



# Temperature Dependent Rheological Behaviour of Plastisols Used in Coated Fabrics

Ali Zadhoush<sup>1\*</sup>, Mohammad Ali Alsharif<sup>1</sup>,

Mohammad Reza Moghareh-Abed<sup>1</sup>, and Majid Haghghat<sup>2</sup>

(1) Textile Engineering Department; (2) Chemical Engineering Department,  
Isfahan University of Technology, Isfahan-84156, Iran

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## ABSTRACT

The rheological behaviour of plastisols used in coated fabrics was studied by using a computer controlled concentric cylinder viscometer over the temperature range of 20-70°C. The results indicated that these pastes behave as Newtonian to pseudo-plastic fluids over the temperature range studied and the flow behaviour can be adequately described by the power-law model. The changes in the flow behaviour index ( $n$ ) with temperature were statistically negligible ( $P < 0.05$ ). Both consistency index ( $k$ ) and apparent viscosity ( $\eta$ ) decreased with increase in temperature. The effect of temperature on the viscosity of the paste was modelled using the Arrhenius model. The activation energy for the temperature range of 20-42°C, varied between 3.90 and 4.64 kJ.mol<sup>-1</sup>. Increase in the temperature of the paste had influence on Newtonian behaviour of the paste.

## Key Words:

plastisol;  
pseudo-plastic;  
coated fabrics;  
Arrhenius model;  
viscosity;

## INTRODUCTION

Rheological properties are of primary importance in successful application of coating technology. Plastisols have the most complex rheology [1,2]. They are suspensions consisting of particles of a PVC resin in a liquid continuous phase, formed mainly by a plasticizer and the stabilizer [3].

In the majority of transformation processes, PVC plastisols are coated on a substrate or on a mould, and then heated in an oven at about 180-200°C. During this process, the plastisol undergoes two processes, known as gelation and fusion. In the former, the PVC resin is dissolved in the continuous

(\*) To whom correspondence to be addressed.  
E-mail: [zadhoush@cc.iut.ac.ir](mailto:zadhoush@cc.iut.ac.ir)

phase, yielding a gel, where the PVC micro-crystallites exist and form a network structure responsible for its properties [4-8]. In the latter, the network structure is destroyed by fusion of the micro-crystallites, reaching a homogeneous state, which in a further cooling step leads to a solid polymer with all the properties of the plasticized PVC.

The gelation and fusion behaviour of PVC plastisols are very important to determine the process conditions and the features of the end products in the different industrial applications of this material and, consequently, the study of these processes is necessary by using different techniques. Rheology has proven to be a very suitable technique to study and characterize the behaviour of virgin plastisols in the gelation and fusion processes [9].

Extensive work has been carried out on many aspects of plastisol rheology such as temperature effect on the viscosity ageing of plastisols [10]; qualitative model that satisfactorily explains the rheological behaviour of PVC plastisols during gelation and fusion [11]; the influence of K-value and plasticizer [12]; determining viscosities over a wide range of concentrations, temperatures, and shear stresses [13]; and investigating pseudo-plastic behaviour of PVC plastisol [14]. But, literature search shows no publication on the applying Arrhenius model in order to correlate rheological properties of PVC plastisols, at present.

The objectives of this work were to determine and correlate the rheological properties of a typical plastisol paste used in coated fabrics and to determine the influence of temperature on the rheological properties of the paste. The results show that an increase in temperature will reduce the consistency index and viscosity during gelation and causing an increase during fusion. The correlation was modelled by Arrhenius equation adequately.

## THEORY

Two rheological models, namely Newtonian and power-law (also known as the Ostwald-de Waele equation), were fitted to the experimental data. The shear stress and shear rate relation for power-law fluids is given by:

$$\tau = k\dot{\gamma}^n \quad (1)$$

where,  $\tau$  is the shear stress,  $k$  is a constant (named a viscosity constant or consistency index),  $\dot{\gamma}$  is the shear rate,  $n$  is also a constant (named as power-law constant or power-law index). For  $n=1$ , eqn (1) describes a flow curve of a Newtonian fluid. For  $n<1$  a flow curve of a shear-thinning fluid and for  $n>1$  a flow curve of shear-thickening fluid are described.

