

### Front Illustration

85 mm diameter single screw extruder for thermoplastics, fitted with a crosshead die for wire covering; the barrel is electrically heated and air cooled. Reproduced by kind permission of Francis Shaw & Co. Ltd, Manchester.

## Principles of Polymer Processing

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### **Principles of Polymer Processing**

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## Preface

The increasing use of synthetic polymers in preference to metals and other engineering materials for a wide range of applications has been accompanied by the development and improvement of processes for converting them into useful products. Indeed, it is often the comparative ease and cheapness with which polymeric materials can be processed that make them attractive choices. Because of the relatively complex behaviour of the materials, polymer processes may appear to be difficult to understand and analyse quantitatively.

The purposes of this book are to introduce the reader briefly to the main methods of processing thermoplastic polymers, and to examine the principles of flow and heat transfer in some of the more industrially important of these processes. Much attention is devoted to the two most widely used methods — screw extrusion and injection moulding. Quantitative analyses based on mathematical models of the processes are developed in order to aid the understanding of them, and to improve both the performance and design of processing equipment. In addition to algebraic formulae, some worked examples are included to illustrate the use of the results obtained. In cases where analytical solutions are not possible, methods of numerical solution using digital computers are discussed in some detail, and typical results presented.

This book is partly based on courses given by the author to both undergraduate and postgraduate students of mechanical and chemical engineering at Imperial College. The level of continuum mechanics and mathematics employed is that normally taught in undergraduate engineering courses. Although tensor notation is introduced for conciseness in presenting general continuum mechanics equations (in chapter 4), no prior knowledge is assumed, and it is not used in the later practical applications. The book is therefore suitable for engineering undergraduates and postgraduates, and other students at equivalent levels. Practising polymer engineers may also find it useful.

The author wishes to acknowledge the contributions made by many colleagues, students and technical staff to the work on polymer processing at Imperial College, on which this book is firmly based. Much of the material concerned with cable-covering-crosshead design presented in chapter 5 is drawn from the current work of Mrs F. Nadiri. The very skilful typing services of Miss E. A. Quin are also gratefully acknowledged.

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## **Notation**

The mathematical symbols used in the main text are defined in the following list. In some cases, particular symbols have more than one meaning in different parts of the book. Where appropriate, the chapter or section in which a particular definition applies is indicated in parentheses. Many of the symbols used in the appendixes are the same as in the main text, although some new notation is introduced and defined within each appendix.

```
dimensionless coordinate in direction of flow (section 4.5)
Α
\boldsymbol{A}
                    a function defined in equation 5.101 (section 5.5.1)
                    cross-sectional area of a screw channel (section 6.2.1)
\boldsymbol{A}
\boldsymbol{A}
                    integration constant in equations 5.6 and 7.5
                    radius of a cone-and-plate-rheometer cone
а
                    constants in general thermal boundary condition equation 4.47
a_1, a_2, a_3
В
                    integration constant in equation 5.35
В
                    a function defined in equation 5.101 (section 5.5.1)
Br
                    Brinkman number
b
                    radius of drum driving cone-and-plate rheometer (section 3.3.1)
b
                    temperature coefficient of viscosity at constant shear rate
\begin{array}{c} b_i \\ C \\ C \\ C \\ C_p \\ C_{p\,\mathrm{m}} \\ C_{1} - C_5 \\ C_1, C_2 \end{array}
                    body force vector
                    consistency (viscosity) in power-law equation
                    integration constant in equation 5.102 (section 5.5.1)
                    a function of screw geometry in equation 6.79 (section 6.2.1)
                    specific heat (of melt)
                    specific heat of a melt
                    specific heat of a solid polymer
                    constants defined in equations 5.5, 5.20, 5.32, 5.40 and 5.81
                    constants defined in equations 6.92 and 6.96
                    radial clearance between barrel and flight tips
D
                    diameter of a circular flow channel, including a rheometer
D
                    internal diameter of an extruder barrel (chapter 6)
D'
                    diameter of a molten extrudate after leaving a capillary
                    reference mould cavity diameter
D_0
                    initial and final diameters of a tapered circular die
                    a function defined in equation 5.37 (section 5.1.3)
\boldsymbol{E}
                    a function defined in equation 5.106 (section 5.5.1)
```

