## 1. INTRODUCTION

1.1 Historical notes of melt spinning process

In many polymer processing operations, molten polymers emerge from dies into a stress field which deforms the melt into a final fabricated shape. This is the case in melt spinning of fiber and in film extrusion. The melt spinning process involves preparation of a spinning fluid, extrusion of the melt through spinneret into a cooling atmosphere, and elongation of the extruded melt. The resulting filaments are then subjected to drawing and anealing. Figure 1.1 shows a schematic diagram of these melt spinning processes. We have obtained the fine and strong filament by these processes in melt spinning.

The process of melt spinning dates from the pioneering efforts of Carothers and Hill<sup>1</sup> reported in 1932. It has been an important manufacturing process, and a large fraction of all synthetic fiber is produced via the melt spinning route.

At present time the melt spinning process is applied to the method of fiber formation from liquid-crystalline material,<sup>2,3</sup> the fabrication method of ultrafine fibers by using of composite spinning,<sup>4</sup> high speed melt spinning method,<sup>5</sup> and so on. The filaments with many useful properties have been developed by these new methods.

The properties of filaments are highly dependent on

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the spinning conditions used in their production. This behavior is due to the effect of process variables on the structure of the spun filament. The formation of structure during melt spinning is complicated by the combined influences of melt structure, rheological factors and non-isothermal effects. Because of these complications, early work was little fundamental study of the rheological behavior untill the work of Ziabick and Kedzierska<sup>6</sup> in 1959-1962.



Figure 1.1 Scheme of melt spinning

After their work, many studies on the elongational region in Figure 1.1 have been carried out; heat transfer of running filament,<sup>7-9</sup> kinematics and dynamics of deformation of the spinline, 10-15 molecular orientation

accompanying fiber spinning,  $^{16-20}$  crystallization behavior of running filament  $^{21-24}$  have been studied. The results of these works have made possible the formulation of a system of mathematical equation describing the melt spinning process as a whole. These studies were reviewed in the book of Ziabicki.<sup>25</sup>

However, there are several unresolved important problems. First, in spite of non-Newtonian viscosity of polymer melt having been known in the study of shear flow, the running filament in melt spinning has been assumed to be Newtonian fluid: three times the zero shear viscosity have been used as the viscosity of running filament. Considerably less effort has so far been invested in studying the nonnewtonian viscosity of elongational flow in melt spinning, although this is of considerable significance. Another problem is the quantitative estimation of crystallization progress in running filament by using of the data of static isothermal crystallization experiments. A third problem is associated with the effects of prehistory in preparation on elongational flow and crystallization behavior.

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## 1.2 Elongational flow

Elongation is the dominant mode of deformation in many important polymer processes. The importance of such flow in processing applications has been indicated by many authors including Cogswell and Lamb,<sup>26</sup> Dealy<sup>27</sup> and Petrie.<sup>28</sup> Moreover, other significant reason for considering elongational flow is the fact that a knowledge of physical behavior in shear does not generally suffice to characterise a material's response in other type of deformation.

Most studies on polymer melt rheology have concentrated on shear flow,<sup>29-31</sup> which were observed in such flow geometries as Poiseuille flow and Couette flow and in parallel plate and cone and plate torsion flows. Simple shear flow in the Cartesian coodinate can be visuallized by considering a liquid between two flat plates, one stationaly and the other moving. We assign labels to the points using the spatial coordinates  $x^{i}$  at time t and the body coordinates  $X^{i}$  at an arbitrary past time t' (Figure 2). We shall write the equation characterizing the deformation in the form

$$x^{1} = x^{1} + \varepsilon x^{2}$$
  
 $x^{2} = x^{2}$   
 $x^{3} = x^{3}$   
1.1

The Finger strain tensors are calculated directly from

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Figure 1.2 Shear flow



Figure 1.3 Elongational flow

$$C_{ij}^{-1} = \delta^{\alpha\beta} \frac{\partial x^{i}}{\partial X^{\alpha}} \frac{\partial x^{j}}{\partial X^{\beta}}$$
 1.2

In terms of physical components  $C_{\mbox{ij}}^{-1}$  , one has

$$\mathbb{C}^{-1} = \begin{pmatrix} 1+\varepsilon^2 & -\varepsilon & 0 \\ -\varepsilon & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}$$
 1.3

Simple shear is accompanied by the rotation of flow unit, as found by equation (1.1) or by Figure 1.2. This causes a macromolecular coil to rotate about its center of gravity, and lessens the orientating effect of flow field. This motion of polymer molecules is considered as a cause of non-Newtonian viscosity of polymer melt,<sup>32</sup> which decreases with increasing strain rate.

