NUMERICAL INVESTIGATION ON THE POLYMER FLOW IN A RESTRICTION FLOW PATH

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By

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DECLARATION

I hereby declare that I have conducted, completed the research work and written the dissertation entitled 'Thesis Title'. I also declare that it has not been previously submitted for the award of any degree and diploma or other similar title of this for any other examining body or University.

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Date:		

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LIST OF ABBREVIATIOS

ABS	Acrylonitrile Butadiene Styrene
LDPE	Low Density Polyethylene
PVC	Polyvinyl Chloride
SAN	Styrene Acrylonitrile
2D	Two Dimensional
3D	Three Dimensional

NOMENCLATURES

$\dot{\gamma}$	Shear Rate
η	Viscosity
λ	Lambda
ρ	Density
Cp	Specific Heat
K	Thermal Conductivity
L	Length of Capillary
n	Power Law Index
r	Radius of capillary
T	Temperature

ABSTRAK

Reologi ialah kajian tentang ubah bentuk dan aliran bahan apabila daya dikenakan ke atasnya. Kajian reologi adalah penting untuk teknik pemprosesan dan kejayaannya bergantung kepada kelakuan reologi yang mengawal leburan polimer. Tingkah laku aliran ricih boleh dikawal dengan melaraskan pembolehubah pemprosesan seperti suhu, ricih, tekanan dan banyak lagi. Kajian ini bertujuan untuk mensimulasikan aliran ABS di dalam rheometer kapilari dan pengindeks aliran cair (MFI), dan juga untuk menentukan kesan suhu terhadap sifat reologi polimer. Model 2D rheometer kapilari dan model 3D bagi Indeks Aliran Leburan (Melt Flow Index-MFI) telah direka bentuk untuk mensimulasikan aliran Akrilonitril-Butadiena-Stirena (ABS) menggunakan Ansys Fluent. Penilaian perbandingan antara eksperimen dan simulasi dilakukan pada suhu yang berbeza. Bagi rheometer kapilari, suhu yang digunakan ialah 200 ℃ dan 220 ℃, manakala untuk MFI, suhu yang digunakan ialah 250 ℃, 260 ℃, 270 ℃. Hasilnya menunjukkan eksperimen dan simulasi membangunkan corak yang hampir sama. Lengkung kelikatan melawan kadar ricih menggambarkan hasil yang hampir serupa. Apabila kadar ricih meningkat, kelikatan berkurangan. Juga apabila suhu meningkat, kelikatan leburan polimer berkurangan akibat penguraian rantai molekul. Secara amnya, suhu mempengaruhi sifat aliran leburan polimer ABS.

ABSTRACT

Rheology is the studies of deformation and flow of material when force is applied on it. Rheology studies is crucial for the processing technique and the successful of it is depends on the controlling rheological behavior of the polymer melts. The shear flow behavior can be controlled by adjusting the processing variable such as temperature, shear, pressure and many more. This study aims to simulate the flow of ABS inside the capillary rheometer and melt flow indexer (MFI), and also to determine the effect of temperature on the rheology properties of the polymer. The 2D model of capillary rheometer and 3D model of melt flow indexer (MFI) were designed to simulate the flow of ABS using Ansys Fluent. Comparative evaluation between experiment and simulation were done at different temperature. For the capillary rheometer, temperature used were 200℃ and 220℃, while for MFI, the temperature used were 250℃, 260℃, 270℃. The result demonstrate the experiment and simulation develop almost the same pattern. The curve of viscosity against shear rate depicts almost similar result. As shear rates increases, the viscosity reduced. Also as temperature was increases, the viscosity of polymer melt reduced due to disentanglement of molecular chain. Generally, the temperature influences the flow properties of the ABS polymer melt.

CHAPTER 1

INTRODUCTION

1.1 Overview

This thesis presents the introduction of plastic polymer, how the demand urges the study on the rheology to improve the processing. The problem statement which described the reason of this research which differ from previous study also presented. The objective of the research, the scope of study and the outline of research also demonstrate in this thesis.

1.2 Introduction

Plastics have extensively been used in the packaging industry due to their exceptional performance, accounting for over 30% of total materials (Fact.MR). According to Mordor Intelligence, the Malaysian plastics market recorded at around 2,500 kilo tons in 2021, and the market is projected to register a CAGR of greater than 3% during the forecast period. In addition, the Malaysian plastics market had a surge in the year 2021 as many governments have lifted off many restrictions in the pandemic scenario. There was a huge demand for flexible packaging owing to raising demand in for personal and health care products, pharmaceuticals and packed food and beverages.

Due to the demand of plastic industries, optimization on the rheology or in simple word the flow behaviour is vital to produce excellent final product. The rheological measurement helpful for quality control and optimization of processing. Munstedt (2021) state that another interesting field of rheology is to provide information about molecular parameters of polymers and the structure build-up in heterogeneous polymeric systems. The investigation on rheology can be employed to qualitatively follow up the change of molecular parameters under external parameters like heat and mechanical deformation that may occur during various processing operations.

Common testing method to study the rheology are rotational rheometer, MFI tester, capillary rheometer etc. This testing provide data regarding on the rheology of the material. Despite on this testing method, simulation method also widely used nowadays to simulate the flow of polymer just like in the experimental one. Many flow phenomena distinctive to polymeric fluids have already been explained, at least qualitatively. However, quantitative predictions remain quite challenging, hampering the computational design of industrial polymer processing Larson (2015).

