

## Studies on Dry Spinning. I. Fundamental Equations

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### Synopsis

One difficulty in analyzing the state of filament in the dry spinning process is that in the boundary conditions required to solve the equations of mass, momentum and energy which are derived on the consideration of balance with respect to infinitesimally small element are not given *a priori*. The equations which include these boundary conditions in themselves are derived by considering mass, momentum, and energy balances with respect to the entire cross section of filament. These additional macroscopic equations are simplified to a great extent by assuming the flat velocity profile through the cross section of filament. Besides, in the steady state, these macroscopic equations are modified to give the equations of average solvent content, spinning tension, cross-sectional area, and average temperature. When the spinning conditions are given and the physical constants are measured for a given polymer and solvent system, it becomes possible to calculate the state of filament by solving these microscopic and macroscopic equations simultaneously without resorting to actual spinning.

### INTRODUCTION

Simultaneous partial differential equations on melt spinning were derived by Kase and Matsuo.<sup>1,2</sup> Both transient and steady-state solutions of the equations were shown to explain systematically the behavior of filament in melt spinning. In this paper, as an extension of the above studies on melt spinning, the fundamental equations on dry spinning are derived, being based on macroscopic and microscopic balances of mass, momentum, and energy.

An essential difference between melt and dry spinning is the number of components involved. In melt spinning we deal with one component, i.e., polymer, whereas in dry spinning we deal with two components, i.e., polymer and solvent. Dry spinning, therefore, involves the problem of mass transport in a binary system. The mass transport inside a filament may be treated as a molecular diffusion process, while that at the boundary layer on the filament surface may be treated by the concept of interphase mass transfer of solvent vapor.

The fundamental equations on dry spinning contain various constants which depend on the physical properties of polymer solution. These constants, therefore, vary both with temperature and concentration of polymer solution. Among the constants are the mass diffusivity, the activity of solvent, and the Trouton viscosity.

A difficulty in analyzing dry spinning mathematically is that a mere set of general spinning conditions and equations of continuity, motion and energy which are derived by considering balances over a infinitesimally small region of the system (hereafter, referred to as microscopic balances), does not completely specify the whole problem, since the boundary conditions to the above equations such as filament diameter or rates of mass, momentum and heat transfer across the filament surface are not yet given. In order to specify the above boundary conditions, another set of equations must be derived in addition to the equations of microscopic balances. These additional equations contain variables related to the above boundary conditions and are derived by considering balances over a region of filament which includes its entire cross section and surface (referred to as macroscopic balances).

Equations of microscopic and macroscopic balances, which constitute the fundamental equations, should be solved as a set of simultaneous partial differential equations. Solutions of these equations should simultaneously give the conditions inside the filament such as temperature and the boundary conditions such as filament diameter.

Because of excessive complexity it is difficult to solve the fundamental equations in their most general form. We must, therefore, reduce them into a simpler form by setting up appropriate assumptions or approximations and discard equations which are less important on the basis of physical considerations.

First we derive the general nonsteady-state equations assuming axial symmetry, then simplify the macroscopic equations by assuming a flat velocity distribution, and finally obtain under the steady-state condition the equations of solvent content, spinning tension, cross-sectional area, and temperature of filament.

Once these steady-state equations are obtained and the physical constants are measured or estimated for a given polymer and solvent system, the state of a filament along its traveling direction during spinning process can be calculated and it may become possible to predict properties of filaments dry spun under various spinning conditions without resorting to actual spinning. From the equation of solvent content, for instance, it becomes possible to calculate the distance from the spinneret required to complete the drying of solvent. This calculation is expected to give us useful knowledge concerning the selection of solvent, design of the length of a spinning column, and the determination of spinning conditions. Furthermore, it will become possible to roughly predict the cross-sectional shape of a dry spun filament by solving the equation for the cross-sectional area by assuming that the perimeter of the cross section of a filament remains unchanged after the polymer solution gains a certain viscosity level which does not allow further stretching of filament. It may also be possible to predict the orientation of polymer chains in a dry spun filament from the value of tensile stress by solving the equation of spinning tension.

Papers hitherto published on dry spinning discuss the effects of spinning conditions on filament properties only on the basis of experimental results. Theoretical analysis of the state of filament during dry spinning under various spinning conditions has not been done except by Sano and Nishikawa.<sup>3,4</sup> They calculated the residual solvent content and the temperature of filament and showed that air temperature and polymer solution throughput had a dominant influence on the residual solvent content.<sup>3,4</sup> Calculation of the deformation of filament, however, was not covered by the previous workers. The present theory, as mentioned above, will make it possible to predict the relation between spinning conditions and filament properties and to calculate the state of filament being dry spun.

### DERIVATION OF THE FUNDAMENTAL EQUATIONS

We adopt the cylindrical coordinate system. The origin of the  $z$  axis is taken to be the position at some short distance from the spinneret where the velocity profile of the jet is considered to become practically flat. The  $z$  axis is positive in the traveling direction of the filament. The origin of the  $r$  axis is taken to be the center of the filament. In the following discussions we will limit our consideration to the state of one filament. A macroscopic balance will be made over the segment of the filament which is bounded by two  $z$  and  $(z + \Delta z)$  planes perpendicular to the  $z$  axis and separated by a small distance  $\Delta z$  (see Fig. 1). Assumptions necessary to derive the equations will be made where they are required.

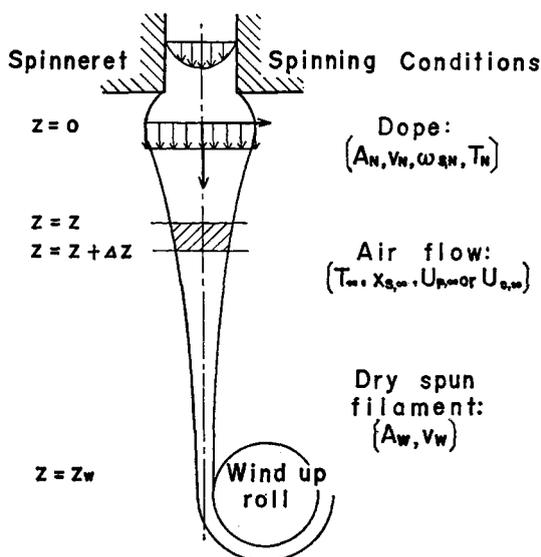


Fig. 1. Dry-spinning system.

## Equations of Continuity

### *Equation of Continuity for the Polymer Component*

First we make the following assumptions.

**Assumption 1.** The cross section of a filament is circular. Since the cross section of a filament produced by means of the dry spinning process often shows the shape of a dumbbell or raisin,<sup>5,6</sup> it might be necessary to make some corrections to accurately analyze the later stages of drying process. At an earlier stage, however, the assumption is considered valid.

**Assumption 2.** The flow velocities  $v_P$  and  $v_S$  of the polymer component and the solvent component, respectively, are both axially symmetrical.

**Assumption 3.** The concentration distribution within a filament is axially symmetrical.

We take the mass fractions  $\omega_P(z, r, t)$  and  $\omega_S(z, r, t)$  as the concentrations of the polymer component and of the solvent component, respectively. Note the relation

$$\omega_P + \omega_S = 1 \quad (1)$$

**Assumption 4.** The temperature distribution  $T(z, r, t)$  within a filament is axially symmetrical.

The density  $\rho(z, r, t)$  of the filament, therefore, is axially symmetrical.

Now we consider the flow of a binary mixture of polymer and solvent across an arbitrary  $z$ -plane. The total mass flow rate of the polymer component,  $W_P(z, t)$ , across the  $z$  plane at time  $t$  will be

$$W_P(z, t) = \int_0^R 2\pi r \rho \omega_P v_{P,z} dr \quad (2)$$

where  $R(z, t)$  denotes the radius of filament and  $v_{P,z}(z, r, t)$  denotes the  $z$  component of the flow velocity of the polymer component.

Similarly, the total mass flow rate of the solvent component,  $W_S(z, t)$ , is represented by the equation:

$$W_S(z, t) = \int_0^R 2\pi r \rho \omega_S v_{S,z} dr \quad (3)$$

where  $v_{S,z}(z, r, t)$  denotes the  $z$  component of the flow velocity of the solvent component.