In elongation, the deformation in one direction, say  $x^{1}$ , is independent of position in the other two direction (Figure 3). The dimensional change is given by

$$x^{1} = sX^{1}$$
  
 $x^{2} = s^{-1/2} X^{2}$   
 $x^{3} = s^{-1/2} X^{3}$   
1.4

Where s is an elongation ratio. The component of  $C^{-1}$  is

$$\mathbf{C}^{-1} = \begin{pmatrix} s^2 & 0 & 0 \\ 0 & s & 0 \\ 0 & 0 & s \end{pmatrix}$$
 1.5

The rate of strain tensor e is also independent of position and is given by

$$e = \begin{pmatrix} 1 & 0 & 0 \\ 0 & -\frac{1}{2} & 0 \\ 0 & 0 & -\frac{1}{2} \end{pmatrix}$$
 1.6

where  $\gamma$  is the rate of strain, and if we consider elongation a cylindrical specimen of length 1 and crosssection area A then,

$$\dot{\gamma} = \frac{1}{I} \frac{dI}{dt} = -\frac{1}{A} \frac{dA}{dt}$$
 1.7

Upon integration, we obtain the following expression for the time dependent length and cross-section area that must be created in order to generate this flow.

$$l(t) = l_0 \exp(\dot{\gamma}t)$$
  
A(t) = A\_0 \exp(-\dot{\gamma}t) 1.8

Thus, the needed elongational strain  $\gamma t$  for elongational flow is

$$\gamma = \dot{\gamma}t = \ln\frac{1}{I_0} = \ln s \qquad 1.9$$

In elongational flow, the velocity gradient develops in the same direction as the velocity itself rather than orthogonal to it as in shear flow, i.e. elongational flow is strain process free from the rotation of flow unit. This favors a large elongation of the long chain molecules from the random coil conformation and a stable orientation of elongated molecules in the direction of flow.

The experiments of elongational flow was first studied by Trouton<sup>33</sup> in 1906 for pitch, tar and shoemaker's wax descending under their own weight. Trouton defined the elongational viscosity as

$$\lambda = \frac{F/A}{dV/dX}$$
 1.10

where F is applied axial force and dV/dx velocity gradient in the axial direction. Nowwe consider the velocity V at a position x and V<sub>0</sub> at an initial position, then the elongational strain at a position x is

$$\gamma = \ln \frac{V}{V_0}$$
 1.11

and

$$\dot{\gamma} = \frac{1}{V} \frac{dV}{dt} = \frac{dV}{dx}$$
 1.12

Hence the Trouton's elongational viscosity is equal to

$$\lambda = \frac{\sigma}{\dot{\gamma}}$$
 1.13

Trouton also showed that the elongational viscosity was three times the shear viscosity for Newtonian fluids:  $\lambda = 3\eta$ .

Recently, the experiments of a constant elongational strain rate for polymer melt have been studied by Ballman, Vinogradov,<sup>35</sup> Meissner.<sup>36</sup> On the other hand, Cogswell<sup>37</sup> has employed a constant stress method and obtained the elongational viscosity. An alternative method studing elongational flow is an isothermal melt spinning method. The isothermal melt spinning has been examined by Acierno et.al.<sup>38</sup> and Han and Lamonte.<sup>39</sup> The elongational experiments for polymer solution have been also carried out by using of a solution spinning method,<sup>40,41</sup> the tubeless-syphon method<sup>42</sup> and the triplet jet method.<sup>43</sup>

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