1.3 Problem Statement

Capillary rheometer is used to identify the flow behaviour of polymeric material. According to Seppo and Johanna (2012), the temperature of the fluid being measured is assumed to remain constant at the set temperature in capillary rheometer experiments. Due to highly viscous nature of polymer Seppo (2012), viscous heating effects may become significant with increasing shear rate, which may cause the actual melt temperature in the capillary to exceed the set temperature. In capillary rheometer, a large pressure drop is commonly associated with the flow in the die entrance (major) and exit (minor), regions (Mitsoulis & Hatzikiriakos, 2003). (Sombatsompop and Intawong, 2001) describes a number of researchers finding such corrections as very difficult to use because: (i) non-linearity of the correction curve, the extension of the imaginary length of the die, (ii) negative *N* values, (iii) *N* depends on the ratio barrel to die diameter, (iv) non-linearity of the correction curve, and (v) the method to determine *N* value is experimental result in difficulty to utilize the process of calculations.

In Melt Flow Index (MFI), consists of a cylindrical barrel and standard weight applied through the piston, a piston head, and dies to withdraw the materials. The concept almost similar with the capillary differ in term of the range of shear rate. Numerous study has been used MFI as testing method to identify the melt of polymer either virgin and blend. However, as far as the author concerned, there was no literature has been investigated on capillary rheometer and MFI using simulation. While the experiment has some limitation to visualize the flow inside the barrel, but it can be seen through simulation in detail by using pathlines to identify the vortex near the wall barrel. Therefore, in this study, the rheology of thermoplastic through the simulation of capillary rheometer and MFI would be identified.

Simulation enables the study on the effect of effect of temperature on the viscosity, flow pattern of polymer inside the barrel. It also allows studies of more complex systems because it creates observations by “moving forward” into the future as it imitates the real working condition inside the barrel, whereas other research methods attempt to look backwards across history to determine what happened Dooley (2002). Axelrod (1997) define different purposes of simulation in the social sciences such as prediction, performance, theory discovery and many more which useful to study the rheology properties of polymers also the improves the processing.

1.4 Objective

i. To simulate the melt of thermoplastic inside the capillary rheometer and MFI using ANSYS FLUENT.

ii. To investigate the effect of temperature on the melt rheology of thermoplastic using capillary rheometer and MFI.

1.5 Scope of Research

Rheology is a broad study on flow of material and one of the complex things to be done among researcher. There are many previous studies on rheology of polymer and their effect on many factors such as shear rate, temperature, types of processing method and etc. One of study done by Ariff, (2003) on the melt rheology of ABS using capillary rheometer. However, in this thesis, the works is aimed to simulate the flow of polymer melt inside the capillary rheometer and MFI (through barrel) moving out through die under different working condition. Then, the result will be discussed further in terms of experiment as well as numerical simulation. The polymer melt with different temperatures will be investigate since it is a major factor which can influence the rheology of thermoplastic.

1.6 Thesis Outline

The layout of the remaining chapters of this thesis is as follow.

First, in CHAPTER 1, a review about the problem statement, objective. It also described the introduction and scope of research.

In CHAPTER 2, a review of previous study related to rheology of polymer using capillary rheometer and Melt Flow Index (MFI). The viscosity of polymer melt, the factor affects, fluid characterization, and the correction procedure was also presented.

In CHAPTER 3, consists of description on the methodology of this study. The procedure to perform the DSC, and to simulate the capillary rheometer and MFI, the numerical method, mathematical modelling presented in this topic.

In CHAPTER 4, demonstrate the result and discussion of this study. The effect of temperature on the rheology and melt flow of thermoplastic will be discussed further.

CHAPTER 5 is the summarize the main conclusion of the study and recommendations for future research

CHAPTER 2

LITERATURE REVIEW

2.1 Overview

In this thesis, the introduction to rheology, the definition, the relationship between rheology and the processing, how important the rheology, factors that can affect rheology properties of thermoplastic was presented. Few of testing method which commonly used to investigate the flow of polymer also presented. The classification of fluid, the flow behavior of fluid and review on thermoplastic which is ABS was further discussed. Lastly, the conclusion remark which is the summarize of this chapter.

2.2 Introduction

Thermoplastic is widely used in the industry to produce various product in a broad range of field. Not just that, the processing uses to produce the product itself play a major role and requires optimum processing parameter. While to obtain optimum parameter, study on the rheology should be done to find out the correct choices during processing especially while the material is melting. As an example, there are few of study has been done to investigate the rheological properties of ABS using capillary rheometer via experiment. Previously, Ariff (2003) investigate the melt rheology of ABS via capillary rheometer and result of the flow pattern of polymer melt shows more parabolic as the temperature increased. Rheology is about the flow of polymer melt. Therefore, from previous study it was found that, rheology can be affected by internal and external issues such as molecular weight of polymer (internal), temperature (external) and etc. This factor influenced the viscosity of the polymer melt upon shear rates.

2.3 Rheology

Rheology is a science of deformation and flow of a material or polymer when stress or any force is applied on it. According Bhat and Kandagor (2014), polymer rheology is a scientific study of polymeric fluids deform when subjected to external stresses such as processing parameter and polymeric material composition. Processing parameters are the temperature, pressure, shear rate, flow geometry and many more. While for the polymeric materials composition, are the polymer blends, fillers or additive. Ianniruberto (2015) stated that synthetic plastic is the most used in plastic industry to produce goods for many different applications. Nowadays, polymer such as thermoplastic are widely use in the industry such as food packaging, kitchen utensil, and many more. However, the crucial part to produce final product is where the polymer needs to be transformed into liquid form before solidify.