Addition of eqs. (2) and (3) yields the total mass flow rate  $W(z, t)$  of the binary mixture:

$$\begin{aligned} W(z, t) &= W_P(z, t) + W_S(z, t) \\ &= \int_0^R 2\pi r \rho (\omega_P v_{P,z} + \omega_S v_{S,z}) dr \\ &= \int_0^R 2\pi r \rho v_z dr \end{aligned} \quad (4)$$

where  $v_z(z, r, t)$  denotes the  $z$  component of the mass average velocity  $v(z, r, t)$  of the mixture.

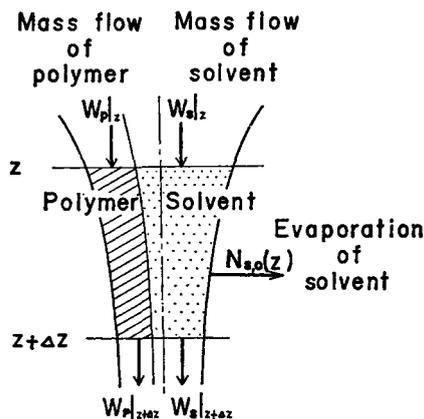


Fig. 2. Schematic description for macroscopic mass balance.

We now consider a macroscopic mass balance of the polymer component over a filament segment bounded by the  $z$  and  $(z + \Delta z)$  planes as shown in Figure 2.

The total mass of the polymer component in this segment is

$$\left( \int_0^R 2\pi r \rho \omega_P dr \right) \cdot \Delta z$$

Hence, the rate of mass accumulation of the polymer component in this segment is

$$\left[ \frac{\partial}{\partial t} \left( \int_0^R 2\pi r \rho \omega_P dr \right) \right] \cdot \Delta z$$

Thus, the contributions to the mass balance over this segment are as follows:

Time rate of total mass accumulation of the polymer component in the segment:

$$\left[ \frac{\partial}{\partial t} \left( \int_0^R 2\pi r \rho \omega_P dr \right) \right] \Delta z$$

Total input of the polymer component across the  $z$  plane:

$$W_P(z, t)$$

Total output of the polymer component across the  $(z + \Delta z)$  plane:

$$W_P(z + \Delta z, t)$$

The mass balance then becomes

$$\left[ \frac{\partial}{\partial t} \left( \int_0^R 2\pi r \rho \omega_P dr \right) \right] \cdot \Delta z = W_P(z, t) - W_P(z + \Delta z, t) \quad (5)$$

By dividing the entire equation by  $\Delta z$  and taking the limit as  $\Delta z$  ap-

proaches zero, we get the macroscopic equation of continuity for the polymer component:

$$\frac{\partial}{\partial t} \left( \int_0^R 2\pi r \rho \omega_P dr \right) + \frac{\partial W_P}{\partial z} = 0 \quad (6)$$

### *Equation of Continuity for the Solvent Component*

The evaporation of the solvent must be complete before a filament is taken up on a winding machine. The movement of the solvent during the drying process in the spinning column consists of two processes: the diffusion from the interior of the filament to its surface and the transfer in the gas phase from the filament surface into the flowing air. The concentration distribution of the solvent at an arbitrary  $z$  plane will be schematically as in Figure 3.

First we consider the movement in the gas phase. The symbol  $x_{s,0}$  is used to denote the mole fraction of solvent vapor at the interface and  $x_{s,\infty}$  to denote the mole fraction contained in the flowing air. The transfer rate of the solvent at the interface is represented by the molar flux  $N_{s,0}$ .

Since the evaporation of the polymer component and the dissolution of air into the polymer solution can be neglected,  $N_{s,0}$  is given by the equation

$$N_{s,0} = k_z(x_{s,0} - x_{s,\infty})/(1 - x_{s,0}) \quad (7)$$

where  $k_z$ , defined by the equation, represents the mass transfer coefficient.<sup>7</sup>

We now apply the law of conservation of mass of the solvent component to the filament segment described in Figure 2. Noting that solvent evaporation is an additional contribution to the solvent mass balance unlike in the case of the polymer component, we have the following list.

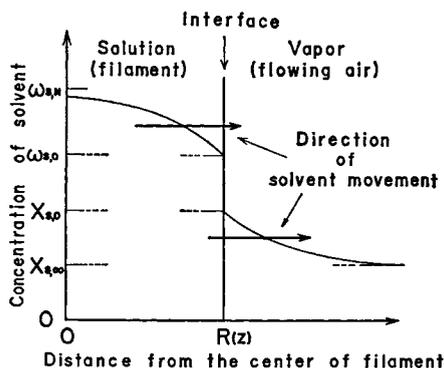


Fig. 3. Concentration distribution of solvent in the filament and in the gas phase.

Time rate of total mass accumulation of the solvent component in the segment:

$$\left[ \frac{\partial}{\partial t} \left( \int_0^R 2\pi r \rho \omega_s dr \right) \right] \cdot \Delta z$$

Total input of the solvent component across the  $z$  plane:

$$W_s(z, t)$$

Total output of the solvent component across the  $(z + \Delta z)$  plane:

$$W(z + \Delta z, t)$$

Rate of evaporation of solvent across the filament surface:

$$M_s N_{s,0} \cdot 2\pi R \Delta z$$

where  $M_s$  denotes molecular weight of solvent.

When the entire mass balance is written down and divided by  $\Delta z$ , we obtain the following macroscopic equation of continuity for the solvent component after letting  $\Delta z$  approach to zero:

$$\frac{\partial}{\partial t} \left( \int_0^R 2\pi r \rho \omega_s dr \right) + \frac{\partial W_s}{\partial z} + 2\pi R M_s N_{s,0} = 0 \quad (8)$$

Addition of eqs. (6) and (8) yields the macroscopic equation of continuity for the mixture:

$$\frac{\partial}{\partial t} \left( \int_0^R 2\pi r \rho dr \right) + \frac{\partial W}{\partial z} + 2\pi R M_s N_{s,0} = 0 \quad (9)$$

In order to know the velocity and concentration distributions within a filament we must make use of the equations of continuity for a binary mixture which are derived by considering the local mass balance with respect to each component.<sup>7</sup>

The general form for the mixture is

$$(\partial \rho / \partial t) + (\nabla \cdot \rho v) = 0 \quad (10)$$

The general form for the solvent is equivalent to the binary diffusion equation:

$$[\partial(\rho \omega_s) / \partial t] + (\nabla \cdot \rho \omega_s v) = (\nabla \cdot \rho D_{PS} \nabla \omega_s) \quad (11)$$

where  $D_{P,S}$  represents the binary diffusivity for the polymer-solvent system.

Since eqs. (10) and (11) are too complicated for solution, we simplify them by making assumptions (5), (6), and (7).

**Assumption 5.** Density  $\rho$  of the mixture is constant irrespective of pressure, composition, and temperature.

This means the assumption of incompressibility of the mixture.

**Assumption 6.** The diffusion in the  $z$  direction is negligible in comparison to that in the  $r$  direction.

**Assumption 7.** The movement of solvent due to average bulk flow  $v_r$  in the  $r$  direction is negligible in comparison to that due to the average bulk flow  $v_z$  in the  $z$  direction.