The degree of pseudo-plasticity can be obtained by determining the shear thinning index ( $n$ ) value of each paste as per the following relationship:

$$\log \tau = \log k + n \log \dot{\gamma} \quad (2)$$

Hence, by plotting  $\log \tau$  versus  $\log \dot{\gamma}$  for different pastes, the value of  $n$  for each paste is the slope of the corresponding plot. The  $k$  values can be determined from the intercept elevation of the corresponding curves.

The apparent viscosity,  $\mu_a$ , for a power-law fluid is expressed by the following formula:

$$\mu_a = \frac{\tau}{\dot{\gamma}} \quad (3)$$

Considering in turn the Ostwald-de Waele model, the apparent viscosity can be written as follows:

$$\mu_a = k\dot{\gamma}^{n-1} \quad (4)$$

Of course, according to eqn (4), the viscosity,  $\mu_a$ , decreases with increase of shear rate,  $\dot{\gamma}$ , for  $n<1$  (a shear-thinning fluid), and  $\mu_a$  increases with increase of  $\dot{\gamma}$  for  $n>1$  (a shear-thickening fluid).

The temperature dependence of the apparent viscosity at constant shear rate can be described by the Arrhenius model [15-18]:

$$\eta = \eta_\infty \exp(E_a / RT) \quad (5)$$

where  $\eta$  is the viscosity,  $\eta_\infty$  is a pre-exponential factor,  $E_a$  is the activation energy for flow,  $R$  is the universal gas constant, and  $T$  is the absolute temperature. Eqn (5) can be expressed in the logarithmic form in order to estimate the parameters  $E_a$  and  $\eta_\infty$  for each concentration. Semi-log plots of viscosity versus inverse of absolute temperature gave significant

correlation coefficients (95% confidence level) when plotted using the linear regression method.

## EXPERIMENTAL

### Materials

A common formulation of plastisols including emulsion polyvinylchloride (PVC-E) with stabilizer (Vestolit) and co-stabilizer (Lankromark LZB 753, Akros Chemicals) used in manufacturing of coated fabrics was selected. Formulation of plastisol used was as follows (phr): PVC-E 100, stabilizer 2, co-stabilizer 2, filler 15, and plasticizer 30.

### Rheological Measurements

The rheological characteristics of the paste were studied by using a computer controlled rotational concentric cylinder viscometer, namely, a Haake Rotovisco RV12 SVI coaxial viscometer (Table 1). The temperature of the circulating bath was kept at  $20 \pm 0.1^\circ\text{C}$ . Various speeds of the drive were used to produce different shear rates. According to eqns (6) to (8), the shear stress (Pa), shear rate ( $\text{s}^{-1}$ ), and viscosity ( $\text{mPa}\cdot\text{s}$ ) were calculated:

$$\text{Shear stress : } \tau = A.S(\text{Pa}) \quad (6)$$

$$\text{Shear rate : } D = M.n(\text{s}^{-1}) \quad (7)$$

**Table 1.** Haake Rotovisco RV12 SVI coaxial viscometer characteristics.

Sensor system	SV I
Inner cylinder (rotor)	
- Radius, $R_i$ (mm)	10.1
- Height, L (mm)	61.4
Outer cylinder (cup)	
- Radius, $R_a$ (mm)	11.55
Radii ratio, $R_a/R_i$	1.14
Sample volume, V (mL)	12
Temperature: max ( $^\circ\text{C}$ )	100
min ( $^\circ\text{C}$ )	-30
Calculation factors	
- A (Pa/scale grad.)	12.4
- M (min/s)	0.89
- G (mPa.s/scale grad. Min)	13920

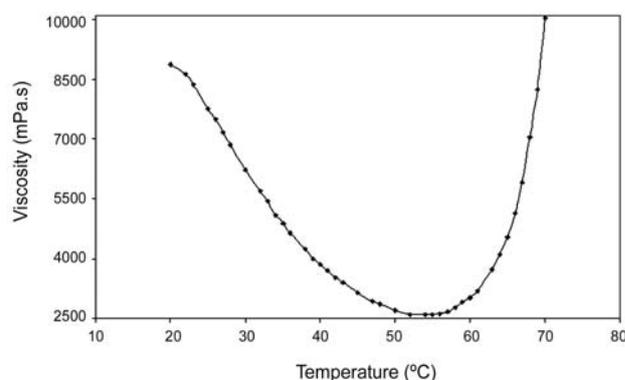
$$\text{Viscosity : } \eta = \frac{G.S}{n} (\text{mPa}\cdot\text{s}) \quad (8)$$

where A is the shear stress factor, depending on the type of measuring drive unit and sensor system (Pa/scale grad.), M is the shear rate factor, depending on the sensor system (min/s), G is the instrument factor, depending on the type of measuring drive unit and sensor system grad., n is the actual test speed ( $\text{min}^{-1}$ ) and S is the measuring value (scale grad.).