2.3.1 Viscosity of Polymer Melt

Viscosity is the resistance of fluid to shearing and is the most important factor in polymer processing Nicholas and John (2019). In polymer melts, viscosity is depending on the shear rate. The higher the viscosity, the lower the shear rate. Shear is the motion between adjacent layer of moving fluid, as shown in Figure 2.1. Shear force acts in parallel and motion to develop shear stresses is due to the particle which moving inside the polymer. In the case of polymer which behave like a non-Newtonian fluid it is mainly depend on the shear rate, viscosity and temperature where it will determine either faster or slower the fluid (polymer) to flow.

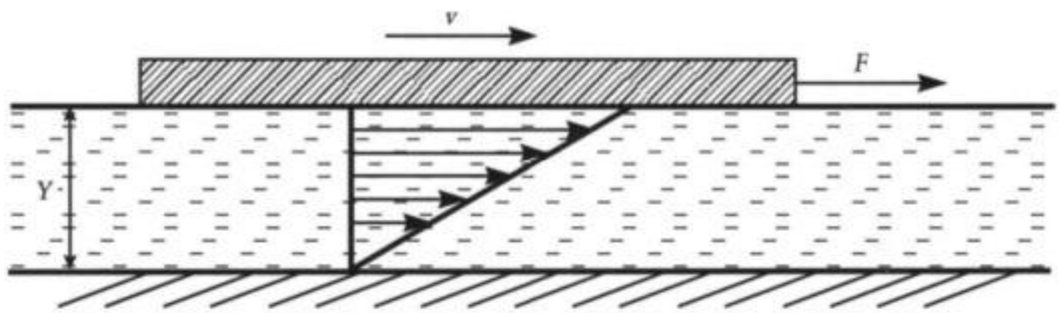


Figure 2.1: Motion between adjacent layer in moving fluid (polymer) Howard (2020).

Equation (2.1) is the formula of viscosity where shear stress divided by the shear rate. Polymer consists of chain which tangle like a spaghetti, when applied to certain temperature can affect the entanglement of molecular chain. This will result in reduction of viscosity due to alignment of long polymer chain disentanglement of the polymer. The higher the shear rate apply to the polymer, the easier for the polymer to moving and flow out through die.

$$\eta, viscosity = \frac{shear\ stress}{shear\ rate}\frac{\tau}{\dot{\gamma}} \quad (2.1)$$

2.3.2 Role of Rheology in Polymer Processing

Processing method are the most important part to convert polymer into fluid and solidify which commonly run under various level of temperature and pressure. Once the optimum temperature is applied on polymer, it will melt (turns into liquid form before solidify), causing the change in their molecular structure. Therefore, polymer rheology data is used in determining whether the polymer can be extruded or moulded into desired product or not. According to (Ariff et al, 2012), knowledge on rheology helps to optimise design of processing equipment such as extrusion die design, screw geometries of an extruder, various mould cavity for injection moulding and mixing devices. On top of that, processing is the fundamental of behaviour of materials such as ABS and PVC which widely used in injection moulding and blown moulding, thermoforming etc. In all these processes, rheological measurement correlates molecular weight, molecular structure to such processing behaviour such as flow rate, die swell, melt stability, and recoverable shear.

Processing problem while polymer in liquid form, issue like flashing might occur on final product in injection moulding while extrudate distortion and uneven surface of extrudate may be observed in profile extrusion Ariff (2003). This happened due to viscosity is not suitable with processing condition. The study by Abdelhalik et al (2017) revealed that for the materials with lower viscosity, the injection speed and barrel temperature, are the most influential parameters for increasing the amount of flash while for material with high viscosity, the barrel temperature is the most influential parameters for increasing the flash amount. Figure 2.2 show how rheology can be used to correlate end-use and processing performance to the polymer structure.

