NUMERICAL STUDY ON THE EFFECTIVENESS OF HOMOGENEOUS MULTIPHASE VOLUME OF FLUID (VOF) MODEL IN RUBBER PROCESSING

LEE EYANN

UNIVERSITI SAINS MALAYSIA

2022

SCHOOL OF MATERIALS AND MINERAL RESOURCES ENGINEERING

UNIVERSITI SAINS MALAYSIA

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By

LEE EYANN

Supervisor: Ir. Dr. Muhammad Khalil

Dissertation submitted in partial fulfillment of the requirements for the degree of

Bachelor of Engineering with Honours

(Polymer Engineering)

Universiti Sains Malaysia

August 2022

DECLARATION

I hereby declare that I have conducted, completed the research work and written the dissertation entitled ‘Numerical Study on The Effectiveness of Homogeneous Multiphase Volume of Fluid (VOF) Model In Rubber Processing’. 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.

Name of Student: Lee Eyann Signature:
Date: 10 AUGUST 2022

Witness by
Supervisor: Ir. Dr. Muhammad Khalil Signature:
Date: 10 AUGUST 2022

ACKNOWLEDGEMENT

First and foremost, I would like to express my gratitude of University Sains Malaysia and School of Materials and Mineral Resources for providing me computers and workplace to complete this thesis. Next, I would like to thank my supervisor Ir. Dr. Muhammad Khalil Abdullah who has guided me in completing this project. Thanks to his patience, motivation, advice and knowledge, I was able to complete the research work and thesis within the given period. In this aspect, I am eternally grateful to you.

Besides my supervisor, I would like to thanks to Prof. Dr. Zulkifli Mohamad Arif for allowing me to use the data from his research to accomplish this thesis. Also, I would like to express my gratitude to Puan Hasnah who have me in solving the problems arise from software and computer.

Lastly, I would like to thank my family, my beloved parent Lee Seng Joo and Leow Kim Har for providing support and encouragement to me during this period. I would also like to express my thanks all my friends who keep support and provide helps to me.

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LIST OF SYMBOLS

τ^*	Critical shear stress
°	Degree
°C	Degree Celsius
P	Density
F	Fraction of cell’s volume occupied by liquid
η_∞	Infinite shear rate viscosity
J	Joules
K	Kelvin
Kg	Kilogram
m^3	Metre cube
mm	Millimeter
mN	millinewton
s	Second
$\dot{\gamma}$	Shear rate
a_T	Shift factor
C_p	Specific heat capacity
Pa	Pascal
n	Power law index
T	Temperature
k	Thermal conductivity
t	Time
λ	Time constant
u	Velocity at x direction
v	Velocity at y direction
w	Velocity at z direction

η	Viscosity
Φ	Volume Fraction
W	Watt
η_0	Zero-shear rate viscosity

LIST OF ABBREVIATIONS

ADS	Air dried sheet
CFD	Computational fluid dynamics
CV	Conventional vulcanization
DPM	Discrete phase model
EBS	Estate brown crepe
EV	Efficient vulcanization
FVM	Finite volume method
PLC	Pale latex crepe
RSS	Ribbed smoked sheet
SBR	Styrene butadiene rubber
TSR	Technically specified rubber
VOF	Volume of fluid
WLF	Williams-Landel-Ferry

NUMERICAL STUDY OF THE EFFECTIVENESS OF HOMOGENEOUS MULTIPHASE VOLUME OF FLUID (VOF) MODEL IN RUBBER PROCESSING

ABSTRAK

Simulasi berangka telah digunakan secara meluas dalam simulasi pemprosesan polimer. Penggunaan simulasi dalam pengacuan suntikan mampu mengesan potensi kecacatan yang boleh membantu mengurangkan kos dan masa yang terbuang. Tujuan tesis ini adalah untuk mengkaji keberkesanan model berbilang fasa homogen “Volume of Fluid (VOF)” dalam pemprosesan getah. Untuk menjalankan simulasi, formula penjanaan, model Cross dan Carreau digunakan. Model acuan diambil daripada kajian penyelidikan terdahulu. Pengesahan Ansys Fluent akan dijalankan terlebih dahulu dengan membandingkan hasil eksperimen dan simulasi daripada penyelidikan itu. Kemudian, tingkah laku pengisian acuan dan keberkesanan model reologi akan dinilai. Acuan akan diubah suai untuk menyiasat kelakuan pengisian acuan getah asli dalam acuan dengan geometri yang berbeza. Ansys Fluent menunjukkan persetujuan yang baik dengan keputusan eksperimen daripada penyelidikan itu dan model Cross menunjukkan ketepatan yang lebih baik. Getah asli akan cenderung mengalir ke dalam rongga dengan ketebalan yang lebih besar dan kadar aliran getah asli dipengaruhi oleh kelajuan masuk atau kelajuan suntikan. Tambahan perangkap cair mampu memerangkap kelikatan cair yang tinggi di hadapan aliran. Kehadiran halangan dalam acuan akan menjejaskan pembentukan kecacatan garis aliran dan mungkin menyebabkan pengisian yang tidak lengkap. Kesimpulannya, penggunaan model VOF dengan model Cross dalam Ansys Fluent merupakan model yang sesuai untuk meramal tingkah laku aliran getah asli.

NUMERICAL STUDY OF THE EFFECTIVENESS OF HOMOGENEOUS MULTIPHASE VOLUME OF FLUID (VOF) MODEL IN RUBBER PROCESSING

ABSTRACT

Numerical simulation has been widely used in simulation of polymer processing. The use of simulation in injection moulding was able to detect potential defects which could help to reduce the cost and time wasted. The purpose of this thesis was to study the effectiveness of homogeneous multiphase Volume of Fluid (VOF) model in rubber processing. To run the simulation, the governing equations, Cross and Carreau models were used. The mould model was taken from a established research paper. Validation of Ansys Fluent was first conducted by comparing the experimental and simulation results from the research paper. Then, the mould filling behaviour and effectiveness of rheology models were evaluated. Next, the mould was modified to investigate the mould filling behaviour of natural rubber in mould with different geometry such as addition of melt traps and implementation of obstacles. Ansys Fluent showed good agreement with the experimental results from the research paper and the Cross model showed better accuracy than the Carreau model. During mould filling, the natural rubber tended to flow into cavity with larger thickness and the rate of flow of natural rubber was affected by the inlet speed or injection speed. Besides that, the implementation of melt trap was able to trap the high viscosity melt front of the flow. Moreover, the presence of obstacles in the mould affected the formation of flow line defects and maybe caused short shot. In conclusion, the use of VOF model with Cross model in Ansys Fluent was a suitable model to predict the flow behaviour of natural rubber.

