linear low density polyethylene solutions, King Saud University, college of engineering, chemical engineering department, (2008).

[2] Morrison, F.A. 2001. Understanding Rheology, Oxford University Press, New York.

[3] Rao, M.A. 1999a. Introduction in rheology of fluid and semisolid foods principles and applications, 1-22. USA: Aspen publishers, Inc.

[4] Steffe, James, F. 1996. Introduction to rheology. In rheological methods in food process engineering. 1-91. USA: Freeman press.

[5] White, J.L., Czarnecki, L., and Tanaka, H. Experimental studies of the influence of particle and fiber reinforcement on rheological properties of polymer melts. Rub. Chem. Technol. 53, no 4 (1980) 823-835.

[6] Mills, N., 2005. Plastics, Microstructure and Engineering Applications, Elsevier.

[7] Alvarez, E., Cancela, M.A. and Maceiras, R. Effect of temperature on rheological properties of different jams. International Journal of Food Properties 9 (2006) 135-146.

[8] Esfe, M.H. and Saedodin, S. An experimental investigation and new correlation of viscosity of Zno-EG nanofluid at various temperatures and different solid volume fractions. Exp. Thermal Fluid Sci. 55 (2014) 1-5.

[9] Ahmmed, N., Asirvatham, L.G. and Wongwises, S. Effect of volume concentration and temperature on viscosity and surface tension of grapheme-water nanofluid for heat transfer applications. J. Therm. Anal Calorim, 123 (2015)1399-1409.

[10] Raymond, B.S. and Charles, E.C. 1981. Polymer Chemistry, Marcel Dekker, Inc., New York.

[11] Tanner, R.I. 2000. Engineering Rheology. 2nd Edition, Oxford University Press, Oxford.

[12] Diego, G.D., Jose, M.N. and Lourdes, C.Q. Influence of concentration and temperature upon rheology of k-carrageen aqueous solutions. EJEAFChe, 5(1) (2006) 1213-1220.

[13] Mohsen, B., Arash, K., Masoud, A. and Somchai, W. Effects of temperature and concentration on the viscosity of nanofluids made of single-wall carbon nanotubes in ethylene glycol. Int. Communications in Heat and Mass Transfer, 74 (2016) 108-113.

[14] Keshani, S., Luqman Chuah, A. and Russly, A.R. Effect of temperature and concentration on rheological properties pomelo juice concentrates. Int. Food Research journal 19(2) (2012) 553-562.

[15] Sehyun Shin and Young I. Cho. Temperature effect on non-Newtonian viscosity of an aqueous polyacrylamide solution. Int. Communications in Heat and Mass Transfer. 20 (1993) 831-844.

[16] Bird, R.B., Armstrong, R.C. and Hassager, O. 1987a. Dynamics of Polymeric Liquids. Vol. 1, Fluid Mechanics, 2nd ed., Wiley-Interscience, New York.

[17] Bird, R.B., Curtiss, C.F., Armstrong, R.C. and Hassager, O. 1987b. Dynamics of Polymeric Liquids. Vol. 2, Kinetic Theory, 2nd ed., Wiley-Interscience, New York.

[18] Giesekus, H. Constitutive equations for polymer fluids based on the concept of configuration-dependent molecular mobility: A generalized mean-configuration model. Journal of non-Newtonian fluid mechanics, 17 (1985) 349-372.

[19] Giesekus, H., J. Non-Newtonian Fluid Mech., 12 (1983) 367.

[20] Mostafaiyan, M., Khodabandehlou, K. and Sharif, F. Analysis of a viscoelastic fluid in an annulus using Giesekus model. Journal of non-Newtonian Fluid mechanics, 118 (2004) 49-55.

[21] Bouldin, M., Kulicke, W. and Kehler, H. Prediction of the non-Newtonian viscosity and shear stability of polymer solutions. Colloid polymer Sci., 266 (1988) 793-805.

[22] Eyring, H. Viscosity, plastic and diffusion as examples of absolute reaction rates. J. Chem. Phys., 4 (1936) 283.

[23] Amin, M.B. and Maddox, R.N. Estimate viscosity vs. temperature. Hydrocarbon Processing, (1980) 131-135.

[24] Al-Zahrani, S.M. 1990. Rheology of polymer solutions and their flow behavior through porous media. Master thesis at chemical engineering department, Kin Saud University.

[25] Al-Fariss, T.F., Al-Zahrani, S.M. Rheology behavior of some dilute polimer solutions. Journal of King Abdulaziz University, Engineering Science, 5 (1993) 95-109.

[26] Denn, M.M. 2008. Polymer Melt Processing: Foundations in Fluid Mechanics and Heat Transfer. Cambridge University Press, New York.

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