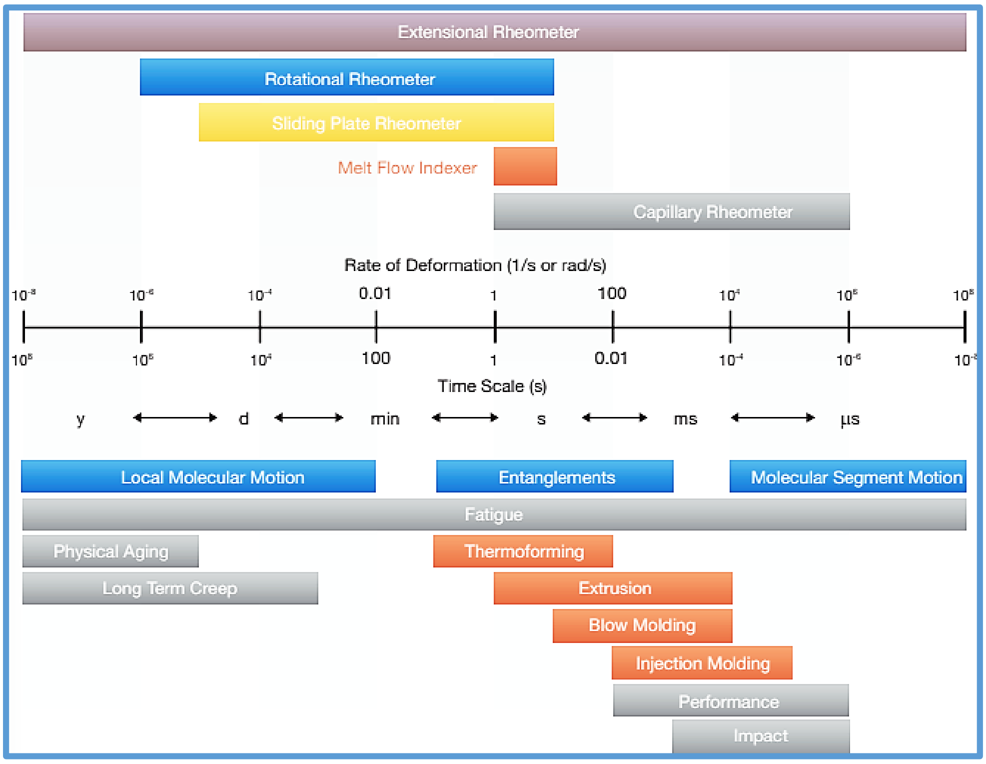


Figure 2.2: Interrelationship of processing conditions, flow properties, mechanical properties Osswald et al (2012).

2.3.3 Factors Affect Properties of Polymer Melt

Polymer melt can be influence by two major factor, internal and external factor. Internal factors are such as molecular weight, molecular weight distribution and polymer structure. External factors such as processing parameter; (temperature, pressure, shear rate, flow geometry) and polymeric materials composition. Study by Asif (2019), observed that rheology when studied under capillary flow conditions, was depended on rubber content (internal factor: molecular structure) on molecular weight (M_w) of SAN as changing M_w also changed interaction with the polybutadiene (PB) rubber particle.

Polymer at sufficiently high shear rates will completely disentangled and fully align. In this region, the viscosity of polymer melt will independent of the shear rate. Ariff (2003) studied on rheology ABS using capillary rheometer at temperature 200, 220, 240 and 260℃ demonstrate the curve of viscosity against shear rates at different temperature. They found that as shear rates is increased, the viscosity of ABS decreased accordingly with increasing temperature as shown in Figure 2.3. This condition is called as shear-thinning condition, where shear rate is inversely with viscosity according to Samuel (2020). The higher the viscosity, the lower the shear rates. As temperature increase, viscosity of the material also tends to increase (easy to flow) due to disentanglement of polymer chain.

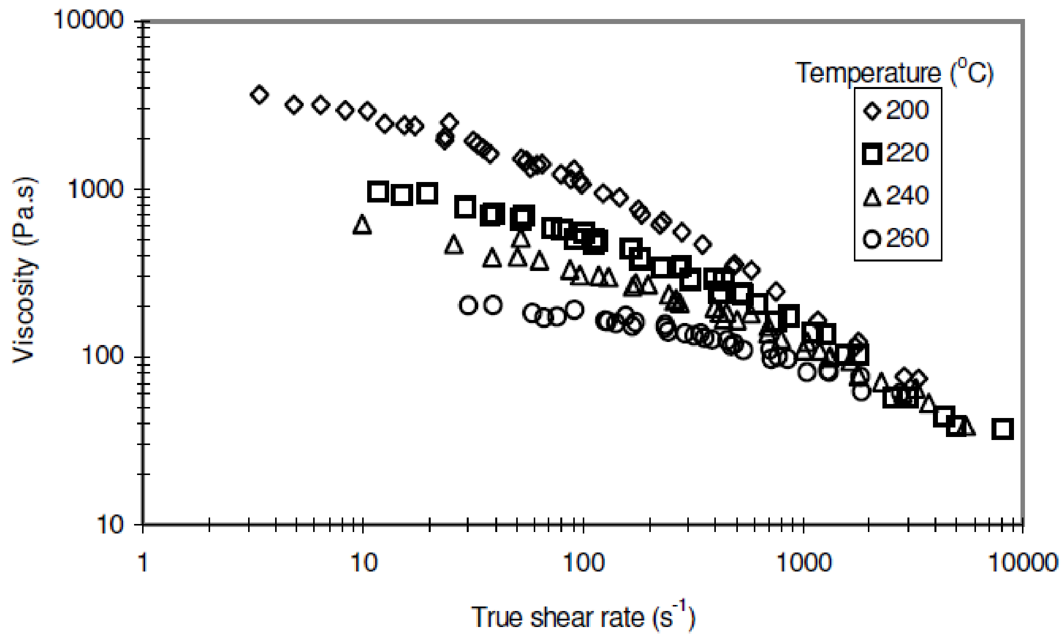


Figure 2.3: Curve of viscosity against shear rate at different temperature Ariff (2003).

2.4 Rheological measurement (Rheometer)

The reasons of the rheological testing are for characterization (molecular weight, molecular weight distribution), process performance (injection moulding, extrusion, blow moulding), and product performance, (temperature, pressure, dimensional stability, etc). Figure 2.4 shows several type of rheometer. Rheological measurement can be categorized into two categories, shear viscometer and shear rheometer. Shear viscometer is used to measure the viscosity of fluid over a limited shear rate range (~0.1-1000s^{-1}) while rheometer has the ability to measure viscosity over wide range of shear rate (0.01-10,000,000 s^{-1}). Hence, rheometer has the capability to measure viscosity over a broad range of shear rates.