CHAPTER 1

INTRODUCTION

1.1 Research Background

Polymer is large macromolecule that made of many repeated subunits called monomers. The monomers are simple organic molecules (Mustafa *et al.*, 2016; Fakirov, 2017). Polymer can be classified into thermoplastic, thermoset, and elastomer (Zainudin *et al.*, 2020; Singh *et al.*, 2015). Elastomer is able to undergo large and reversible deformation because the coiled molecular chains are held together by crosslinking. This crosslinking can hinder the chains from sliding past each other (Osswald, 2017). Natural rubber is one of the elastomers (Sastri, 2010).

Natural rubber is produced from the latex of the Hevea Brasiliensis (Sisanth *et al.*, 2017). The natural rubber has a structure of cis-1,4-polyisoprene as shown in Figure 1.1 (Surya *et al.*, 2018). Natural rubber has been widely used in various fields such as military, medical, industrial, and agricultural fields due to its unique physical and chemical properties such as high tensile strength, resilience, toughness, and good processing characteristics (Zheng *et al.*, 2021; Sasitaran *et al.*, 2017). Hence, the natural rubber is suitable to be manufactured into various products such as tyres, conveyor belts, hoses, and adhesives (Bokobza and Banks, 2017). However, due to the existence of the double bonds and allylic hydrogen in the molecular structure, the natural rubber can be degraded by heat, oxygen, light, and ozone (Zheng *et al.*, 2021; Sasitaran *et al.*, 2017).

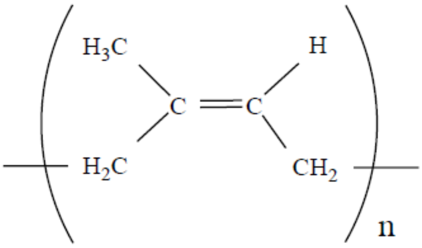

Figure 1.1 Structure of cis-1,4-polyisoprene (Surya *et al.*, 2018)

Generally, compression moulding is used for production of rubber product in large amount (Izamshah *et al.*, 2020). This is because the operation of compression moulding is simple and the cost for construction of mould is low (Tatara, 2017). However, in recent years, the injection moulding of rubber has become a desired method to manufacture several products such as sealings, housing, and dampings due to short processing times (Traintinger *et al.*, 2021). The injection moulding of rubber is ideal for production of small or complicated products. As compared to compression moulding, it can improve the uniformity of product, allow more versatility of shape and able to reduce material cost (Khang and Ariff, 2014). For rubber injection moulding, the mould is used to heat the rubber compound to promote vulcanization instead of cooling for solidification (Manas *et al.*, 2011; Skrobak *et al.*, 2018).

Nowadays, computer simulation has been widely used to simulate polymer processing include injection moulding process. Through simulation, potential defects such as air trap, weld line and sink mark can be predicted (Tran and Gehde, 2019). As the defects are predicted before the run begin, both consumption on cost and time can be reduced in term of design and production (Liu and Gehde, 2015; Amran *et al.*, 2016). One of the methods to analyse the complex transport phenomena is through the use of computational fluid dynamics (CFD) (Tekasakul *et al.*, 2015). The use of CFD allow the properties of fluid, operating conditions, and geometric characteristics to be adjusted in a flexible but defined way (Ahsan and Hussain, 2017).

With the use of the finite volume method (FVM), partial differential equations that express conservation laws over differential volumes can be converted into discrete algebraic equations over finite volumes. Because it can transform the conservation equations into face fluxes and be assessed at the finite volume faces, it is a favoured model in CFD. On unstructured polygonal meshes, the FVM can also be formulated in physical space. Due to these qualities, FVM is an effective technique for simulating fluid flow (Moukalled *et al.*, 2016).

A multiphase system or models consists of materials of two or more phases (Kreith, 2017). There are primary and secondary phases in the flow of multiphase. The primary phase is continuous while the secondary phases are dispersed within the continuous phase (Jadidi, 2017). The multiphase flow in CFD is considered as an Eulerian reference. The dispersed phase can be treated with Lagrangian or Eulerian representations (Kreith, 2017). In the Eulerian representation or Euler-Euler approach, three multiphase models are available which are volume of fluid (VOF), mixture model and Eulerian model (Ansys Fluent, 2009).

The VOF model is able to describe the location and shape of the free surface accurately. The technique for computing the movement is simple (Katopodes, 2019). The moving interface is tracked and reconstructed using the VOF model, which can also serve as an indication that indicates how much fluid of each phase is contained in each of the grid cells (Chen *et al.,* 2015). Besides, in VOF, the volume fraction, ϕ is evolved for a certain fluid (fluid A) in any cell that houses the interface. Consequently, only the following values can be assigned to ϕ: 1 denotes that the cell only contains fluid A: 0 indicates that the cell contains the other fluid, while $0 < \phi < 1$ indicates that the cell contains both (Shonibare, 2017).

Several research on numerical investigation of polymer have been conducted. For example, Satin and Bílik (2016) has use Moldex3D to investigate the effect on viscosity on the mould filling behaviour of polymer. Manas *et al.* (2011) have also investigate the effectiveness of Cadmold rubber in predicting the flow of rubber in injection mouldings. Next, Mitsoulis *et al*. (2017), with the use of finite element method (FEM), is able to simulate the flow of polymer in nozzle of injection moulding and capillary rheometer. The use of different softwares and models have several effects on the calculated results. Also, the numerical simulation on mould filling behaviour of rubber is less. Hence, this thesis is conducted to study the effectiveness of homogeneous multiphase Volume of Fluid (VOF) model in rubber processing.