Assumptions (2) through (7) simplify eqs. (10) and (11) to eqs. (12) and (13), respectively:

$$\frac{1}{r} \cdot \frac{\partial}{\partial r} (rv_r) + \frac{\partial v_z}{\partial z} = 0 \quad (12)$$

$$\frac{\partial \omega_S}{\partial t} + v_z \frac{\partial \omega_S}{\partial z} = \frac{1}{r} \cdot \frac{\partial}{\partial r} \left( r D_{PS} \cdot \frac{\partial \omega_S}{\partial r} \right) \quad (13)$$

### Equation of Motion

Over a segment of filament bounded by the  $z$  and  $(z + \Delta z)$  planes as shown in Figure 4, we write a momentum balance in the  $z$  direction in the form:

$$\left\{ \begin{array}{l} \text{rate of} \\ \text{momentum} \\ \text{accumulation} \end{array} \right\} = \left\{ \begin{array}{l} \text{net flow of} \\ \text{momentum} \\ \text{into the segment} \end{array} \right\} + \left\{ \begin{array}{l} \text{Sum of forces} \\ \text{acting on} \\ \text{the system} \end{array} \right\} \quad (14)$$

The rate at which total momentum enters the  $z$  plane by convection is

$$\int_0^R 2\pi r \rho v_z v_z dr$$

The net convective momentum flow into this segment becomes, when  $\Delta z$  is small enough,

$$\begin{aligned} \int_0^R 2\pi r \rho v_z^2 dr \Big|_{z=z} - \int_0^R 2\pi r \rho v_z^2 dr \Big|_{z=z+\Delta z} \\ = - \left[ \frac{\partial}{\partial z} \left( \int_0^R 2\pi r \rho v_z^2 dr \right) \right] \cdot \Delta z \quad (15) \end{aligned}$$

The total upward force  $F(z,t)$  acting on the upper part end of the filament segment is

$$F = \int_0^R 2\pi r \tau_{zz} dr \quad (16)$$

The resultant force acting on this segment due to mechanical stresses in the  $z$  direction will be

$$\begin{aligned} - \int_0^R 2\pi r \tau_{zz} dr \Big|_{z=z} + \int_0^R 2\pi r \tau_{zz} dr \Big|_{z=z+\Delta z} \\ = \left[ \frac{\partial}{\partial z} \left( \int_0^R 2\pi r \tau_{zz} dr \right) \right] \cdot \Delta z \quad (17) \end{aligned}$$

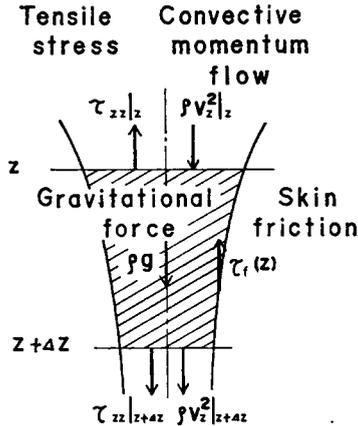


Fig. 4. Schematic description for macroscopic momentum balance.

For the notation, the positive direction, and the system for the subscripts of the stresses we adopt the same convention used in the textbook by Bennett and Myers.<sup>8</sup>

Other forces acting on the segment will be those arising from the gravitational force and the skin friction.

Hence the various contributions to the momentum balanced in the  $z$ -direction over the segment are as follows:

Time rate of momentum accumulation in the segment:

$$\left[ \frac{\partial}{\partial t} \left( \int_0^R 2\pi r \rho v_z dr \right) \right] \cdot \Delta z$$

Net convective momentum flow into the segment:

$$- \left[ \frac{\partial}{\partial z} \left( \int_0^R 2\pi r \rho v_z^2 dr \right) \right] \cdot \Delta z$$

Resultant tensile force acting on the segment:

$$\left[ \frac{\partial}{\partial z} \left( \int_0^R 2\pi r \tau_{zz} dr \right) \right] \cdot \Delta z$$

Gravitational force acting on the segment:

$$\left[ \int_0^R 2\pi r \rho g dr \right] \cdot \Delta z$$

Skin friction acting on the surface of the segment:

$$- \tau_f \cdot 2\pi R \Delta z$$

In the above list  $\tau_f$  denotes the local shearing stress or skin friction on the filament surface and  $g$  is the gravitational acceleration. We now substitute these expressions into eq. (14).

By dividing the entire resulting equation by  $\Delta z$  and taking the limit as  $\Delta z$  approaches zero, we obtain the macroscopic equation of motion:

$$\frac{\partial}{\partial t} \left( \int_0^R 2\pi r \rho v_z dr \right) = - \frac{\partial}{\partial z} \left( \int_0^R 2\pi r \rho v_z^2 dr \right) + \frac{\partial}{\partial z} \left( \int_0^R 2\pi r \tau_{zz} dr \right) + \int_0^R 2\pi r \rho g dr - 2\pi R \tau_t \quad (18)$$

Velocity and stress distributions within the filament are governed by eqs. (19) and (20) derived on consideration of the local momentum balance under the assumptions (2), (3), and (4).

$z$  component:

$$\rho \left( \frac{\partial v_z}{\partial t} + v_r \frac{\partial v_z}{\partial r} + v_z \frac{\partial v_z}{\partial z} \right) = \frac{\partial \tau_{zz}}{\partial z} + \frac{1}{r} \cdot \frac{\partial}{\partial r} (r \tau_{rz}) + \rho g \quad (19)$$

$r$  component:

$$\rho \left( \frac{\partial v_r}{\partial t} + v_r \frac{\partial v_r}{\partial r} + v_z \frac{\partial v_r}{\partial z} \right) = \frac{\partial \tau_{rr}}{\partial r} + \frac{\partial \tau_{rz}}{\partial z} + \frac{\tau_{rr} - \tau_{\theta\theta}}{r} \quad (20)$$

In order to solve eqs. (19) and (20) for velocities, we must express the various stresses on the right-hand side in terms of velocity gradients and fluid properties. Under assumption (8) below and assumption (5), eqs. (21)–(25) give the expressions for various stresses.

**Assumption 8.** The rheological properties of the fluid obey the law of the Newtonian fluid.

$$\tau_{zz} = -p + 2\mu(\partial v_z/\partial z) \quad (21)$$

$$\tau_{rr} = -p + 2\mu(\partial v_r/\partial r) \quad (22)$$

$$\tau_{\theta\theta} = -p + 2\mu(v_r/r) \quad (23)$$

$$\tau_{zr} = \mu(\partial v_z/\partial r + \partial v_r/\partial z) \quad (24)$$

$$\tau_{r\theta} = \tau_{z\theta} = 0 \quad (25)$$

where  $\mu$  is the viscosity of the fluid and is a function of temperature and composition of the fluid.

### Equation of Energy

We write the statement of the law of conservation of energy over the same filament segment as in mass and momentum balances (see Fig. 5). First make the following assumptions.

**Assumption 9.** Energy transport by radiation is negligible.

**Assumption 10.** Energy in and out by conduction in the  $z$  direction is negligible.

**Assumption 11.** The contributions of kinetic energy and work done by such forces as pressure, gravitational, viscous, and external forces are negligible.

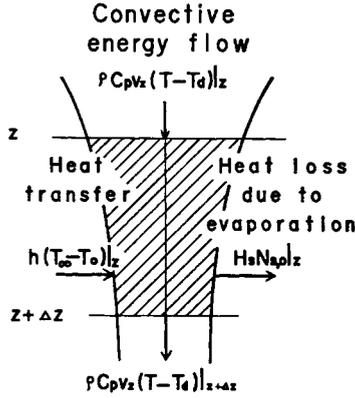


Fig. 5. Schematic description for macroscopic energy balance.

Unlike in melt spinning, we must take account of the additional contribution to the energy balance resulting from the difference in enthalpy of the solvent in the solution state and in the gas mixture.

The rate at which the energy enters the  $z$  plane with the flow of the mixture will be

$$\int_0^R 2\pi r \rho C_P v_z (T - T_d) dr$$

where  $C_P$  is the specific heat of the filament at constant pressure and  $T_d$  is a datum temperature.

The net convective energy flow into this segment becomes, when  $\Delta z$  is small enough:

$$- \left\{ \frac{\partial}{\partial z} \left[ \int_0^R 2\pi r \rho C_P v_z (T - T_d) dr \right] \right\} \cdot \Delta z$$

Hence, the various contributions to the macroscopic energy balance over the segment will be as follows.

Time rate of energy accumulation in the segment:

$$\left\{ \frac{\partial}{\partial t} \left[ \int_0^R 2\pi r \rho C_P (T - T_d) dr \right] \right\} \cdot \Delta z$$

Net convective energy flow into the segment:

$$- \left\{ \frac{\partial}{\partial z} \left[ \int_0^R 2\pi r \rho C_P v_z (T - T_d) dr \right] \right\} \cdot \Delta z$$

Energy loss due to evaporation of the solvent across the filament surface:

$$H_s N_{s,0} \cdot 2\pi R \Delta z$$

Energy gained from air flow by heat transfer across the filament surface

$$h(T_\infty - T_0) \cdot 2\pi R \Delta z$$

In the above list  $H_s$  denotes the partial molal enthalpy of the solvent at the filament surface in the gas mixture with respect to  $T_a$ ,  $h$  denotes the overall heat-transfer coefficient,  $T_\infty$  denotes the temperature of the air flow, and  $T_0$  denotes the temperature of filament surface.