Concentric/Coaxial cylinder rheometer, capillary rheometer, torque rheometer, and extensional flow rheometer are few of rheometer testing method. However, capillary rheometer is the mostly used as it has more advantage in term of handling and also shear rate and flow geometry are similar to the actual condition found in the process of extrusion and injection moulding according Gupta (2000). Tien (2009) stated that the design and operational principle of capillary rheometer are conducive to provide rheological data under high shear rates (on the order of $10^7 s^{-1}$) and from the mode of deformation of a material it will undergo in processes like extrusion, film blowing and injection moulding.

Coaxial cylinder rheometer is used for the low viscosity fluid, polymer solution, solid-in-solid suspension and emulsion. Halley (1992) stated that coaxial cylinder rheometer measure stress on cylinder after applied rotation of concentric surface. For torque rheometer, it is to measure stress on plate after rotation of adjacent plate. For capillary rheometer, pressure is applied to produces flow through capillary.

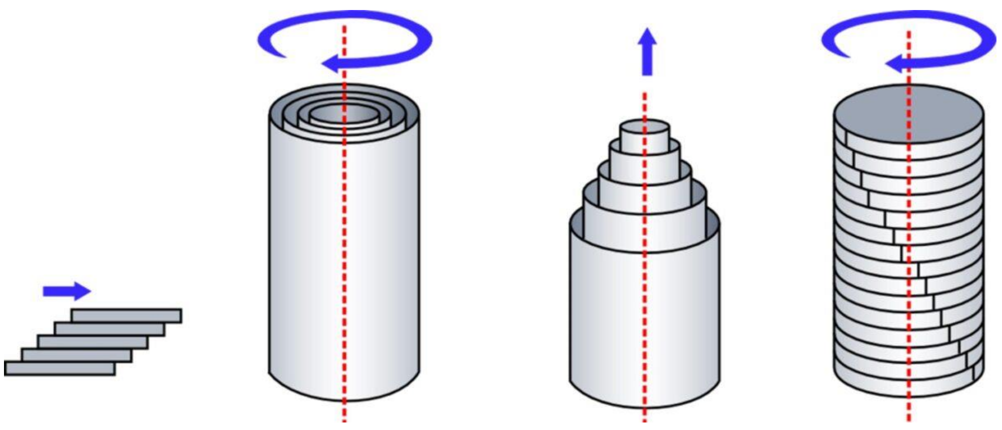


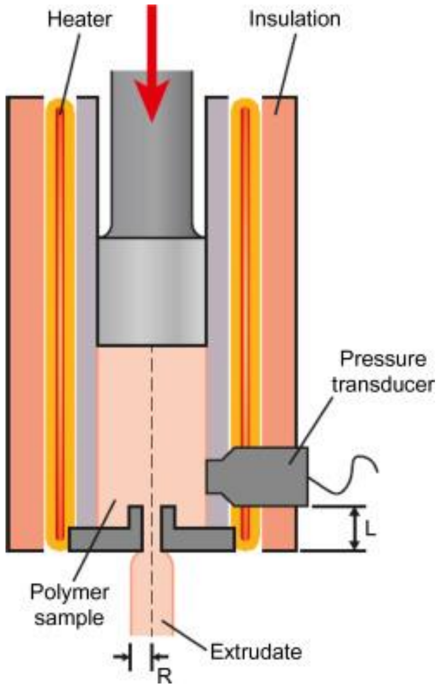
Figure 2.4: Type of rheometer.

2.4.1 Capillary Rheometer

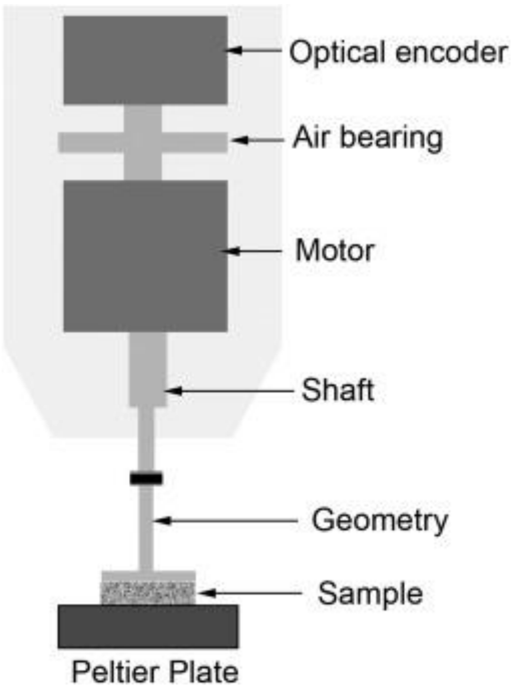
Capillary rheometer consists of heated barrel where polymer sample will be charge. There are two types of capillary rheometer, constant- rate and constant-stress as show in Figure 2 5. In the controlled stress-rate rheometer, Figure 2 5(b), the material is placed between two surfaces: bottom surface fixed and upper surface which rotates; hence, torque (stress) is independent that is apply to the geometry (Peyronel, 2018).