1.2 Problem Statement

Many studies have been conducted to investigate the effectiveness of numerical simulation in polymer processing. However, the accuracy of numerical simulation could be varied as it depends on the model chosen and parameters inserted. Different rheology model such as Cross and Carreau models and multiphase models will result in deviation between results due to different formulations and equations used. Some researchers have conduct numerical simulation on natural rubber to predict its cure behaviour and to validate the use of Computational Fluid Dynamics (CFD) software in predicting the flow behaviour of polymer. However, the research on mould filling behaviour and injection moulding of natural rubber is still lacking. Hence, in this thesis, the numerical simulation on mould filling behaviour of natural rubber is investigated. It is believed that this thesis is able to provide the fundamental ideas on numerical simulation on flow properties of natural rubber and injection moulding of natural rubber. It was also hoped that through this thesis, the mould filling behaviour of natural rubber could be further investigated and applied in industries.

1.3 Objectives of Launching Thesis Template

- To validate the effectiveness of homogeneous multiphase Volume of Fluid (VOF) model in rubber processing.

- To investigate the effectiveness of Cross and Carreau model in numerical simulation of rubber processing.

- To investigate the effect of certain parameters such as injection speed and melt trap on mould filling behaviour of natural rubber in numerical simulation.

- To investigate the effects of mould geometry to flow properties of natural rubber in numerical simulation.

1.4 Scopes of research

In this thesis, the numerical simulation on mould filling behaviour of natural rubber will be conducted by using Ansys Fluent. The VOF model are selected to conduct the numerical simulation with the use of Cross and Carreau model respectively. The obtained result will be compared with the results from from Khang and Ariff (2014). After that, Cross model will be used to run the remaining simulation to investigate the mould filling behaviour of natural rubber, effect of inlet velocity and geometry of mould to flow of natural rubber. Besides, the rheology model used in this thesis do not include the WLF-equation, hence the rheology properties are assumed to be affected by shear stress and shear rate only. Also, this thesis only focusses on the filling stage on injection moulding therefore, the curing or vulcanization of natural rubber are ignored.

1.5 Thesis Organization

In CHAPTER 1, an introduction on the tittle of thesis background will be provided. The fundamental of polymers and numerical simulations about the thesis will be discussed briefly. Besides, the problems to be investigated and the objectives of the thesis will be stated. Also, the scope of this research is reported.

In CHAPTER 2, the background of natural rubber including polymer and elastomer will be discussed. The unique properties of natural rubber and its difference with other classes of polymer are stated. Furthermore, the information of processing of natural rubber and concept of injection moulding are also reported. Next, the fundamental concept on numerical simulation and multiphase model are also stated in this chapter.

In CHAPTER 3, the equations used in VOF model, Cross model and Carreau model are stated. The discretization or meshing results for the moulds or models used will be listed in detail. Also, the boundary conditions which include the material properties of natural rubber, inlet velocity, wall temperature and melt temperature are stated.

In CHAPTER 4, the results from the validation on the use of Ansys Fluent and the difference between the Cross and Carreau model are discussed first. Next, the results obtain from mould filling behaviour of natural rubber such as filling rate, viscosity, flow rate and void content are discussed. Then, different inlet velocity is applied on the simulation to find out its effect on the mould filling behaviour of natural rubber. Lastly, the effect of mould with different geometry and modified runner on mould filling behaviour of natural rubber are discussed.

The CHAPTER 5 summarize all the finding in CHAPTER 4 and some comments for future work or research are provided.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

This chapter has briefly discussed the concept and theory behind natural rubber, injection moulding and numerical simulations. From the literature review, it was found out that the numerical simulation of polymer processing has been widely used in various fields due to cost and time saving. The use of simulation in polymer processing is able to predict various defects such as voids, sink marks, weld line and short shot, hence, parameters for the process can be optimized through simulation to prevent or reduce the occurrence of these defects. However, the accuracy of numerical simulation could be varied as it depends on the model and data inserted. In the past decade, various study on numerical simulation of polymer processing has been conducted by using different materials, software, and rheology models but the numerical simulation on mould filling behaviour of natural rubber is a few. Hence, in this thesis, numerical study on the effectiveness of homogeneous multiphase Volume of Fluid (VOF) model in rubber processing was conducted.

2.2 Polymer

The term polymer stems from the Greek roots poly which means many and meros which means part. Hence, this word means “many parts”. Polymers contains thousands to millions of atoms in a large molecule which is known as macromolecule. They are formed by joining numerous small and simple organic molecules called monomers (Fakirov, 2017). There are two types of polymers which are natural polymer and synthetic polymer. The process of formation of these polymers from monomers is polymerization. Due to large molecular mass relative to small molecules compound in these polymers, they exhibit unique physical properties such as toughness, viscoelasticity and ability to form glasses and semi crystalline structures rather than crystals (Mustafa *et al.*, 2016).

Natural polymers, such as proteins, cellulose, starch, resins, etc., are polymers derived naturally from sources in plants and animals. Natural polymers are superior to synthetic ones in a number of ways, including cost, availability, versatility, possibility for biodegradability, and compatibility. Natural polymers are also biogenic, making them ideal drug delivery materials due to their biological characteristics such cell recognition and interaction, enzymatic degradability, similarity to the extracellular matrix, and chemical flexibility. Natural polymers therefore have a lot of potential in the pharmaceutical, food, and cosmetic industries (Rajeswari *et al.*, 2021).

The synthetic polymers are manmade polymers and are usually derived from petroleum oil. They can be classified into thermoplastics and thermosets (Zainudin *et al.*, 2020). Thermoplastic can be melted and reshaped by applying heat and pressure, therefore, it can be recycled easily. While thermosets tend to decompose when heated (Charles and Carraher, 2017). This is because thermoset undergo chemical reaction when heated and result in an insoluble material that cannot be melted. One of the major advantages of synthetic polymers are their high chemical purity. Medical devices such as suture materials, medical implants, fracture fixation devices, and dialysis tubing make use of the biological inertness of synthetic polymers and reduce the host response to the material to a minimum level (Zainudin *et al.*, 2020). Other than thermoplastic and thermoset, there is still one type of polymer called elastomer (Singh *et al*., 2015; Peters, 2015).