When the entire energy balance is written down and divided by  $\Delta z$ , we obtain the following macroscopic equation of energy after letting  $\Delta z$  approach to zero

$$\frac{\partial}{\partial t} \left[ \int_0^R 2\pi r \rho C_P (T - T_a) dr \right] = - \frac{\partial}{\partial z} \left[ \int_0^R 2\pi r \rho C_P v_z (T - T_a) dr \right] - 2\pi R H_s N_{s,0} + 2\pi R h (T_\infty - T_0) \quad (26)$$

The temperature distribution within the filament will be obtained by solving the microscopic equation of heat conduction. The equation is given by eq. (27) under the assumptions (12), (13), and (14) below and those previously made.

**Assumption 12.** The specific heat  $C_P$  of filament is constant irrespective of temperature and composition.

**Assumption 13.** The thermal conductivity  $\lambda$  of filament is constant irrespective of temperature and composition.

**Assumption 14.** The energy movement due to the flow  $v_r$  in the  $r$  direction is negligible in comparison to that in the  $z$  direction.

$$\rho C_P \left( \frac{\partial T}{\partial t} + v_z \frac{\partial T}{\partial z} \right) = \frac{\lambda}{r} \cdot \frac{\partial}{\partial r} \left( r \frac{\partial T}{\partial r} \right) \quad (27)$$

### FUNDAMENTAL EQUATIONS UNDER FLAT VELOCITY ASSUMPTION

Although simplified in the previous section by assuming axial symmetry and other limiting conditions, the fundamental equations of dry spinning are still too complicated for solution. In this section it is shown that the macroscopic equations become quite simple under the two important assumptions (15) and (16) set forth below.

**Assumption 15.** The  $z$  component  $v_{p,z}$  of the velocity of the polymer component is equal to the  $z$  component  $v_{s,z}$  of the velocity of the solvent component. Hence both  $v_{p,z}$  and  $v_{s,z}$  are equal to the  $z$ -component  $v_z$  of the mass average velocity.

**Assumption 16.** The velocity  $v_z$  is constant over the filament cross-section (the assumption of the flat velocity).

Since in dry spinning the solvent concentration varies greatly in the radial direction, the viscosity near the surface is expected to be considerably higher than that at the center. Therefore, validity of assumption (16)

may be questioned. However, we first try to solve the equations under this assumption before discussing its validity.

### Equations of Continuity

Equations (2), (3), and (4) for the total mass flow rates  $W_P$ ,  $W_S$ , and  $W$  of the polymer component, the solvent component, and the mixture, respectively, reduce to eqs. (28), (29), and (30) under the flat velocity assumption.

$$W_P = A \rho v_z \langle \omega_P \rangle \quad (28)$$

$$W_S = A \rho v_z \langle \omega_S \rangle \quad (29)$$

$$W = A \rho v_z \quad (30)$$

where  $\langle \omega_P(z) \rangle$  and  $\langle \omega_S(z) \rangle$  denote the average mass fractions at  $z$  of the polymer component and of the solvent component, respectively, and  $A(z)$  denotes the cross-sectional area of filament at  $z$ . These quantities are given by eqs. (31)–(33).

$$\langle \omega_P \rangle = \frac{1}{A} \int_0^R 2\pi r \omega_P dr \quad (31)$$

$$\langle \omega_S \rangle = \frac{1}{A} \int_0^R 2\pi r \omega_S dr \quad (32)$$

$$A = 2\pi R^2 \quad (33)$$

The macroscopic equations of continuity for the polymer component, the solvent component, and the mixture are rewritten in terms of  $W_P$ ,  $W_S$ , and  $W$ , respectively, as eqs. (34)–(36),

Macroscopic equation of continuity for the polymer component:

$$\frac{\partial}{\partial t} \left( \frac{W_P}{v_z} \right) + \frac{\partial W_P}{\partial z} = 0 \quad (34)$$

Macroscopic equation of continuity for the solvent component:

$$\frac{\partial}{\partial t} \left( \frac{W_S}{v_z} \right) + \frac{\partial W_S}{\partial z} + 2\pi R M_S N_{S,0} = 0 \quad (35)$$

Macroscopic equation of continuity for the mixture:

$$\frac{\partial}{\partial t} \left( \frac{W}{v_z} \right) + \frac{\partial W}{\partial z} + 2\pi R M_S N_{S,0} = 0 \quad (36)$$

### Equation of Motion

Noting eq. (30), we can rewrite the macroscopic equation of motion (18) in the form

$$\frac{\partial F}{\partial z} = \frac{\partial W}{\partial t} + \frac{\partial}{\partial z} (v_z W) - A \rho g + 2\pi R \tau_t \quad (37)$$

where  $F$  denotes the spinning tension which arises from the normal stress in the  $z$  direction and is expressed by the equation

$$F = \int_0^R 2\pi r \tau_{zz} dr \quad (38a)$$

$$= A \langle T_{zz} \rangle \quad (38b)$$

Now we try to express  $F$  through the rheological equation. The isotropic component  $p$  of the stresses is defined as

$$p = -(\tau_{rr} + \tau_{\theta\theta} + \tau_{zz})/3 \quad (39)$$

The stresses arising from the surface tension will be expressed as

$$\tau_{rr} = \tau_{\theta\theta} = -\gamma/R \quad (40)$$

where  $\gamma$  is the surface tension.<sup>9</sup>

Thus we get from eqs. (39) and (40)

$$p = -(\tau_{zz}/3) + (2\gamma/3R) \quad (41)$$

Combination of eq. (41) with eq. (21) yields

$$\tau_{zz} = 3\mu(\partial v_z/\partial z) - (\gamma/R) \quad (42a)$$

$$= \beta(\partial v_z/\partial z) - (\gamma/R) \quad (42b)$$

In eq. (42b) above we used the relation that the Trouton viscosity  $\beta$  is three times the shear viscosity  $\mu$  in the Newtonian fluid.<sup>10</sup>

Inserting eq. (42b) into eq. (38a) and noting that  $\partial v_z/\partial z$  is independent of  $r$ , we obtain

$$F = A\langle\beta\rangle(\partial v_z/\partial z) - \gamma\sqrt{\pi A} \quad (43)$$

where  $\langle\beta\rangle$  is given by

$$\langle\beta\rangle = \frac{1}{A} \int_0^R 2\pi r \beta dr \quad (44)$$

### Equation of Energy

The macroscopic eq. (26) of energy will be simplified into the form

$$\begin{aligned} \frac{\partial}{\partial t} \left[ \frac{C_P W}{v_z} (\langle T \rangle - R_d) \right] + \left[ \frac{\partial}{\partial z} [C_P W (\langle T \rangle - T_d)] \right] \\ + 2\pi R H_s N_{s,0} - 2\pi R h (T_\infty - T_0) = 0 \end{aligned} \quad (45)$$

where the average temperature  $\langle T(z,t) \rangle$  is given by

$$\langle T \rangle = \frac{1}{A} \int_0^R 2\pi r T dr \quad (46)$$

After the execution of differentiation, eq. (45) is rearranged to eq. (47):

$$\frac{C_P W}{v_z} \left( \frac{\partial \langle T \rangle}{\partial t} + v_z \frac{\partial \langle T \rangle}{\partial z} \right) = 2\pi R h (T_\infty - T_0) = 2\pi R H_S N_{S,0} - C_P (\langle T \rangle - T_d) \left[ \frac{\partial}{\partial t} \left( \frac{W}{v_z} \right) + \frac{\partial W}{\partial z} \right] \quad (47)$$

Substitution of the macroscopic eq. (36) of continuity for the mixture into the third term of the right-hand side of eq. (47) yields

$$\frac{C_P W}{v_z} \left( \frac{\partial \langle T \rangle}{\partial t} + v_z \frac{\partial \langle T \rangle}{\partial z} \right) = 2\pi R h (T_\infty - T_0) - 2\pi R N_{S,0} [H_S - M_S C_P (\langle T \rangle - T_d)] \quad (48)$$

Now we make assumption (17).