x NOTATION

E	a function of screw geometry in equation 6.79 (section 6.2.1)
E È	rate of working per unit area of extruder-barrel surface
E	
$E_z$ ,	power consumption per unit downstream length of channel
$E_{z\atop E_{z'}}$	power consumption per unit downstream length of flight
$E_1$	power consumption per unit length of upper calender roll
e	capillary end correction (section 3.3.3)
e	rate of extension of a filament (section 4.3.4)
e	width of a screw flight (chapter 6)
$e_{ij}$	rate-of-deformation tensor
<b>F</b>	force on a calender roll, per unit length of roll (section 5.4.1)
$\boldsymbol{F}$	net transverse force at the flights in a screw-feed section (section
-	6.2.1)
r	·
$F_D$	drag flow rate shape factor
$F_{P}$	pressure flow rate shape factor
$f_D$	drag flow velocity shape factor
	pressure flow velocity shape factor
$\stackrel{f_P}{G}$	Griffith number
Gz	Graetz number
$G_1, G_2$	functions introduced in equations 6.84 and 6.85
g	summation counter used in equations 6.46 and 6.47
$g_{ij}$	velocity gradient tensor
H	depth of a flow channel, including screw channel and mould
	cavity
$H_{s}$	depth of solid bed
$H_0$	distance between the lips of a flat film die (section 5.3.1)
$H_0$	minimum distance between the rolls of a calender (section 5.4)
$H_{0}$	initial thickness of a thin melt film (section 5.5.2)
$H_0$	initial screw-channel depth (section 6.1.4)
$H_0$	reference-mould-cavity depth (section 7.3.3)
$H_1$	initial channel depth in a flat film die (section 5.3.1)
$H_1$	distance between calender rolls at point of separation (section 5.4)
$H_1, H_2$	initial and final depths of a tapered flat-slit die (section 5.1.2)
$H_1, H_2$	functions introduced in equations 6.84 and 6.85
h	height of a flow element in a cone-and-plate rheometer (section
	3.3.1)
h	heat-transfer coefficient (section 4.4)
h	length of a striation (section 4.7)
h	thickness of solid skin in a mould cavity (section 7.3.4)
$I_1, I_2, I_3$	principal invariants of the rate-of-deformation tensor
$I_2^*$	dimensionless form of the second invariant
K	ratio between outer and inner radii of a flow channel (section
	5.1)
K	a function introduced in equation 6.78 (section 6.2.1)
k	thermal conductivity (of melt)
<i>k</i> <sub>m</sub>	thermal conductivity of a melt
$k_{\rm s}$	thermal conductivity of a solid polymer
$k_1, k_2, k_3$	ratios between compressive stresses at barrel, channel sides and
	screw root, respectively, and stress in downstream direction

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L	length of a flow channel, including rheometer capillary
L	axial length of a screw
L'	length of flow channel required for fully developed flow
$L_0$	distance between pressure initiation and nip in a calender
$L_1$	length of the die lips at the centre of a film die (section 5.3.1)
$L_1$	distance of point of separation from calender nip (section 5.4)
$L_1, L_2, L_3$	lengths of three rheometer capillaries (section 3.3.3)
l	fluid displacement in mixing analysis (section 4.7)
l	screw-flight pitch (chapter 6)
M	degree of distributive mixing
<u>M</u>	a function introduced in equation 6.80 (section 6.2.1)
$\overline{\underline{M}}$	mean degree of distributive mixing
$\overline{M}_{x}$	mean distributive mixing per unit channel length
$M_{\mathbf{m}z}$	downstream mass flow rate in the melt pool
$M_{\mathrm{T}}$	total downstream mass flow rate in a screw channel
m	number of screw channels in parallel (number of starts)
m	total mass flow rate from an extruder
$m_{fx}$	leakage mass flow rate per unit downstream distance
$m_1$	resultant upper melt film mass flow rate per unit width of film
$m_{1x}, m_{1z}$	upper melt film mass flow rates in $x$ and $z$ directions, per unit
	width of film in the z and x directions
$m_{2z}$	lower-melt-film downstream mass flow rate per unit width of film
$m_{3z}$	side melt film downstream mass flow rate per unit width of film
N n	screw speed power-law index
n	direction normal to a flow boundary (section 4.4)
n'	gradient of shear-stress curve plotted logarithmically against
<i>"</i>	apparent shear rate
<b>P</b> .	pressure drop over rheometer capillary (section 3.3.3)
P	pressure just after the gate to a mould cavity (section 7.3.3)
Pe	Peclet number
$P_r$	pressure gradient in the radial direction
$P_x$	pressure gradient in the x direction
$\hat{P_{x}}_1$	pressure gradient in the x direction at channel inlet
$\hat{P_z}$	pressure gradient in the z direction
$P_{z1}^{z}$	pressure gradient in the z direction at channel inlet
$P_0$	pressure drop for a zero-length capillary
p	pressure
p	compressive stress in a solid plug, acting in the downstream
	direction (section 6.2)
$p_0$	downstream pressure at the beginning of a feed section
$p_{1}, p_{2}$	pressures at flow-channel inlet and outlet
$\Delta P$	pressure difference across an injection nozzle
Q	volumetric flow rate (per unit width in the case of an infinitely
	wide channel, per screw channel in the case of an extruder)
Q'	volumetric flow rate at a particular position along the manifold of
	a flat film die
$Q_{ m F}$	volumetric downstream flow rate in the clearance