2.3 Elastomer

Elastomer is a type of polymer which their chains are long, randomly coiled, curled, loosely crosslinked, and held together by weak attractive forces. Due to this unique property, the elastomer is said to have high degree of flexibility and mobility that allow it to be stretched easily and return to original shape when the applied force or stress is removed (Mustafa *et al.*, 2016; Sastri, 2010; Bokobza and Banks, 2017). During the deformation, the coiled and curled polymer chains are stretched. As the force continue to be applied, chains will be stretched fully until hindered by the crosslinks. During this, stage, if the force is removed, the chains will recoil and return to its original state, however, if further force is applied, breakage of the crosslinking and chains will occur and result in fracture of the component (Osswald, 2017). Even though the occurrence of crosslinking exist in thermoset also, but unlike elastomer which is ductile, the thermoset are hard, rigid and brittle (Sastri, 2022). Figure 2.1 shows an illustration of movement of the polymer chains when stretched. Example for elastomers include natural rubbers, styrene-butadiene block copolymers, polyisoprene, polybutadiene, ethylene propylene rubber, ethylene propylene diene rubber, silicone elastomers, fluoroelastomers, polyurethane elastomers, and nitrile rubbers Also elastomer can be manufactured into gaskets, seals, rubber septum, automobile tyres, conveyor belts, hoses, and adhesives (Sastri, 2010; Bokobza and Banks, 2017).

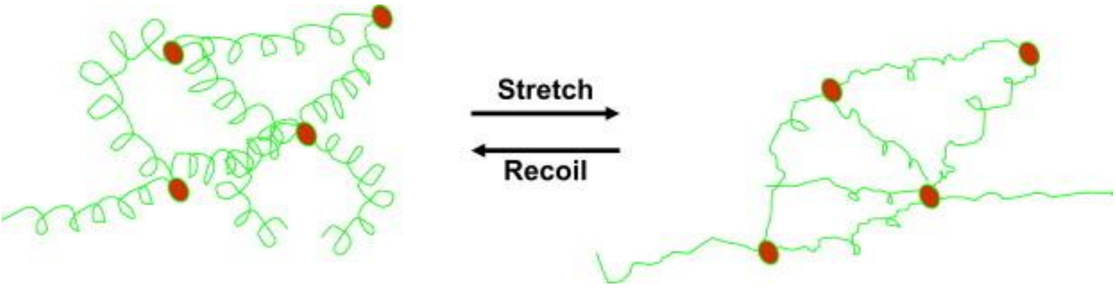

Figure 2.1 Movement of polymer chains under deformation (Sastri, 2010)

2.3.1 Natural Rubber

One of the elastomers, natural rubber is produced from the latex of Hevea Brasiliensis tree (Surya *et al.*, 2018). The latex is an aqueous colloidal dispersion of rubber particles, and it is extracted through the tapping process. Fresh latex coagulates spontaneously within few hours, but this can be prevented by adding anticoagulants such as ammonia, or formalin (Sisanth *et al.*, 2017). Through a step known as primary process, the latex can be converted into natural rubber which is more suitable to be stored and preserved. These rubbers are known as ribbed smoked sheet (RSS), air dried sheet (ADS), technically specified rubber (TSR), pale latex crepe (PLC) and estate brown crepe (EBC) (Kurian and Mathew, 2011).

Natural rubber has a structure of cis-1,4 polyisoprene as shown in Figure 2.2 (Surya *et al.*, 2018). Natural rubber has an average molecular weight ranging from 100, 000 to 1,000,000. The higher the molecular weight, the higher the viscosity. During storage, the viscosity of natural rubber will be increased, and it is dependent on the time of storage. The increase in viscosity is due to occurrence of crosslinking between the rubber molecules with the amino acids and protein. Furthermore, due to high structural regularity of the structure, natural rubber is capable to crystallise spontaneously when stored at low temperature or stretched (Kurian and Mathew, 2011). Furthermore, natural rubber undergoes strain-induced crystallization that result in significant increase in modulus under high deformation, this phenomenon is due to formation of crystallites that act as additional crosslinks in the network (Bokobza and Banks, 2017).

Other than excellent tensile strength, natural rubber also various unique properties such as good abrasion resistance, resilience, toughness, good process characteristics and good dynamics properties (Izamshah *et al.*, 2020; Zhao *et al.*, 2019; Sasitaran *et al.*, 2017). Due to its unique properties, natural rubber is an ideal material for many applications. It has been received great attention in the military, medical, industrial, and agricultural fields (Li *et al.*, 2015; Zheng *et al.*, 2021). The applications of natural rubber in industrial and engineering are various such as hoses, sealant, seals, O-rings, safety boots, tires, and mounting materials (Surya *et al.*, 2018). Generally, rubber-based products can be manufactured through various processes such as extrusion, compression moulding and calendaring (Izamshah *et al.*, 2020).

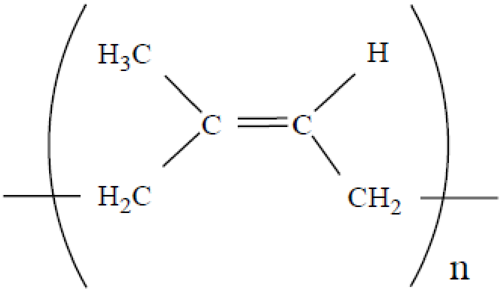

Figure 2.2 Structure of natural rubber (Surya *et al.*, 2018)

However, there are still some limitations on natural rubber such as low hardness, and low resistance to oil. Besides, due to the existence of unstable double bonds and allylic hydrogen in the molecular structure of natural rubber, it can be degraded easily by exposing to heat, light, oxygen and ozone (Sasitaran *et al.*, 2017; Zheng *et al.*, 2021).

Next, the properties of natural rubber can be altered through a process known as compounding in conducted on rubber which various ingredients are added to the rubber matric. The reasons for using compounding include to satisfy the end properties, to achieve better processing requirements and to maintain a balance between the properties and price (Sisanth *et al.*, 2017). Generally, ingredients such as vulcanizing agents, activators, accelerators, antioxidants, and fillers are used in compounding of natural rubber to modify its properties for specific applications (Ngamsurat *et al.*, 2011; Zhao *et al.*, 2019).

Besides, natural rubber can be blended with synthetic rubber to overcome its limitation and production of static and dynamic applications such as tires, vibration dampers, seals, isolators, and shock mounts (Dick *et al.*, 2001). Blending is a process of mixing two or more polymer together to produce a blend with desired characteristics without the combination of particular polymer network (Reddy *et al.*, 2015). An example of natural rubber blend is blending of natural rubber with styrene butadiene rubber (SBR) to improve its processing characteristics and reduce the cost of final products. Through blending, the resistance to abrasion, resilience, flexibility at low temperature and resistance to groove cracking of natural rubber can be improved (Moustafa *et al.*, 2016).