**Assumption 17.** The difference between the enthalpy of the solvent in the solution phase and that in the gas mixture is approximated by the heat of vaporization  $L_S$  of the pure solvent at its boiling temperature. Then eq. (48) is rewritten with the use of  $L_S$  as:

$$\rho C_P \left( \frac{\partial \langle T \rangle}{\partial t} + v_z \frac{\partial \langle T \rangle}{\partial z} \right) = 2 \sqrt{\frac{\pi}{A}} [h(T_\infty - T_0) - L_S N_{S,0}] \quad (49)$$

## FUNDAMENTAL EQUATIONS IN THE STEADY STATE

One of the objects of this series of paper is to see how the changes of residual solvent content, cross-sectional area, and filament temperature along the  $z$  direction are affected by various spinning conditions. This object is satisfied by solving the steady-state equations, in which partial differentiation with respect to  $t$  is taken to be zero and that with respect to  $z$  is replaced by ordinary differentiation. In this section we give the steady-state equations of solvent content, spinning tension, cross-sectional area, and temperature of filament.

### Equation of Solvent Content

In the steady state, the macroscopic eq. (34) of continuity of the polymer component becomes

$$dW_P/dz = 0 \quad (50)$$

After integration we get

$$W_P = C \quad (51)$$

Equation (51) shows that the total mass of polymer which passes across any  $z$  plane per unit time is constant. In other words, the polymer component which comes out of the spinneret is all wound by the wind-up roll.

Therefore, by rewriting the constant of integration  $C$ , we get the steady-state macroscopic equation of continuity for the polymer component:

$$W_P = A(z)v_z(z)\rho\langle\omega_P(z)\rangle \quad (52a)$$

$$= A_N v_N \rho \omega_{P,N} \quad (52b)$$

$$= A_W v_W \rho \quad (52c)$$

where the subscripts N and W designate the positions at  $z = 0$  and the wind-up roll, respectively.

We define at this stage the position  $z_W$  of the wind-up roll by the distance from the nozzle where the evaporation of solvent entirely completes, i.e., where the relation  $\langle\omega_P\rangle = 1$  holds.

From eq. (35) of continuity of the solvent component, we get

$$dW_S/dz = 2\pi T M_S N_{S,0} = 0 \quad (53)$$

On integrating eq. (53) from  $z = 0$  to  $z = z$ , we obtain

$$W_S(z) - W_S(0) + \int_0^z 2\pi R M_S N_{S,0} dz = 0 \quad (54)$$

Since the total mass flow rate  $W_S$  of the solvent component is equal to zero at  $z_W$  by the above definition, the replacement of the upper limit  $z$  of the integration by  $z_W$  yields

$$W_S(0) = \int_0^{z_W} 2\pi R M_S N_{S,0} dz \quad (55a)$$

$$= A_N v_N \rho \omega_{S,N} \quad (55b)$$

Equation (55) shows that the total mass of the solvent component which comes out of the nozzle per unit time is equal to the rate of evaporation from the entire filament surface from  $z = 0$  to  $z = z_W$ .

Thus, the steady-state macroscopic equation for the solvent component becomes

$$W_S(z) = A(z)v_z(z)\rho\langle\omega_S(z)\rangle \quad (56a)$$

$$= A_N v_N \rho \omega_{S,N} - \int_0^z 2\pi R M_S N_{S,0} dz \quad (56b)$$

$$= \int_0^{z_W} 2\pi R M_S N_{S,0} dz \quad (56c)$$

The sum of eqs. (52) and (56) yields the equation of continuity for  $W$ :

$$W(z) = A(z)v_z(z)\rho \quad (57a)$$

$$= A_N v_N \rho - \int_0^z 2\pi R M_S N_{S,0} dz \quad (57b)$$

$$= A_W v_W \rho + \int_0^{z_W} 2\pi R M_S N_{S,0} dz \quad (57c)$$

Now we substitute eq. (48-1) into eq. (46) and carry out the differentiation to get

$$\frac{1}{\langle \omega_P \rangle} \cdot \frac{d\langle \omega_P \rangle}{dz} + \frac{1}{A} \cdot \frac{dA}{dz} + \frac{1}{v_z} \cdot \frac{dv_z}{dz} = 0 \quad (58)$$

Similarly, the substitution of eq. (56a) into eq. (49) yields

$$\frac{1}{\langle \omega_S \rangle} \cdot \frac{d\langle \omega_S \rangle}{dz} + \frac{1}{A} \cdot \frac{dA}{dz} + \frac{1}{v_z} \cdot \frac{dv_z}{dz} + \frac{2\pi R M_S N_{S,0}}{W_S} = 0 \quad (59)$$

On subtracting eq. (59) from eq. (58) and taking eq. (1) into consideration, we obtain the ordinary differential equation of the first order for the average mass fraction of solvent,  $\langle \omega_S(z) \rangle$ :

$$\frac{d\langle \omega_S \rangle}{dz} + \frac{2\sqrt{\pi M_S}}{W_P} \cdot \sqrt{A} N_{S,0} (1 - \langle \omega_S \rangle)^2 = 0 \quad (60)$$

In order to evaluate  $N_{S,0}$  we make the following assumption:

**Assumption 18.** Immediately after the polymer solution comes out of the spinneret, there exists at the surface of filament an equilibrium relationship (61) between the solvent concentration  $\omega_S$  in the liquid phase and the solvent concentration  $x_S$  in the gas phase.

$$x_S = f(\omega_S) \quad (61)$$

The explicit form of eq. (61) is dependent on a given polymer and solvent system and is governed by the activity of solvent. Theoretically, it is given by the Flory-Huggins equation.<sup>11,12</sup>

If we take eq. (61) as given, we finally obtain from eqs. (6), (60), and (61) an ordinary nonlinear differential equation of the first order for the average mass fraction of solvent,  $\langle \omega_S(z) \rangle$ :

$$\frac{d\langle \omega_S \rangle}{dz} + \frac{2\sqrt{\pi M_S} \sqrt{A}}{W_P} \cdot \frac{k_r [f(\omega_{S,0}) - x_{S,\infty}]}{1 - f(\omega_{S,0})} (1 - \langle \omega_S \rangle)^2 = 0 \quad (62)$$

When the molecular weight of solvent  $M_S$ , the spinning conditions  $W_P$ ,  $x_{S,\infty}$ ,  $\omega_{S,N}$ , the mass-transfer coefficient  $k_z(z)$ , the cross-sectional area  $[A(z)]$  and the mass fraction of solvent at the filament surface  $\omega_{S,0}(z)$  are given, eq. (62) can be solved to get the average mass fraction of solvent,  $\langle \omega_S(z) \rangle$ .

The concentration distribution within the filament is given by solving under the boundary conditions (64) and (65) the diffusion eq. (63) which is derived from eq. (13) under the steady-state condition: in other words, if a spinning condition  $[\omega_{S,N}]$ , the surface concentration  $[\omega_{S,0}(z)]$  the velocity  $[v_z(z)]$  and the diffusivity  $D_{PS}(z,r)$  are known, eq. (63) can be solved.

$$v_z \frac{\partial \omega_S}{\partial z} = \frac{1}{r} \cdot \frac{\partial}{\partial r} \left( r D_{PS} \frac{\partial \omega_S}{\partial r} \right) \quad (63)$$

with boundary conditions:

$$\begin{cases} z = 0 & 0 \leq r \leq R_N & \omega_S = \omega_{S,N} \\ z > 0, & r = R(z) & \omega_S = \omega_{S,0}(z) \end{cases} \quad (64)$$

Thus, in order to solve both eqs. (62) and (63), the surface concentration  $\omega_{S,0}(z)$  should be known. A suggested method of solving both equations simultaneously is to determine  $\omega_{S,0}(z)$  point by point so that the average weight fraction of solvent,  $\langle \omega_S(z) \rangle$ , obtained from eq. (62) and from eq. (32) by using the solution of eq. (63) are equal to each other.