In the controlled-rate rheometer, Figure 2 5 (a), the material is placed between two plates, where the bottom moves at fixed speed and the torsional force produced on top plate is measured, thus, strain-rate is the independent variable and stress is the dependent one (Peyronel, 2018). For the controlled stress, the material is placed in between of two plates, upper known as geometry (rotate) and bottom (fixed). According to Ariff (2003), constant-stress rheometer uses constant gas pressure to move the molten polymer inside the capillary rheometer. Gas pressure which usually is nitrogen is assumed equal to pressure at the die entrance.

However, controlled-rate rheometer is the better approach as the stress can be increased in gradual controlled until yield point is observed. Semancik (1997) stated that the yield has to occur before the measurement can be performed using the controlled-rate rheometer. While for the controlled stress, there is no transducer and not automated just like the other one, hence it required longer time to complete.



(a) Controlled rate.



(b) Controlled stress

Figure 2 5: Schematic diagram of (a) Controlled-rate and (b) controlled-stress Osswald (2012).

2.5 Classification of Fluid

There are two basic types of fluid which are Newtonian and non-Newtonian fluids. Newtonian fluid is where the relationship between shear rate and shear stress is linear and the viscosity of fluid is constant no matter amount of shear is applied for a constant temperature. This type of fluid obeys Newton's law equation (eq. 2.2). In non-Newtonian fluid the relationship between shear rate and shear stress is not linear. For non-Newtonian fluid, the viscosity is not constant and among others it is depend on shear rate.

$$\tau = \eta \times \dot{\gamma} \tag{2.2}$$

Where, τ is shear stress, $\dot{\gamma}$ is shear rate and η is the viscosity.

Mashelkar (1992) observed that the flow of non-Newtonian fluid showing that flow of polymer is quantitatively different from the Newtonian fluid. In Figure 2.6 is the surface of the Newtonian fluid is depressed the rod, while non-Newtonian (polymeric fluid) tries to climb the rod. This climbing is known as the 'Weissenberg effect'. He added that Newtonian is not suit to describe the polymeric fluid because it deals with the large molecules, whose molecular weights may range from 10^5 to 10^8. Collyer (1973) stated that the properties of non-Newtonian behaviour are caused by the viscous dissipation of energy due to collision between large particles, or to distortion of or collision between colloidal structure.

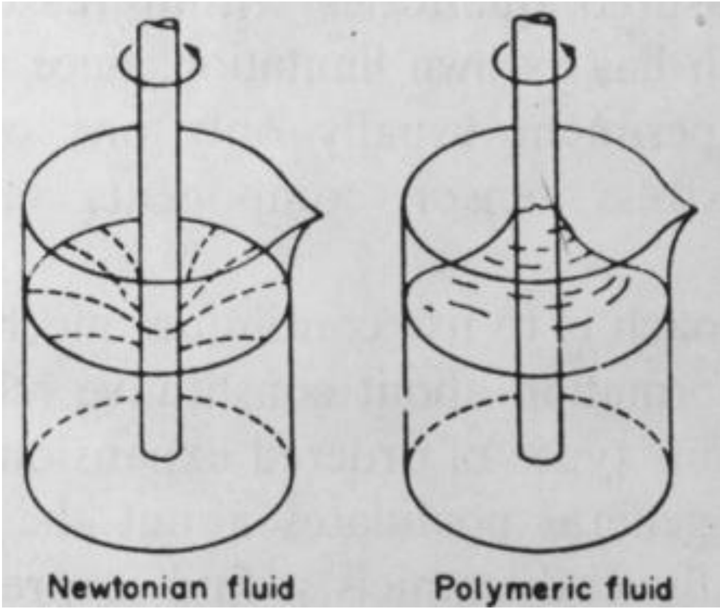


Figure 2.6: ‘Weissenberg effect’ of Newtonian and polymeric fluid Mashelkar (1992).

2.5.1 Time-independent Fluid Behaviour (non-Newtonian)

Non-Newtonian fluid can be categorized into two: time-independent fluid behaviour and time dependent fluid behaviour. Time-independent fluid consists of three different types: Bingham, pseudoplastic fluids and dilatant fluids Figure 2 7.

Bingham model in Figure 2 7 shows that shear stress is proportional with the shear rate. (Bill, 2012) Bingham plastic model most common rheological model used in the drilling industry and it is widely used because it is simple and estimate pressure loss in a turbulent condition with accuracy close to the other models. However, Bingham body is not observed in polymer melts although it has been referred to as the ‘ideal plastic’.

Shear thinning which often called *pseudoplastic* characterize by apparent viscosity that gradually decreases with increasing shear rate. As shear stresses is increased, molecules aligned to the direction of flow which result in the reduction the internal friction and allow greater shear rate.

Shear thickening effect was first observed by Reynolds in 1885 since this model required a dilation upon shearing. This model also requires that all suspensions of solids in liquid should exhibit dilatant behaviour when solid content is high Collyer (1973).