The distinction that made natural rubber differ with thermoplastic is the occurrence of vulcanization. Vulcanization is a process generally applied to rubbery or elastomeric materials. During vulcanization, rubber change from the linear structure of the macromolecules crosslinking to a three-dimensional network structure of macromolecules that improve the physical and mechanical properties of rubber significantly. Thus, vulcanization increase the elasticity of natural rubber but decrease its plasticity. During moulding, vulcanization of natural rubber occurs under high temperature and pressure (Zhang *et al.*, 2013; Mark *et al.*, 2013).

The vulcanization process consists of three stages as shown in Figure 2.3, during the first stage which is called as induction period or time, slow chemical reactions occur between the rubber and the additives. This stage is best for processing as it provide good flow for the rubber compound to fill the mould cavity. The second stage begin at the end of induction. During this stage curing occur where the crosslinking form between the rubber molecular chains to produce a network structure as shown in Figure 2.4. Three results can occur at the last stage, and it is dependent on the vulcanization agent and temperature applied. Plateau is an ideal case where equilibrium degree of vulcanization is achieved. However, reversion might occur when the rubber compound is overheated. This will cause the breakdown of crosslinks occurs faster than their formation. In some cases, marching will occur which is a result of formation of crosslinks (Khimi and Pickering, 2014; Isayev, 1989).

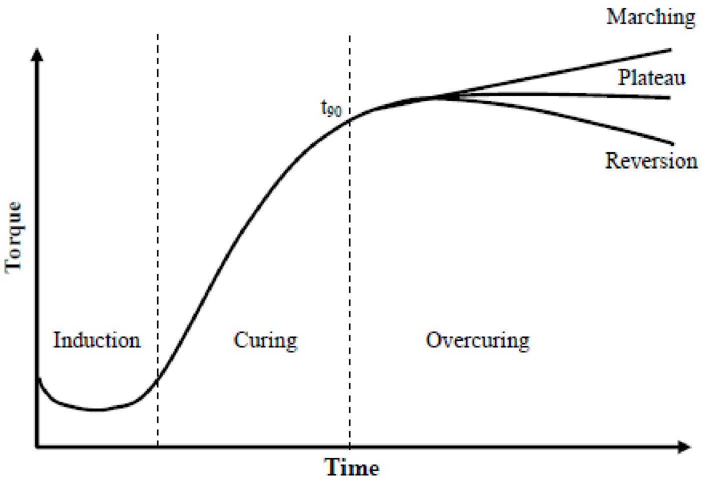

Figure 2.3 Vulcanization curve of rubber (Khimi and Pickering, 2014)

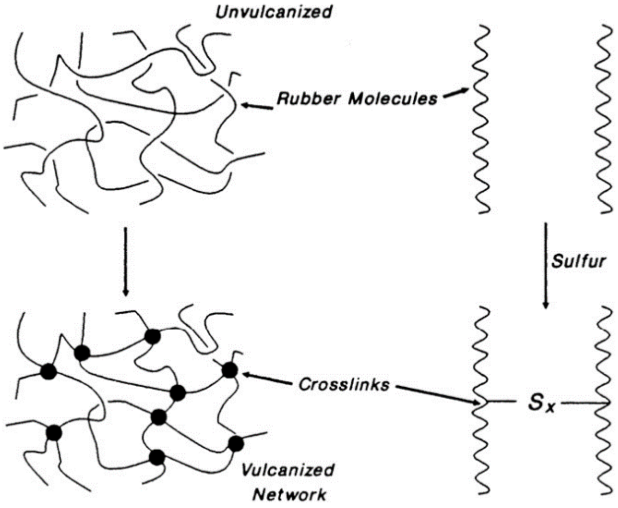

Figure 2.4 Formation of vulcanization (Mark *et al.*, 2013)

There are three types of sulphur vulcanization system available for rubber and it is dependent on the ratio of incorporate accelerator to sulphur. With a ratio of 0.1 – 0.6, conventional vulcanization (CV) system is obtained, with a ratio of 0.7 – 2.5, semi-efficient vulcanization (semi-EV) system is obtained and with ratio of 2.5 – 12, efficient vulcanization (EV) system is achieved. The number of crosslinking formed between the rubber chains are affected by the amount of sulphur used and it will affect the curing properties, mechanical properties thermal stability and aging properties of the rubber compound. The CV system provide higher crosslink density to the rubber compound, and it has more polysulphides crosslinks as compared to monosulphides or disulphides as a result, it has higher mechanical properties and resistance to chemical, but it has poor thermal stability. Conversely, the EV system which has lower crosslinks density and contains monosulphides or disulphides are more resistance to fatigue, oxidation, and aging, but it has lower mechanical properties. While the semi-EV system has moderate fatigue life and mechanical properties (Kurian and Mathew, 2011; Mark *et al.*, 2013; Lee *et al.*, 2020).

2.3.2 Processing of Natural Rubber

For economical purposes, the mass production of rubber products is usually done by compression moulding (Izamshah *et al.*, 2020). During rubber compression moulding, a preheated rubber is first placed in a heated mould cavity, then the mould is closed, and pressure is applied on it to force the rubber to take the shape of the cavity as shown in Figure 2.5. Heat and pressure are applied continuously until the rubber has cured (Ramorino *et al.*, 2020). The low mould construction cost with simple operation step has allow compression moulding to manufacture products with different size and shape, as a result it is often chosen for mass production of medium hardness compounds or application that required expensive material. Compression moulding require low maintenance and operating costs because it only has less movement on machine and mould components as well as little temperature zones. The short distance for material flow in the mould cause the polymer chains does not have sufficient time to orient themselves, as a result, the product has uniform part properties and density in all three dimensions as well as fewer residual stresses (Ramorino *et al.*, 2020; Tatara, 2017).