### Equations of Spinning Tension and Cross-Sectional Area

The steady-state macroscopic equation of motion is obtained from eq. (37):

$$\frac{dF}{dz} = \frac{d}{dz} (v_z W) - A\rho g + 2\pi R\tau_t \quad (66)$$

Integration of eq. (66) from  $z = z$  to  $z = z_w$  yields the equation of spinning tension  $F(z)$ :

$$F = F_w - \rho(A_w v_w^2 - A v_z^2) + \rho g \int_z^{z_w} A dz - [F_d(z_w) - F_d(z)] \quad (67)$$

where  $F_d(z)$  represents the total drag force acting at the entire surface of the filament from  $z = 0$  to  $z = z$  and is given by the equation

$$F_d(z) = \int_0^z 2\pi R\tau_t dz \quad (68)$$

The physical meaning of each term on the right-hand side of eq. (67) is as follows. The first term  $F_w$  represents the force acting between a filament and a wind-up roll at  $z = z_w$  and is a constant determined by the spinning state. The second term represents the force required to accelerate a filament. The third term is due to the weight of filament hanging from  $z = z$  to  $z = z_w$ . The fourth term arises from the drag force acting at the surface of filament running through the surrounding air. Note that the first and the third terms increase the spinning tension, while the second and the fourth terms decrease it.

When we express by  $F_z$  the terms of eq. (63) which depend on  $z$ , eq. (67) is rewritten to the form

$$F = F_w + F_z \quad (69)$$

where

$$F_z = -\rho(A_w v_w^2 - A v_z^2) + \rho g \int_0^{z_w} A dz - [F_d(z_w) - F_d(z)] \quad (70)$$

The spinning tension, on the other hand, is expressed by eq. (71), which is derived from the rheological eq. (43):

$$F = A\langle\beta\rangle(dv_z/dz) - \gamma\sqrt{\pi A} \quad (71)$$

By combining eq. (71) with eq. (54) and using the relations (1) and (69), we obtain the equation for the change of cross-sectional area:

$$\frac{1}{A} \cdot \frac{dA}{dz} = \frac{1}{1 - \langle\omega_S\rangle} \cdot \frac{d\langle\omega_S\rangle}{dz} - \frac{\rho(1 - \langle\omega_S\rangle)}{W_P\langle\beta\rangle} (F_W + F_z + \gamma\sqrt{\pi A}) \quad (72)$$

The first term on the right-hand side of eq. (72) represents the thinning of cross-sectional area due to the solvent evaporation and the second term represents the thinning due to the elongational flow caused by the internal tension.

On integrating eq. (72) from  $z = 0$  to  $z = z$ , we obtain the equation of cross-sectional area of the form

$$\ln \frac{A(z)}{A_N} = \ln \frac{1 - \omega_{S,N}}{1 - \langle\omega_S\rangle} - \frac{\rho}{W_P} \int_0^z \frac{(1 - \langle\omega_S\rangle)(F_W + F_z + \gamma\sqrt{\pi A})}{\langle\beta\rangle} dz \quad (73)$$

where the constant  $F_W$  is determined by eq. (74) which is derived from eq. (73) by replacing  $z$  by  $z_W$  and solving for  $F_W$ :

$$F_W = \frac{\ln \frac{A_N(1 - \omega_{S,N})}{A_W} - \frac{\rho}{W_P} \int_0^{z_W} \frac{1 - \langle\omega_S\rangle}{\langle\beta\rangle} (F_z + \gamma\sqrt{\pi A}) dz}{\frac{\rho}{W_P} \int_0^{z_W} \frac{1 - \langle\omega_S\rangle}{\langle\beta\rangle} dz} \quad (74)$$

Thus  $F_W$  is a constant which depends on the state of filament. It is seen that the cross-sectional area  $A(z)$  can be calculated from eqs. (1), (52), (73), and (74), when the properties of a polymer and solvent system,  $[\rho, \gamma]$ , the spinning conditions  $[W_P, A_N, A_W, \omega_{S,N}]$  and the values of  $\langle\omega_S(z)\rangle$ ,  $\langle\beta(z)\rangle$ , and  $F_d(z)$  are given.

The flat velocity of filament,  $v_z$ , will be obtained from eq. (28), when  $A(z)$  and  $\langle\omega_P(z)\rangle$  are calculated. The average flow in the  $r$  direction,  $v_r(z, r)$ , resulting from the thinning of a filament, will be calculated from eq. (75), which is obtained by integrating eq. (12) with respect to  $r$  on the assumptions (15) and (16):

$$v = -\frac{1}{2} \cdot r \frac{dv_z}{dz} \quad (75)$$

### Equation of Temperature

In the steady state the macroscopic eq. (49) of energy is rewritten to the form:

$$\rho C_P v_z \frac{d\langle T \rangle}{dz} = 2\sqrt{\frac{\pi}{A}} [h(T_\infty - T_0) - L_S N_{S,0}] \quad (76)$$

Substituting eqs. (1), (52a) and (60) into eq. (76) we get the ordinary differential equation of the first order which gives the average temperature of filament,  $\langle T(z) \rangle$ :

$$\frac{d\langle T \rangle}{dz} = \frac{1}{C_P} \left[ \frac{1 - \langle \omega_S \rangle}{W_P} \cdot 2h\sqrt{\pi A}(T_\infty - T_0) + \frac{L_S}{M_S(1 - \langle \omega_S \rangle)} \cdot \frac{d\langle \omega_S \rangle}{dz} \right] \quad (77)$$

where the first term on the right-hand side of the equation represents the rate of temperature increase due to the heat transfer from the air flow, while the second term represents the rate of temperature decrease due to the loss of enthalpy accompanied by the evaporation of solvent.

When the polymer-solvent system  $[C_P, M_S, L_S]$ , the spinning conditions  $[W_P, T_N, T_\infty]$ , and the values of  $h(z)$ ,  $\langle \omega_S(z) \rangle$ ,  $A(z)$ , and  $T_0(z)$  are given, eq. (76) can be solved to get the average temperature of filament,  $\langle T(z) \rangle$ .

The temperature distribution within the filament is given by solving under the boundary conditions (79) and (80) the steady-state eq. (78) of energy which is derived from eq. (27): in other words, if the properties of the polymer-solvent system  $[C_P, \lambda]$ , a spinning condition  $[T_N]$ , the surface temperature  $[T_0(z)]$ , the velocity  $[v_z(z)]$  are known, eq. (78) can be solved.

$$\rho C_P v_z \frac{\partial T}{\partial z} = \lambda \cdot \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial T}{\partial r} \right) \quad (78)$$

with boundary conditions:

$$z = 0, \quad 0 \leq r \leq R, \quad T = T_N \quad (79)$$

$$z > 0, \quad r = R(z), \quad T = T_0(z) \quad (80)$$

In order to calculate  $T(z, r)$  and  $\langle T(z) \rangle$  from eqs. (77) and (78), a similar method suggested concerning the diffusion eq. (63) will also be applied here.

## CORRELATIONS RELATED TO INTERPHASE TRANSPORT

In order to calculate the equations of solvent content [eq. (62)], cross-sectional area [eq. (73)], spinning tension [eq. (69)], and temperature [eq. (77)], mathematical expressions are required for the quantities related to interphase transport such as skin friction  $\tau_t$ , heat-transfer coefficient  $h$ , and mass-transfer coefficient  $k_r$ .

### Skin Friction

Sakiadis calculated various characteristic boundary-layer parameters by assuming the logarithmic velocity profile for the laminar boundary-layer on the continuous cylinder of radius  $a$  moving with constant velocity

$U$  through the stagnant fluid.<sup>13,14</sup> According to his theory, the friction factor for total drag  $\bar{C}_f$  is given by the form

$$\bar{C}_f = \frac{\Theta}{\pi a^2} \cdot \frac{a}{z} \quad (81)$$

where the dimensionless momentum area  $\Theta/\pi a^2$  is calculated as a function of  $\xi$  which is defined by

$$\xi = 4 \sqrt{\frac{2(z/a)}{\text{Re}}} \quad (82)$$

where  $\text{Re}$  is the Reynolds number based on  $U$  and is given by

$$\text{Re} = 2aU/\nu^* \quad (83)$$

in which  $\nu^*$  denotes the kinematic fluid viscosity. The result is shown in Figure 6 as the relationship between  $\bar{C}_f$  and  $\text{Re}$  for values of  $z/a$  of  $10^3$ ,  $10^4$ , and  $10^5$ .