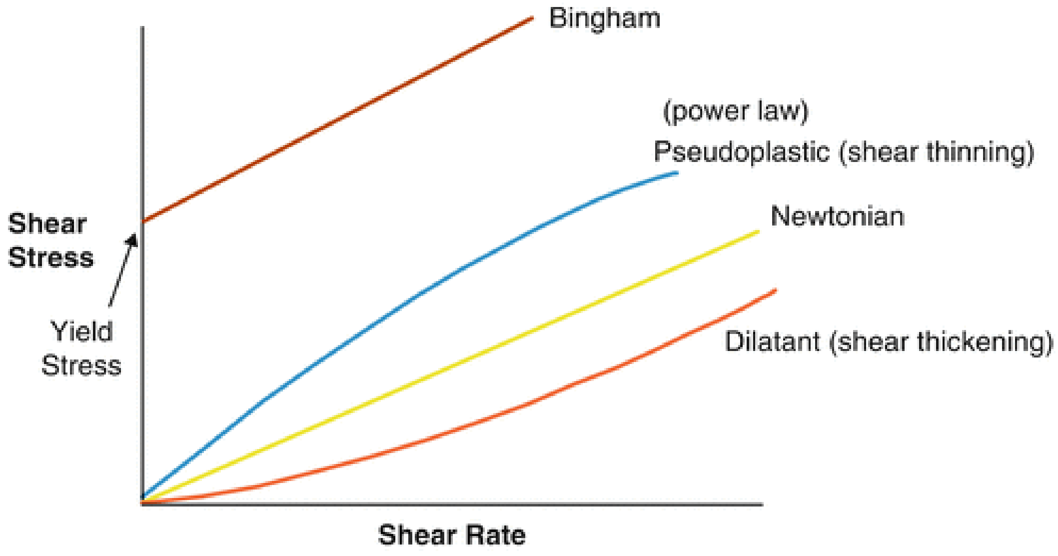


Figure 2 7: Curve of shear stress as a function of shear rate (Time-independent fluid).

2.5.2 Time-dependent Fluid Behaviour (non-Newtonian)

Figure 2.8 demonstrate the curve of time-dependent fluid: thixotropic and rheopectic. Thixotropic is the condition where the viscosity of some fluid, thus thixotropic is time-dependent phenomenon according to Koleske (2012), Mezger (2014), and Wilczynski (2001). Therefore, the viscosity of thixotropic fluid decreases time upon shearing, and recovered their original viscosity as a function of time without shearing. When subjected to varying shear rate, thixotropic will demonstrated ‘hysteresis loop’. The flow curve of a thixotropic fluid is shown in Figure 2.8. A simple loop test with the increase in the shear rate from zero to a maximum value and then a decrease to zero in the same way designates the flow curve according to Zolek (2016) as shown in Figure 2.9. Highly viscous printing inks is one of thixotropic fluid, thus the viscosity of a printed ink is higher than that of the printing ink on the rollers in the printing unit. Therefore, thixotropic phenomenon dries the highly viscous printing.

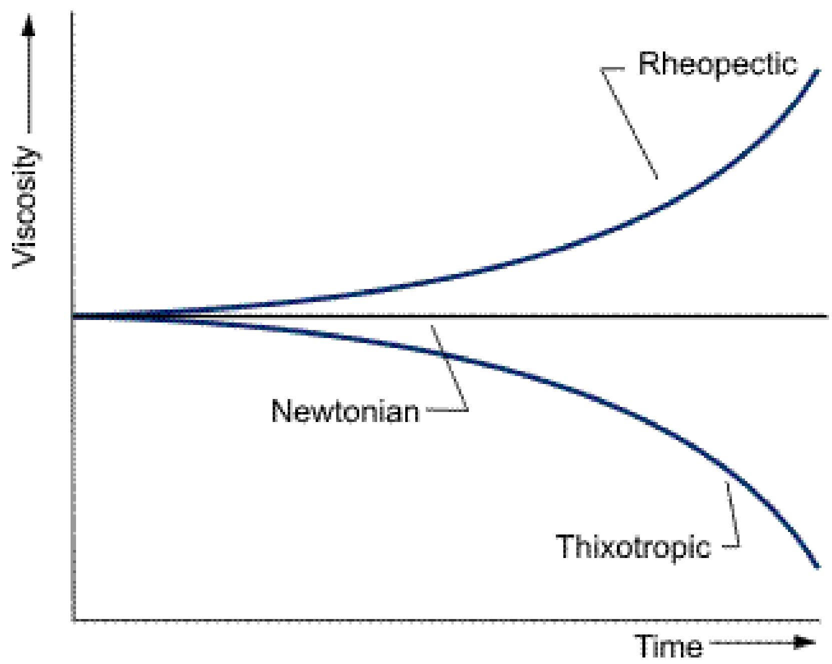


Figure 2.8: Time-dependent fluid.

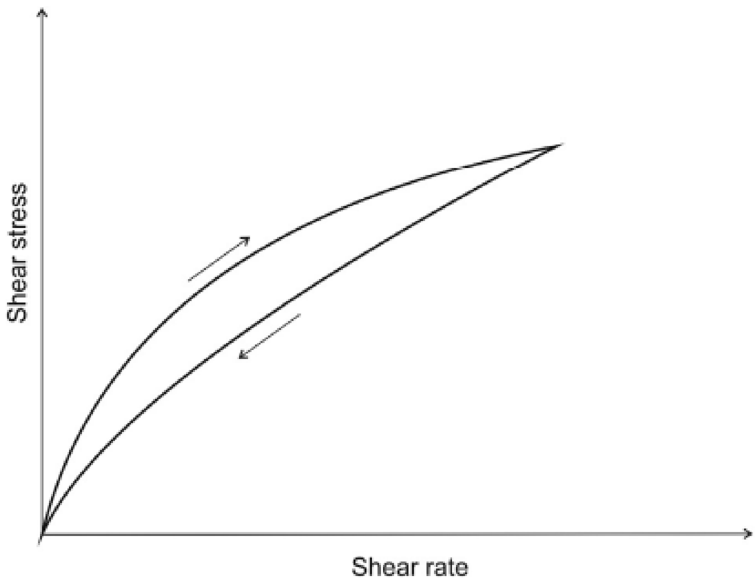


Figure 2.9: Hysteresis loop of thixotropy (Zolek, 2016).