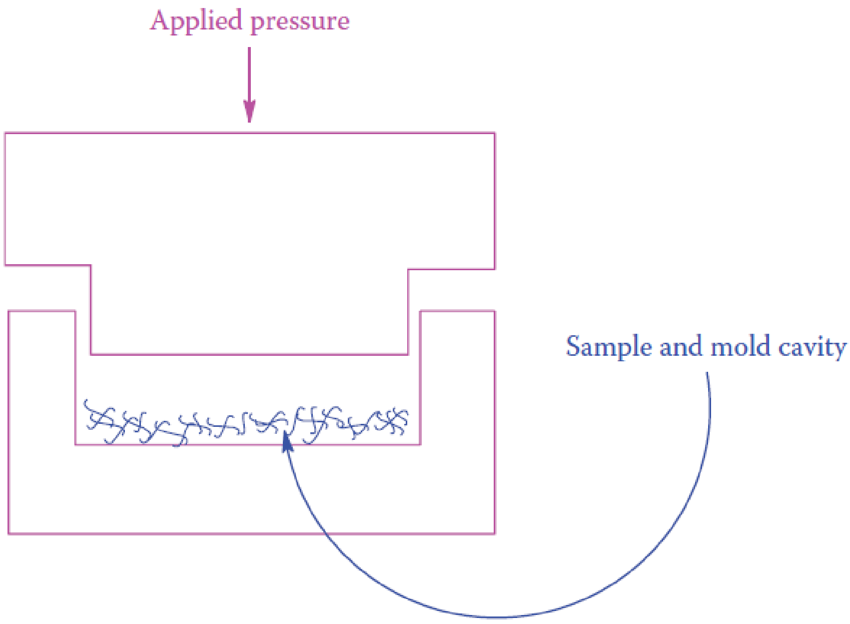

Figure 2.5 Compression Moulding (Charles and Carraher, 2017)

One of the disadvantages of this process is part inconsistency that is related to the process parameter. Moreover, compression moulding of natural rubber will lead to part distortion and shrinkage easily. This is due to the flexibility of natural rubber, and it is affected by temperature. Hence, distortion might occur when removed from the mould through stretching it over a core (Izamshah *et al*., 2020).

As compared to compression moulding, injection moulding has become a good manufacturing method to produce small and complicate parts that required precision and high productivity such as sealing, housing, and damping applications in shorter processing time (Traintinger *et al.*, 2021; Khang and Ariff, 2012). Injection moulding of rubber is an automatic process that feed heated or plasticized rubber stock into the injection chamber at temperature under the vulcanization temperature. The material is injected through a nozzle, sprue, runner, and gate into a cavity having a wall temperature that is high enough to initiate the vulcanization process. After the vulcanization process is completed inside the cavity, the moulded part will be ejected (Isayev *et al.*, 1988).

Normally, rubber injection moulding is used for high production runs and parts that require tighter tolerances as it is able to produce part with high dimensional accuracy, low shrinkage and warpage, excellent surface structure and strength properties. With the use of some type of moulds, finishing steps on parts can be prevented or reduced also (Baum and Anders, 2021; Ramorino *et al.*, 2020). But the process of rubber injection moulding is quite sensitive, and its success rate is dependent on the properties of rubber compound and its parameters. One of the challenges in rubber injection moulding is the flow behaviour of rubber compound. Different from thermoplastic that can be injected at melt temperature to obtain required flow properties, rubber compound can only be injected at temperature between 70-110°C. A higher temperature will lead to premature start of the curing process. In this phase, the rubber compound will exhibit high viscosity and provide more resistance on flow (Traintinger *et al.*, 2021).

2.4 Injection Moulding

In term of thermoplastic, the injection moulding is a two-step cyclical process. The first step is melting of material by screw rotation and the second step is filling the molten material into the mould by forward ramming of the screw, followed by a short packing stage to compensate the shrinkage after cooling and solidification (Polychronopoulos and Vlachopoulos, 2019). As shown in Figure 2.6, the injection moulding machine can be separated into two units which are the plasticizing unit and the clamping unit. In the plasticizing unit, the polymer is fed, melted, homogenized and metered up to the required quantity in this unit. Then the melt is injected through a nozzle at high pressure into the cavity of mould that located at the clamping unit. After the cavity is filled, the melt is compressed, cooled and the volume shrinkage is partly compensated in the holding pressure stage. After, solidification, the mould will be opened and the part is ejected from the mould (Baum and Anders, 2021; Baum *et al.*, 2022).

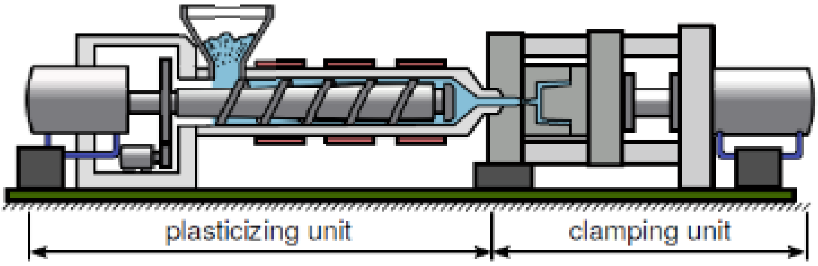

Figure 2.6 Injection moulding machine (Baum and Anders, 2021).

The injection moulding of rubber is almost same as the thermoplastic. Figure 2.7 shows the rubber injection moulding cycle. Same as thermoplastic, the rubber compound is fed, melted, and homogenized inside the plasticizing cylinder or plasticizing unit. Then, the mould is closed, and the melt is injected into the cavity of mould under pressure. Inside the mould, the melt is heated to promote vulcanization instead of cooled (Manas *et al.*, 2011). Besides, the rubber compound also required pre-heating before feed into the injection moulding machine to reduce the vulcanization period. To prevent premature of curing, the compound cannot be heated above 100 °C (Skrobak *et al.*, 2018).

The mould is the most crucial element in an injection moulding. The mould not only determines the shaped of the moulded part, but it also conducts the hot plasticized material from the plasticizing unit to the cavity, vents the trapped air, solidified the part and eject the part (Chanda, 2018). However, the flow of polymer melt in the mould cavity is complex as the flow behaviour of polymer is affected by flow rate, viscosity, melt temperature and wall thickness of mould (Yizong *et al.*, 2017).

Figure 2.7 Rubber injection moulding cycle (Manas *et al.*, 2011)

In order to achieve optimum result of injection moulding process, the parameters, part properties, mould design as well as material properties required to be considered (Baum *et al.*, 2022). One of the problems in injection moulding is entrapment of air. The present of air inside the mould cavity will lead to incomplete mould filling, voids, short shot or burned marks (Miranda and Nogueira, 2019). Hence, to avoid the present of such defects, venting could be applied at area where the mould fills last to release the entrapped air inside the cavity (Abdullahi *et al.*, 2016; Zhang *et al.*, 2018).