For the laminar boundary layer on the cylindrical surface of finite length, Glauert and Lighthill present a recommended curve for  $\Theta/\pi a^2$ .<sup>15</sup> This curve coincides with that for the moving continuous cylinder approximately to within 30% in the calculated range of  $\xi$ .

Kase and Matsuo measured the drag force exerted by air stream flowing parallel to a single wire whose value of  $z/a$  is the order of  $10^4$ .<sup>2</sup> The relationship between  $\bar{C}_f$  and  $\text{Re}_P$  was given by the formula

$$\bar{C}_f = 1.24 \text{Re}_P^{-0.81} \quad (84)$$

where

$$\text{Re}_P = 2aU_{P,\infty}/\nu^* \quad (85)$$

in which  $U_{P,\infty}$  represents the velocity of air flowing parallel to the wire. This experimentally obtained value of  $\bar{C}_f$  is about twice that theoretically calculated for  $z/a = 10^4$ . Although more elaborate experiments are necessary for precise discussion, we take for the present eq. (85) as representation of the average friction factor for the laminar boundary layers both on the moving continuous cylinder surface and on the cylindrical surface of finite length in the range of Reynolds number between 1 and  $10^2$  and  $z/a$  between  $10^3$  and  $10^5$  (see Fig. 6).

The average skin friction  $\bar{\tau}_f$  is related to  $\bar{C}_f$  by the equation

$$\bar{C}_f = \frac{\bar{\tau}_f}{\frac{1}{2} \rho^* U_{P,\infty}^2} \quad (86)$$

where  $\rho^*$  denotes the fluid density.

From eqs. (84), (85), and (86),  $\bar{\tau}_f$  is expressed in terms of  $a$  and  $U_{P,\infty}$  as:

$$\bar{\tau}_f = 0.353 \rho^* \nu^{*0.81} a^{-0.81} U_{P,\infty}^{1.19} \quad (87)$$

Hence, when the air flows with velocity  $U_{P,\infty}$  parallel to the moving cylinder of velocity  $U$ , the average skin friction  $\bar{\tau}_f$  on the cylindrical sur-

face may be given by replacing  $U_{P,\infty}$  in Eq. (82) by the relative velocity  $|U \pm U_{P,\infty}|$ . Therefore, eq. (87) for the countercurrent flow will be given by eq. (88a)

$$\bar{\tau}_f = 0.353\rho^*\nu^{*0.81}a^{-0.81}(U + U_{P,\infty})^{1.19} \quad (88a)$$

and for the co-current flow by eq. (88b)

$$\bar{\tau}_f = \pm 0.353\rho^*\nu^{*0.81}a^{-0.81}|U - U_{P,\infty}|^{1.19} \quad (88b)$$

where the positive sign is for  $U > U_{P,\infty}$  and the negative sign is for  $U < U_{P,\infty}$ .

### Heat-Transfer Coefficient $h$

Sano et al.<sup>16</sup> measured experimentally heat-transfer coefficients of a fine wire in air flow. Their experiments covered the following conditions: (1) heat transfer by forced convective flow parallel to both fixed and moving wires, (2) heat transfer of a fine wire traveling through stagnant air, and (3) free convection heat transfer from a vertical wire. On the basis of the experimental results they discussed the heat-transfer coefficients of filament in spinning processes where these three sources of heat transfer are superposed.<sup>17,18</sup> Their conclusion is that in usual dry spinning conditions the effect of forced convection is dominant and that entrained and free convective air flow are expected to increase the Nusselt number by 20% at most.

For the forced convection heat transfer they presented the following expressions.

For parallel air flow

$$\text{Nu} = 0.35 + 0.146\text{Re}_P^{0.50} \quad (89)$$

for cross air flow

$$\text{Nu} = 0.35 + 0.50\text{Re}_C^{0.50} \quad (90)$$

Here,

$$\text{Nu} = 2ah/\lambda^* \quad (91)$$

$$\text{Re}_C = 2aU_{C,\infty}/\nu^* \quad (92)$$

and  $\lambda^*$  and  $U_{C,\infty}$  denote the thermal conductivity of flowing air and the velocity of cross air flow, respectively.

Kase and Matsuo also made experiments on forced convection heat transfer and obtained similar results.<sup>1</sup> In the Reynolds number ranging from 1 to  $10^2$ , they approximated eqs. (89) and (90) by eqs. (93) and (94) for parallel air flow and for cross air flow, respectively,

$$\text{Nu} = 0.42\text{Re}_P^{1/3} \quad (93)$$

$$\text{Nu} = 0.84\text{Re}_C^{1/3} \quad (94)$$

We use these formulae for writing heat-transfer coefficients. Then the expressions for these coefficients are given for parallel air flow from eqs.

(85), (91) and (93) and for cross air flow from eqs. (91), (92), and (94) are given by eqs. (95) and (96) for parallel air flow and for cross air flow, respectively.

$$h = 0.26(\lambda^* \nu^{*-1/3}) a^{-2/3} U_{P,\infty}^{1/3} \quad (95)$$

$$h = 0.52(\lambda^* \nu^{*-1/3}) a^{-2/3} U_{C,\infty}^{1/3} \quad (96)$$

### Mass-Transfer Coefficient $k_x$

As far as we know, experimental data for the mass-transfer coefficient of a fine cylinder seem to be lacking for the condition encountered in the dry spinning process in which the Reynolds number  $Re$  usually ranges from 1 to 10 and the Schmidt number  $Sc$  from 0.5 to 2.0. However, it is known that the analogy between heat and mass transfer permits us to derive mass-transfer correlations from heat-transfer correlations for equivalent boundary conditions by simply replacing the Nusselt number  $Nu$  by the Sherwood number  $Sh$  and the Prandtl number  $Pr$  by the Schmidt number  $Sc$ .<sup>7</sup> It is reasonable, therefore, to apply this analogy to the dry spinning process and to predict the mass-transfer coefficient from the above mentioned experimental results on heat transfer of a fine wire.

$$Nu = \text{Const. } Re^m Pr^n \quad (97)$$

is used for heat transfer in certain regions of  $Re$  and  $Pr$ .<sup>19</sup>

In eq. (97),  $Pr$  is given by

$$Pr = C_P^* \mu^* / \lambda^* \quad (98)$$

where  $C_P^*$  denotes the specific heat of flowing gas at constant pressure.

The corresponding correlation for mass transfer is

$$Sh = \text{Const. } Re^m Sc^n \quad (99)$$

where

$$Sh = 2ak_r / c^* D_{AS}^* \quad (100)$$

and

$$Sc = \mu^* / \rho^* D_{AS}^* \quad (101)$$

where  $c^*$  denotes molar density of flowing gas and  $D_{AS}^*$  denotes the mass diffusivity for the solvent vapor and air system.

From eqs. (97) and (99) we have

$$Sh/Nu = (Sc/Pr)^n \quad (102)$$

In the range of  $Pr$  and  $Sc$  numbers between 0.5 and 2.5, a value of  $n = 0.5$  is suggested for both heat and mass transfer:<sup>19</sup>

$$Sh/Nu = (Sc/Pr)^{1/2} \quad (103)$$

From eqs. (91), (100), and (103), the mass-transfer coefficient  $k_x$  is expressed by

$$k_x = (c^* D_{AS}^* / \lambda^*) (Sc/Pr)^{1/2} h \quad (104)$$

Hence, on substituting eqs. (95) and (96) into eq. (104), we obtain the

expressions for the mass-transfer coefficient of a fine cylinder for parallel flow and cross flow, respectively.