v	1	3
л	1	3

### NOTATION

$Q_{ m L}$	volumetric leakage flow rate over the flights
$Q_{s}, Q_{x}, Q_{y}$	volumetric flow rates in the s, x and y directions, per unit width
$Q_0$	of channel reference volumetric flow rate in a mould cavity
$Q_1, Q_2, Q_3$	components of the volumetric flow rate in the melt pool
q	surface heat transfer rate (section 4.4)
$\dot{q}$	volumetric flow rate per unit width of a flat film die (section
	5.3.1)
q	volumetric rate of melt influx per unit surface area (section 5.5.2.)
R	radius of calender rolls (section 5.4)
R	radial distance of flow front from the gate (section 7.3.3)
Re	Reynolds number
$R_0$	reference flow front radius in a mould cavity
$R_1$	outer radius of disc mould cavity
r <del>r</del>	radial coordinate in cylindrical polar system
-	mean distance of a flow channel from the axis of symmetry reference radius in a mould cavity
$r_0$ $r_1, r_2$	inner and outer radii of an annular flow channel
S S	swelling ratio (section 3.4)
S	striation thickness (section 4.7)
S	dimensionless shear stress (sections 5.5.1 and 6.3.3)
S	screw channel depth ratio (section 6.1.4)
S'	striation thickness modified by mixing
S	coordinate in the resultant direction of flow
$\frac{T}{T}$	temperature
$rac{T^*}{T}$	dimensionless temperature
	bulk mean temperature
$T_{b}$	temperature of a flow boundary, including extruder barrel
$T_{ m m} \ T_{ m s}$	melting-point temperature temperature of the screw surface
$T_{\bullet}^{s}$	reference temperature for viscosity measurements
$T_0 \\ T_0 *$	a dimensionless temperature introduced in equation 5.98
$T_1$	temperature at flow inlet
$\overline{T}_1, \overline{T}_2$	bulk mean temperatures in the upper and lower melt films
$\overline{T}_{ extsf{s} extsf{b}}$	bulk mean temperature of the solid bed
$T_{\infty}$	temperature remote from a flow boundary
$\Delta T$	mean temperature rise in a flow
t	time
$t_{ij}$	total-stress tensor
$t_{m}$	local memory time
t <sub>m</sub> *	dimensionless local memory time
$egin{array}{c} U \ U \end{array}$	boundary velocity in the $x$ direction (section 3.2) dimensionless velocity in the $x$ direction
	dimensionless velocity in the $x$ direction (section 6.1.4)
$rac{U^{f *}}{\overline{U}}$	mean or characteristic velocity in the $x$ direction (section of the section)
$U_1$	dimensionless x direction velocity at flow channel inlet (section
1	4.5)