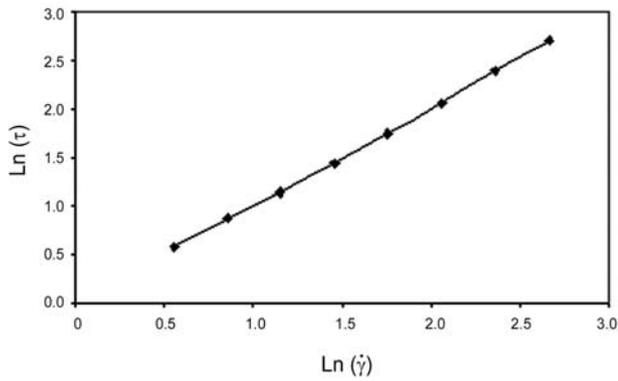
## RESULTS AND DISCUSSION

Values of apparent viscosities of the paste versus temperature are shown in Figure 1. As the paste is heated, there is initially a lowering of paste viscosity due to a drop in viscosities of the plasticizer with temperature. This is dependent on the nature of the plasticizer. A sharp rise of viscosity then occurs mainly due to adsorption of plasticizer by the polymer and due to solution of polymer in the plasticizer-gelation region. The temperature at which the sharp rise occurs is known as the gelation temperature. The system behaves as a suspension of non-interacting PVC particles in the plasticizer, which constitutes the continuous phase, whose viscosity decreases with temperature. Gelation begins to produce a marked increase in both the moduli and the viscosity, due to the swelling of the PVC particles and the interaction among themselves which develops a gel structure [5].

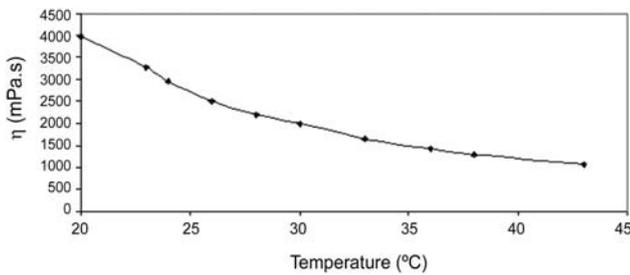
Several temperatures were chosen below the gelation temperature and the effect of temperature on the rheological behaviour was investigated.



**Figure 1.** Viscosity of plastisol at different temperatures.

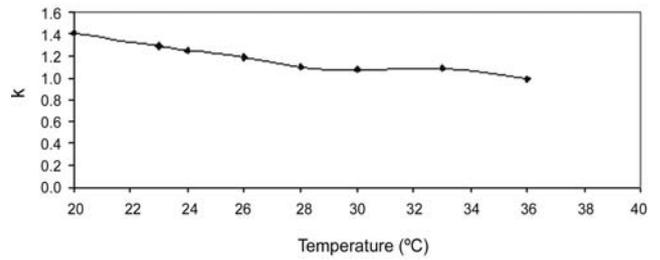


**Figure 2.** Logarithmic curve for shear stress versus shear rate at 44°C.

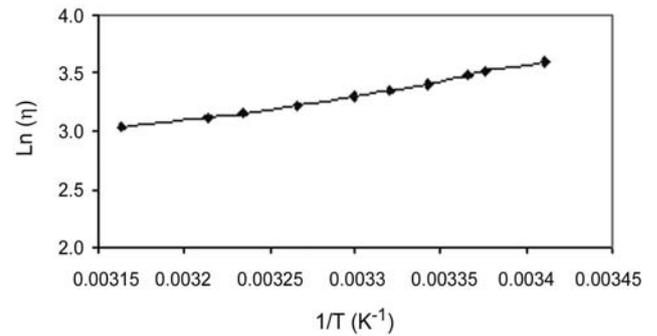


**Figure 3.** Apparent viscosity changes with temperature at constant shear rate (256 s<sup>-1</sup>).

Typical shear stress versus shear rate responses of the paste is shown in Figure 2, for the constant temperature of 44°C. As expected, shear stress is increased with increasing shear rate in a fairly linear fashion.