In addition, in rubber injection moulding, the injection rate needs to be optimized to prevent premature curing and to reduce cycle time. Low injection rate will cause short shot and surface defects due to premature curing. Besides, the packing pressure need to be optimized to prevent occurrence of flash and to decrease shrinkage of part. The holding pressure also play an important role in affecting the injection moulding process. The holding pressure is important in preventing the backflow of material, but it will cause flash. High holding pressure also able to increase the part weight and density (Bont *et al.*, 2021).

2.5 Rheological Properties of Polymer

The rheology properties of material affect the shape and flow of the material significantly which is also known as flow properties. Generally, the flow properties is represented by a flow curve which express the ratio of shear stress and shear rate. The flow properties of a fluid can be classified into two classes which are Newtonian fluid, and non-Newtonian fluid. Liquid with low molecular weight is categorized as Newtonian fluid. These fluids obey the Newton's law which the viscosity of fluid is independent of time, and speed of shearing but depends on the temperature, pressure, and molecular properties of the material. Conversely, fluid with high molecular weight is categorized into non-Newtonian fluid. The viscosity of these fluids is not constant and depend on the strain rate (Satin and Bílik, 2016).

Figure 2.8 shows the flow curve of Newtonian fluid and non-Newtonian fluid. The flow curve of Newtonian fluid always shows a straight line that indicate that the shear stress is directly proportional to the shear rate. Any point on the line is enough to define the viscosity of the fluid (Zuzanna, 2016). Besides, the Newtonian fluid also show constant viscosity along any shear rate. Hence, viscosity of Newtonian fluid is independent of any fluid deformation and also do not exhibit elasticity properties (Escudier, 2017). The non-Newtonian fluids can be classified into time independent and time dependent fluid. Time independent fluid include shear thinning fluids and shear thickening fluids. Shear thinning fluids also known as pseudo-plastic fluid which its viscosity will decrease with increasing the shear rate while shear thickening fluid or dilatant fluids shows an increase in viscosity with the increase in viscosity (Barman *et al.*, 2016).

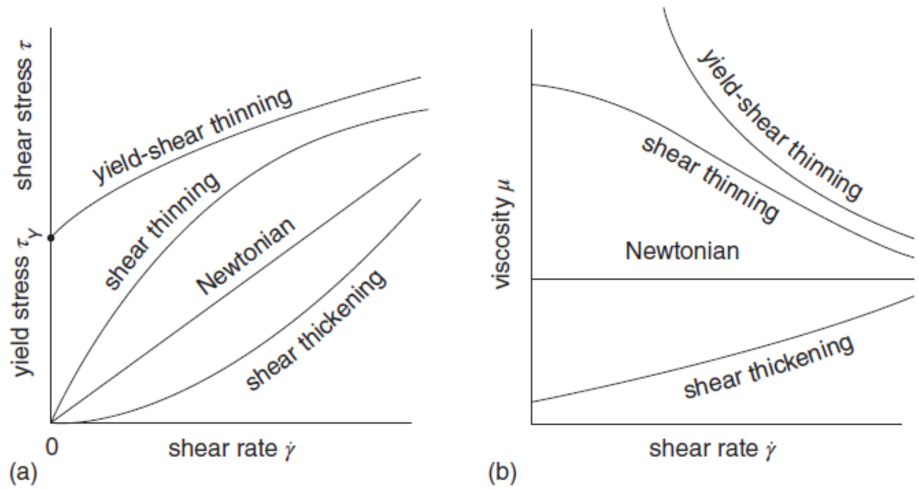

Figure 2.8 Flow curve of Newtonian and non-Newtonian fluid (Escudier, 2017).

The rheology properties of polymer melt are strongly determined by their molecular structure. Structure such as long chain branches has more effect on the rheological properties of polymer melt as compared to short chain branches (Münstedt, 2021). During deformation, as the shear rate increase, the molecular chains will be disentangled and aligned, as result, less resistance on flow and viscosity decrease (Polychronopoulos and Vlachopoulos, 2019). Therefore, polymer melt can be classified as non-Newtonian fluid (Zhang and Chen, 2020).

In order to study the rheology behaviour of fluids, several models have been developed such as Carreau, Ellis and power-law models that describe time independent rheology, Herschel-Bulkley model that includes yield stress and upper-convected Maxwell model and Oldroyd-B that include viscoelastic properties (Zamani *et al.*, 2017). For inelastic non-Newtonian liquids, power law model which consist of only two parameter is widely used. This model provides the slope shear stress and shear rate curve for modest range of shear range(Hina *et al.*, 2020). The power law index, *n* is able to determine the fluid behaviour of the fluid. For power law index of $0 < n < 1$, the fluid behaves as pseudoplastic, for $n > 1$, the fluid behaves as dilatant, and for $n = 1$, the fluid is a Newtonian fluid (Khan *et al.*, 2018). However, the power law model is not suitable to use on fluids that exhibit zero shear viscosity and infinite shear viscosity. Hence, the power law model is only able to describe the range of shear rate at the Newtonian plateaus (Schneider and Gerber, 2020). Hence, the Cross and Carreau model are used to correct the shortness of power law model as they are able to cover the entire range of shear rate (Hina *et al.*, 2020; Schneider and Gerber, 2020).

The Carreau and Cross models are based on the power law model, but with two low- and high-shear Newtonian plateaus to limit the shear rate dependence in order to match experimental observations of real polymeric liquids and to take advantage of the illogical behaviour that the power law model predicts at very low and very high shear rates (Sochi, 2015). The Carreau model is more useful in describing the flow behaviour of fluids at high shear rate (Khan *et al.*, 2018). Even thought, the Carreau model is able to predict the shear thinning behaviour of polymer, but it fails to predict the onset rate and shear thickening regime of viscoelastic polymer. It also under predicts the apparent viscosity of viscoelastic polymers by a considerable margin (Azad and Trivedi, 2019).