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** **	
$U_1, U_2$	peripheral speeds of calender rolls
и	velocity component in the $x$ or $r$ direction
V	velocity of capillary rheometer piston (section 3.3.3)
V	dimensionless velocity in the $y$ direction (section 4.5)
V	wire speed (section 5.1.4)
V	barrel velocity relative to the screw (chapter 6)
$V_{\rm r}$	resultant velocity of barrel relative to the solid bed
$\dot{V_{ m s}}$	resultant relative boundary velocity
$V_{sz}$	downstream velocity of the solid bed relative to the screw
$V_x^{32}$	boundary velocity in the x direction
$V_z$	boundary velocity in the z direction
$\overrightarrow{V}_1$	dimensionless y-direction velocity at flow-channel inlet (section
· 1	4.5)
$V_1, V_2$	relative velocities in the plug-flow analysis (section 6.2.1)
ν 1, , , 2	velocity component in the $y$ or $\theta$ direction
$v_i$	velocity component in the general $x_i$ direction
W	load applied to drive cone-and-plate rheometer (section 3.3.1)
W	over-all width of a flat film die (section 5.3.1)
W	width of a screw channel (chapter 6)
W*	dimensionless velocity in the $z$ direction (section 6.1.4)
$W_{m}$	width of the melt pool
γm w	velocity component in z direction
X	dimensionless x direction coordinate
X	width of the solid bed (section 6.3)
x	cartesian coordinate
	general coordinates
$\stackrel{x_i}{Y}$	dimensionless y direction coordinate
<i>Y</i> *	
	dimensionless y direction coordinate (section 6.1.4)
<i>y</i>	cartesian coordinate
y'	cartesian coordinate measured from stress neutral surface (section
	6.1.3)
<i>y</i> <sub>0</sub>	position of stress neutral point (section 5.4.2)
Z	helical length of a screw channel
Z*	dimensionless z direction coordinate
Z	cartesian coordinate
$z_{\mathbf{m}}$	helical length of a screw channel required for melting
α	angle of cone-and-plate-rheometer cone (section 3.3.1)
α	pressure coefficient of viscosity at constant shear rate
α	angle of rotation of striation produced by mixing (section 4.7)
α .	semiangle of taper of flat film die lips (section 5.3.1)
α	feed angle for solid plug in a screw channel (section 6.2)
β	angle of inclination of flat film die manifold arms
γ	shear rate
$ar{ar{\gamma}}$ .	mean shear rate
$\dot{\gamma}_{a}$	apparent shear rate at the wall of a capillary
${\gamma_{t}}'$	true shear rate at the wall of a capillary
γο	reference shear rate for viscosity measurements
· -	

xiv	NOTATION
$\delta_{ij}$	Kronecker delta
$\delta_1, \delta_2, \delta_3$	thicknesses of the melt films at the barrel, screw root and flight
01,02,03	surfaces
$\epsilon_1$	initial elastic strain in a cone-and-plate rheometer
$\epsilon_2$	recovered elastic strain in a cone-and-plate rheometer
$\eta_1$	generalised viscosity in stokesian constitutive equation
$\eta_2$	cross viscosity in stokesian constitutive equation
$\theta$	angle of rotation of cone-and-plate-rheometer cone (section 3.3.1)
$\theta$	angular coordinate in cylindrical polar system
θ	an angle introduced in equation 5.77 (section 5.4)
$\theta$	helix angle of an extruder screw
$\overline{ heta}$	helix angle at the mean channel depth
$\theta_{ m b}$	helix angle at the barrel surface
$\theta_{s}$	helix angle at the screw root
$\overset{\circ}{\theta}_{1}^{s}$	an angle introduced in equation 5.77
λ	Trouton extensional viscosity (section 4.3.4)
λ	latent heat of fusion
λ	parameter defining the position of the stress neutral surface
	(section 6.1.3)
λ	pressure profile parameter introduced in equation 6.81 (section
	6.2.1)
$\lambda_1, \lambda_2$	pressure profile parameters introduced in equation 6.87
μ	shear viscosity
$\frac{\mu}{\overline{\mu}}$	mean viscosity
$\mu'$	viscosity in the clearance between screw flight and barrel
$\mu_{\mathrm{b}},\mu_{\mathrm{f}},\mu_{\mathrm{s}}$	coefficients of friction at the barrel, flight sides and screw root
	surfaces
$\mu_0$	reference viscosity for viscosity measurements
$\pi_E$	dimensionless channel power consumption
$\pi_P$	dimensionless pressure gradient
$\pi_Q$	dimensionless volumetric flow rate
$\pi_{Q1}, \pi_{Q2}$	dimensionless volumetric flow rates in the upper and lower melt
	films
$\pi_X$	dimensionless pressure gradient in the transverse channel direction
ρ	density (of melt)
$ ho_{m}$	density of a melt
$ ho_{ extsf{S}}$	density of a solid polymer
τ 	shear stress
$\overline{\tau}$	mean shear stress
τ'	shear stress at the wall of a capillary
$ au_{ij}$	viscous-stress tensor
$\tau_{xx}^*, \tau_{xy}^*$	dimensionless stress components
$ au_{xy}$ , $ au_{zy}$	screw channel shear-stress components evaluated at the barrel
_	surface
$\overline{ au}_{0}$	mean shear stress at ambient pressure
<u></u>	resultant shear stress in the upper melt film
$ar{ au}_1,ar{ au}_2$	mean shear stresses in the upper and lower melt films