**Figure 4.** Variations in the consistency index with temperature.



**Figure 5.** Semi-logarithmic curve for viscosity versus inverse absolute temperature at constant shear rate (256 s<sup>-1</sup>).

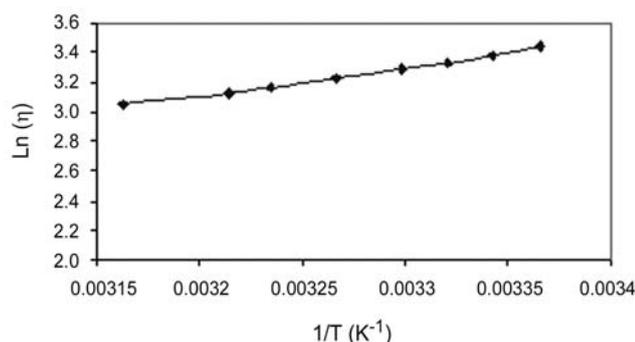
The power-law model had the least standard error and described the flow behaviour of the paste more adequately. Values of the parameters for the power-law at the different temperatures are presented in Table 2, within the temperature range of 42°C to 44°C,

**Table 2.** Estimated values of power-law model parameters.

T= 42°C	Slope=n	Intercept=Ln(K)	K=exp(Intercept)	Correlation coefficient	(Correlation coefficient) <sup>2</sup>
	1.0040	0.0804	1.0837	0.9987	0.9974
T= 44°C	Slope=n	Intercept=Ln(K)	K=exp(Intercept)	Correlation coefficient	(Correlation coefficient) <sup>2</sup>
	1.0130	-0.0130	0.9871	0.9995	0.9990

**Table 3.** Estimated values of E<sub>a</sub> and η<sub>∞</sub> at two constant shear rates.

γ̇ = 256 s <sup>-1</sup>	Slope=(E <sub>a</sub> /R)	E <sub>a</sub> =Slope*R	Intercept=Ln(η <sub>∞</sub> )	η <sub>∞</sub> =exp(Intercept)	Correlation coefficient	(Correlation coefficient) <sup>2</sup>
	2335.8	4.6413	-4.3930	0.0124	0.9942	0.9885
γ̇ = 512 s <sup>-1</sup>	Slope=(E <sub>a</sub> /R)	E <sub>a</sub> =Slope*R	Intercept=Ln(η <sub>∞</sub> )	η <sub>∞</sub> =exp(Intercept)	Correlation coefficient	(Correlation coefficient) <sup>2</sup>
	1962.2	3.8988	-3.1755	0.0418	0.9962	0.9925



**Figure 6.** Semi-logarithmic curve for viscosity versus inverse absolute temperature at constant shear rate ( $512 \text{ s}^{-1}$ ).

the flow behaviour indexes were closer to unity varying between 1.004 and 1.013. The consistency index varied between 1.084 and 0.987 for the same temperature range. The results obtained in this study over the temperature range used show that the paste was predominately Newtonian, whereas very slight pseudo-plasticity was shown at lower temperatures.

Figures 3 and 4 show the temperature dependence of the apparent viscosity at constant shear rate and consistency index, respectively. Apparent viscosity and consistency index decreased significantly with an increase in paste temperature.

The effect of temperature on viscosity coefficient can be correlated by an Arrhenius-type equation (Figures 5 and 6). Viscosity, as expected, decreased with increasing temperature. Estimated values of  $E_a$  and  $\eta_\infty$  at constant shear rates are found in Table 3. The activation energy for the temperature range of 20–42°C varied from 3.90 to 4.64  $\text{kJ}\cdot\text{mol}^{-1}$ .

## CONCLUSION

The paste formulation used in this study exhibited non-Newtonian shear-thinning to Newtonian behaviour over the temperature range of 20 to 42°C. The flow properties of the paste were described by using the well-known power-law model. The apparent viscosity and the consistency index were found to be a function of temperature, while the changes in the flow behaviour index with temperature were statistically negligible as compared with the former ones. The effect of temperature on the viscosity was very well

correlated with the Arrhenius equation ( $r^2 > 0.988$ ). The values of activation energy decreased with temperature from 4.64 to 3.90  $\text{kJ}\cdot\text{mol}^{-1}$ .

The viscosity of the plasticizer and its solvating power affect the viscosity of the paste, hence, lowering of paste viscosity is due when the paste is heated below the gelation temperature.

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