The Cross model not only able to describe the flow of fluid in the power law region but also region where shear rates are very high and low (Khan *et al.*, 2017). As compared to the power law model which describe the behaviour of fluid at limited range of shear rate, the Cross model is able to characterize the more complex behaviour of non-Newtonian fluids. Even though the Cross model characterize the behaviour of fluid same as the power law model over a range of shear rates, but it shows difference results at very low and very high shear rate as shown in Figure 2.9. As a result, Cross model is more preferred when modelling the flow of fluid especially in a 3D system (Hauswirth *et al.*, 2020).

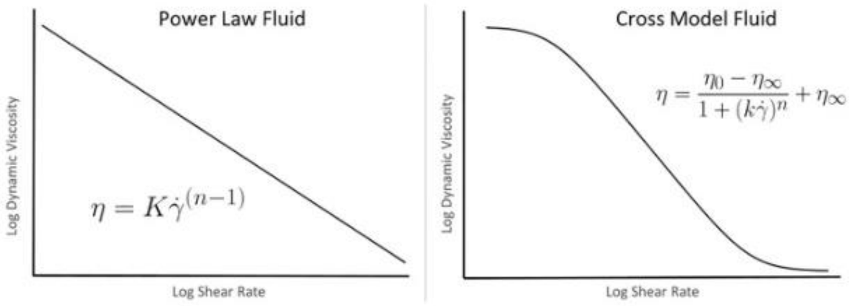

Figure 2.9 Comparison of power law model to cross model (Hauswirth *et al.*, 2020)

Polymer is also a unique material that can behave as liquid or solid under specific temperature and molecular weight. This behaviour is dependent on the rate of deformation and is called as viscoelastic behaviour (Osswald, 2017). Viscoelastic indicate the present of viscous and elastic properties together in a material. Viscous properties are related to the irreversible deformation. While the elastic properties are related to reversible deformation. The amount of elastic or viscous in a material is dependent on the deformation time. A rapid deformation will lead to greater elasticity, conversely, a slower deformation will lead to greater viscosity (Zuzanna, 2016). Under viscous state, the viscosity polymer is affected by shear rate and behave as non-Newtonian liquid. While, when under elastic state, the polymer behaves as a rubber-like solid. This phenomenon causes them has unique flow properties and have significant impact on processing (Polychronopoulos and Vlachopoulos, 2019). However, this viscoelastic behaviour of polymer is usually neglected when running flow simulation. Instead, the Newtonian fluid flow models are used (Friesenbichler *et al.*, 2020).

For injection moulding, the rheology behaviour of a polymer melt in a closed cavity is complicated as the flow behaviour of the polymer is contributed by the flow rate, viscosity, melt temperature and wall thickness. All these factors have effect on each other. For an example, under high flow rate, more friction will be generated between the melt and wall of cavity, as a result more heat provided to the melt and reducing its viscosity (Yizong *et al.*, 2017). Furthermore, the rheological properties of polymer melt are very important in processing especially in injection moulding of rubber as shear rate, pressure and temperate are involved during the process. In rubber industry, uniformity and consistency in the flow behaviour of rubber are important to improve the productivity, quality of product and energy saving (Rahim *et al.*, 2016). The processability of rubber are strongly dependent on the cure characteristics and rheology properties such as cure time, scorch time and viscosity (Sisanth *et al.*, 2017).

2.5.1 Carreau Model

According to Osswald and Rudolph (2015), the Carreau model consists of five parameter and is developed to observe the Newtonian plateau and available for a wide range of strain rates.

$$\frac{\eta_{\dot{\gamma}} - \eta_{\infty}}{\eta_0 - \eta_{\infty}} = (1 + |\lambda\dot{\gamma}\,|^{a})^{\frac{(n-1)}{a}} \tag{2.1}$$

where η_0 is the zero-shear rate viscosity, η_∞ is an infinite shear rate viscosity of the second Newtonian plateau, λ is a time constant and *n* is the Power Law index, which accounts for the shear thinning behaviour. The parameter a accounts for the width of the transition region between the zero shear viscosity and the Power Law region.

Normally, the infinite shear rate is negligible, and the equation become:

$$\eta(\dot{\gamma}\,) = \frac{\eta_0}{(1 + |\lambda\dot{\gamma}\,|^{a})^{\frac{(n-1)}{a}}} \tag{2.2}$$

The equation can be further modified to include a temperature dependence using a WLF relation. The modified model is often used in commercial polymer data base:

$$\eta = \frac{k_1 \cdot a_T}{(1 + k_2 \dot{\gamma}\ a_T)^{K_3}} \tag{2.3}$$

where, a_T is the shift factor:

$$log\ a_T = \frac{8.86(k_4 - k_5)}{101.6 + k_4 - k_5} - \frac{8.86(T - k_5)}{101.6 + T - k_5} \tag{2.4}$$

where k_1 in term of viscosity, k_2 in term of second, k_3 unitless, k_4 in term of temperature and k_5 in term of temperature are the constant for Carreau-WLF equation.

2.5.2 Cross Model

Osswald and Rudolph (2015) have also stated that the Cross model is a 6-parameter model that consider the effects of shear rate and temperature on the viscosity. Similar to the Carreau model, this model describes both Newtonian and shear thinning behaviour of a fluid. The shear thinning part is modelled by the general Cross equation:

$$\frac{\eta_{\dot{\gamma}} - \eta_\infty}{\eta_0 - \eta_\infty} = \frac{1}{1 + (\lambda\ \cdot \dot{\gamma}\ )^{1-n}} \tag{2.5}$$

Where, η_0 is the zero-shear rate viscosity, η_∞ is an infinite shear rate viscosity, λ is a time constant and n is the Power Law index, which accounts for the shear thinning behaviour. When $\eta_{\dot{\gamma}} << \eta_0$ and $\eta_{\dot{\gamma}} >> \eta_\infty$, the Cross model will be reduced to the Power Law model.

When the infinite shear rate viscosity is negligible, the Cross model become:

$$\eta(\dot{\gamma}\ ) = \frac{\eta_0}{1 + \left(\frac{\eta_0 \dot{\gamma}}{\tau^*}\right)^{1-n}} \tag{2.6}$$

Where, τ^* is the critical shear stress at the transition from the Newtonian plateau with $\lambda = \eta_0 / \tau^*$. If this model is used to fit the data prior making the Weissenberg-Rabinowitsch correction, the apparent shear stress can be shifted by letting