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$\tau_{1z}, \tau_{2z}, \tau_{3z}$	downstream shear-stress components in the upper and lower melt
127 227 02	films
Φ	a function introduced in equation 5.77 (section 5.4.1)
$\phi$	a function introduced in equation 5.99 (section 5.5.1)
$\phi$	dimensionless pressure-dependence parameter
Ψ	stream function
$\dot{\Omega}$	rate of rotation of cone-and-plate-rheometer cone
$\omega$	rate of solidification per unit area (section 7.3.4)
$\omega_{ii}$	rate-of-rotation tensor
$\omega_1, \omega_2, \omega_3$	rates of melting per unit area from the top, bottom and side of
	the solid bed

# 1

## Introduction

The last few decades have seen the rapid development of synthetic polymeric materials to the point where, in a number of countries, their total rate of production exceeds that of metals on a volume basis, and will in due course do so in terms of weight. Along with the expansion in the manufacture of polymers has come the development and improvement of processes to convert them into useful products.

### 1.1 Polymeric Materials

Polymeric materials, for present purposes, are those synthetic high molecular weight materials that are of commercial importance. They include plastics — of both the thermoplastic and thermosetting type — and elastomers (rubbers). The most important of these are thermoplastics, and the main emphasis of this book is on the processing of such materials, although at least some of the principles discussed can be applied to thermosets and rubbers.

As engineering materials, polymers compare unfavourably with many older materials, notably metals. They lack strength and stiffness, show time-dependent behaviour, are often unexpectedly brittle and can be used only over limited temperature ranges. Their one main advantage, however, which frequently overrides other considerations, is the ease and cheapness with which they can be processed. A single operation can often be used to produce a finished article of considerable geometric complexity, but of high dimensional accuracy and surface finish. The cost of processing rarely exceeds that of the raw material. The mechanical properties of polymers may be improved by adding reinforcing fillers or by making composite materials. Although a wide range of both organic and inorganic fillers are used in thermosets, this is much less commonly done with thermoplastics, for the very good reason that the resulting mixtures are less easy to process.

The most important thermoplastics, in terms of rates of production, are low- and high-density polyethylenes, polypropylene, polystyrene, polyvinyl chloride and nylons. All of these are available in many grades, having properties appropriate for different applications and processing techniques. Differences between grades are often due to differences in mean *molecular weight* (reflecting the average size of the

long-chain molecules) and molecular weight distribution (reflecting the variation of molecule size about the mean). In addition to reinforcing fillers and colourants, various additives may be used to affect the properties of the material either during processing or in later use. These include lubricants and plasticisers to facilitate processing, stabilisers against degradation (breakdown of the molecules) by heat or light, and fire retardants to make the product less inflammable.

Thermoplastics, as their name suggests, melt reversibly on heating and are usually shaped into the required form while in the molten or *melt* state. Although melts can best be described as fluids, they behave very differently from most familiar fluids. For example, they display significant elastic properties, and should therefore be regarded as *viscoelastic*. Also, melt viscosities are both very high and non-newtonian.

#### 1.2 Polymer Processing

Polymer processing is concerned with the operations carried out on polymeric materials to form them into useful products. The chemical processes involved in the manufacture of polymers from their monomers are specifically excluded. In the case of thermoplastics, the main steps in any process are first to melt, then to shape and finally to cool the material in its new form. The heat required for melting may be supplied by radiation or conduction, or by mechanical work. Mixing of the melt is desirable to improve the properties of the product.

Thermoplastics are normally obtained from the polymerisation reactor in the form of either powder or melt. While it is possible for a processor to purchase some materials in powder form for direct conversion into a finished product, the polymer manufacturer often carries out an homogenising or compounding operation after polymerisation. Homogenisation serves to mix the raw polymer thoroughly, in order to break up lumps of high-molecular-weight material, for example, and also to remove unconverted monomer or other volatiles. It also permits the inclusion of additives, and sale of the material in the form of granules, which are easier to use than powder in the subsequent product-processing operations. Homogenisation is carried out in very large screw extruders or other continuous mixing equipment. Screw extruders and product processing equipment are discussed in chapter 2.

Two main methods are used to make polymer granules. With a technique known as underwater die face cutting, the melt is forced through a multihole die into water, where the emerging strands are cut at the die face to form discrete particles; the particles are roughly spherical in shape when solidified. Alternatively, extruded strands are solidified by cooling in a water bath before being cut; this method, known as lace cutting, gives granules of cylindrical shape. The term granulation in the present context of polymer manufacture should not be confused with the methods used to reduce the size of process scrap prior to reprocessing.

### 1.3 Analysis of Polymer Processes

Although polymer processes have often been evolved by largely trial-and-error methods, the performances currently being demanded make it increasingly important not only to understand how such processes function, but also to be able

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