2.6 Flow Behaviour of Real Fluid

Figure 2.10 is the flow and viscosity curve for a typical fluid of low solids concentration which indicates, at low shear rates, the viscosity is constant, while at high shear rate, viscosity constant but low. The viscosities in the first and second Newtonian region are referred as the zero-shear viscosity, μ_0 and the infinite shear viscosity, μ_∞.

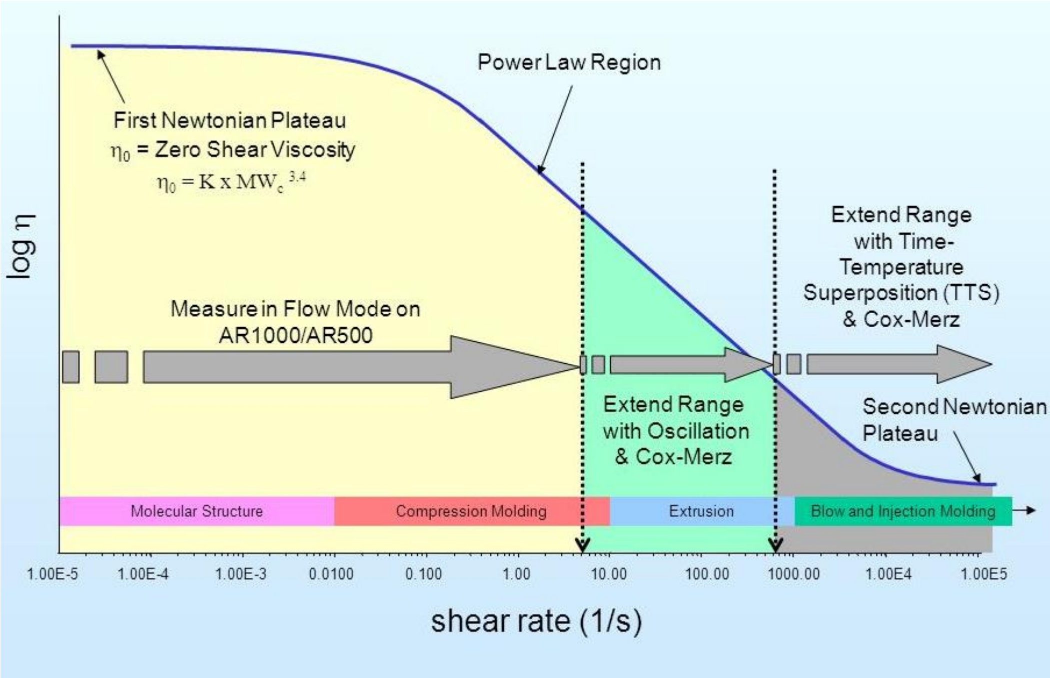


Figure 2.10: Typical flow curve of polymer melts.

2.7 Cross Model

The Cross model is an empirical equation that is used to fit non-Newtonian data according to Cross (1979). Hackley and Ferraris (2001) stated that this model describes *pseudoplastic* flow with asymptotic viscosities at zero (n_0) and infinite shear rates (μ_∞), and no *yield stress*. According to Reig (2005), Cross model is appropriate for post filling analysis (packing phase), because the temperature and pressure sensitivities of the zero-shear-rate viscosity are better presented. The expression of model is presented in equation below:

$$\eta = \frac{\eta_o}{1 + \left(\frac{\eta_o \dot{\gamma}}{\tau^*}\right)^{1-n}} \quad (2.3)$$

Where:

η_o is the viscosity of the material under-zero-shear rate condition, τ^* is the model constant that show the critical shear stress, from which the pseudoplastic behaviour of the material starts, and n is the model constant which symbolizes the pseudoplastic behaviour slope of the material as so: (1-n).

2.8 Rabinowitsch Correction

Rabinowitsch are considered the non-Newtonian behavior where apparent shear rate ($\dot{\gamma}_a$) was converted into true shear rate ($\dot{\gamma}$) as in the equation below. (Asif et al, 2009) state that, Rabinowitsch correction is the most significant one.

$$\dot{\gamma} = \left[\frac{4n}{3n+1}\right]\dot{\gamma}_a \qquad (2.4)$$

2.9 Acrylonitrile Butadiene Styrene (ABS)

Figure 2.11 demonstrate the chemical structure of ABS while Figure 2.12 is the physical and mechanical properties of ABS plastic. ABS is an amorphous thermoplastic which consists of three constituent or monomer; Acrylonitrile, Butadiene, and Styrene. Asif (2009) stated that Butadiene rubber phase, both its content and morphology, play vital role in the rheology of ABS resin. ABS resin contains acrylonitrile-styrene copolymer, SAN, in matrix and grafted polybutadiene rubber in a dispersed phase Sohn (2007). The binding matrix layer of SAN makes the two phases of the polymer compatible Alfredo (2008). According to Anand (2021):