For parallel flow:

$$k_x = 0.26(c^*D_{BS}^*\nu^{*-1/3})(Sc/Pr)^{1/2}a^{-2/3}U_{P,\infty}^{1/3} \quad (105)$$

For cross flow:

$$k_x = 0.52(c^*D_{AS}^*\nu^{*-1/3})(Sc/Pr)^{1/2}a^{-2/3}U_{C,\infty}^{1/3} \quad (106)$$

It will not be worthless to briefly examine here whether the analogy between momentum and heat transfer holds or not. The Chilton-Colburn  $j$  factor defined in eq (107a) for heat transfer can be calculated from eq. (107b), which is derived from eq. (93), when air flows parallel to a fine cylinder:

$$j_H = NuRe^{-1}Pr^{-1/3} \quad (107a)$$

$$= 0.42Re^{-2/3}Pr^{-1/3} \quad (107b)$$

In Figure 6 a curve for  $2j_H$  is shown with a dotted line, by putting  $Pr$  equal to 0.70. It is seen that the curve for  $2j_H$  approximately coincides with that for  $\bar{C}_f$ . This indicates that the Chilton-Colburn analogy holds and suggests that the mechanisms of momentum and heat transfer may be the same. For more precise discussions, however, the accumulation of experimental data will be necessary.

### Correlations Applied to the Dry Spinning Process

It is necessary to make the following assumption in order to apply the correlations for interphase transport concerning a fine cylinder having a constant radius  $a$  and running with constant velocity  $U$  to the condition where a radius changes (therefore, running velocity changes) along its length as is seen with filament in dry spinning.

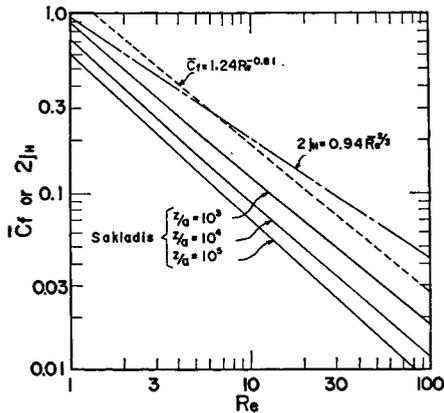


Fig. 6. Momentum and heat-transfer correlations.

**Assumption 19.** The correlations related to interphase transport of filament at  $z$  in dry spinning can be approximated by the correlations which would be given if the filament traveled from the spinneret to  $z$  with constant velocity  $v_z(z)$  and a constant radius  $R(z)$  which the filament possesses at  $z$ .

In other words, the correlations at  $z$  would be given by replacing  $a$  by  $R(z)$  and  $U$  by  $v_z(z)$ , where  $R(z)$  and  $v_z(z)$  are the quantities which the filament actually has at  $z$ .

Thus, if we may assume that the skin friction is approximated by eq. (84) for any given value of  $z/a$ , the skin friction at  $z$ ,  $\tau_f(z)$ , will be expressed by

$$\tau_f(z) = \pm 0.353 \rho^* \nu^{*0.81} R(z)^{-0.81} |v_z(z) \pm U_{P,\infty}|^{1.19} \quad (108)$$

where the sign convention is the same as in eq. (87). Similarly the heat-transfer coefficients at  $z$  will be as follows:

For parallel flow:

$$\begin{aligned} h(z) &= 0.26(\lambda^* \nu^{*-1/3}) R(z)^{-2/3} U_{P,\infty}^{1/3} \\ &= 0.39(\lambda^* \nu^{*-1/3}) A(z)^{-2/3} U_{P,\infty}^{1/3} \end{aligned} \quad (109)$$

For cross flow:

$$\begin{aligned} h(z) &= 0.52(\lambda^* \nu^{*-1/3}) R(z)^{-2/3} U_{C,\infty}^{1/3} \\ &= 0.78(\lambda^* \nu^{*-1/3}) A(z)^{-2/3} U_{C,\infty}^{1/3} \end{aligned} \quad (110)$$

The mass-transfer coefficients at  $z$  will be as follows.

For parallel flow:

$$\begin{aligned} k_z &= 0.26(c^* D_{AS}^* \nu^{*-1/3}) (Sc/Pr)^{1/2} R(z)^{-2/3} U_{P,\infty}^{1/3} \\ &= 0.39(c^* D_{AS}^* \nu^{*-1/3}) (Sc/Pr)^{1/2} A(z)^{-2/3} U_{P,\infty}^{1/3} \end{aligned} \quad (111)$$

For cross flow:

$$\begin{aligned} k_z &= 0.52(c^* D_{AS}^* \nu^{*-1/3}) (Sc/Pr)^{1/2} R(z)^{-2/3} U_{C,\infty}^{1/3} \\ &= 0.78(c^* D_{AS}^* \nu^{*-1/3}) (Sc/Pr)^{1/2} A(z)^{-2/3} U_{C,\infty}^{1/3} \end{aligned} \quad (112)$$

From the above formulae it is seen that the skin friction  $\tau_f(z)$ , the heat-transfer coefficient  $h(z)$ , and the mass-transfer coefficient  $k_z(z)$  are calculated, when the physical constants for air flow [ $\rho^*$ ,  $\nu^*$ ,  $\lambda^*$ ,  $c^*$ ,  $D_{AS}^*$ ], the spinning conditions [ $U_{P,\infty}$  or  $U_{C,\infty}$ ] and the state of the filament [ $v(z)$ ,  $R(z)$ , or  $A(z)$ ] are given.

To sum up, when various physical constants for a given polymer and solvent system are measured as a function of temperature and concentration, the procedure of calculating the steady state of filament for a given spinning condition is as follows. First, the concentration distribution of solvent  $\omega_S(z,r)$  within the filament is obtained by solving the eqs. (52)

and (63) simultaneously. For this aim the knowledge on the quantities  $k_z(z)$ ,  $A(z)$ ,  $v_z(z)$ ,  $D_{PS}(z,r)$ , and  $f(\omega_{S,0}(z))$  are required. Second, the spinning tension  $F(z)$  and the cross-sectional area  $A(z)$  are calculated from eqs. (69) and (73) if  $\tau_i(z)$ ,  $\langle\beta(z)\rangle$ ,  $\langle\omega_S(z)\rangle$ , and  $v_z(z)$  are known. Finally, the temperature distribution  $T(z,r)$  within the filament is obtained from eqs. (77) and (78) based on the knowledge of  $h(z)$ ,  $A(z)$ ,  $v_z(z)$ , and  $\langle\omega_S(z)\rangle$ . Among the above quantities the filament velocity  $v_z(z)$  is connected with  $A(z)$  and  $\langle\omega_S(z)\rangle$  by eq. (52). Therefore, the quantities  $D_{PS}(z,r)$ ,  $\langle\beta(z)\rangle$ , and  $f(\omega_{S,0}(z))$  related to physical properties of the system and the quantities  $\tau_i(z)$ ,  $h(z)$ , and  $k_z(z)$  related to interphase transport are calculated when  $\omega_S(z,r)$ ,  $A(z)$ , and  $T(z,r)$  are given.

Since the quantities  $\omega_S(z,r)$ ,  $A(z)$ , and  $T(z,r)$  are mutually included in the equations of solvent content, spinning tension, cross-sectional area, and temperature, it is concluded that these equations should be solved simultaneously. The examples of calculation will be given in the subsequent series of this study.

### CONCLUSION

The fundamental equations for dry spinning were obtained by making clear the assumptions made.

(1) The nonsteady-state equations were obtained by considering both microscopic and macroscopic balances of mass, momentum and energy on the assumption of axial symmetry.

(2) It was shown that the macroscopic equations can be simplified to a great extent by assuming a flat velocity profile.

(3) The equations of average solvent content  $\langle\omega_S(z)\rangle$ , spinning tension  $F(z)$ , cross-sectional area  $A(z)$ , and average temperature  $\langle T(z)\rangle$  of filament were derived by modifying the macroscopic equations on condition of steady state.

When the spinning conditions are given and the physical constants are measured for a given polymer and solvent system, it becomes possible to calculate the state of filament in dry spinning by solving simultaneously the microscopic and macroscopic equations.

Since the spinning conditions [ $W_P$ ,  $\omega_{S,N}$ ,  $A_N$ ,  $A_W$ ,  $T_N$ ,  $T_\infty$ ,  $x_{S,\infty}$ ,  $U_{P,\infty}$ ,  $U_{C,\infty}$ ] are independently selectable, it is possible to calculate the behavior of filament for any given spinning condition composed of an arbitrary combination of these parameters without resorting to actual spinning.

Among the physical constants of polymer solution important are such as the mass diffusivity, the activity of solvent and the Trouton viscosity. All these are functions of both composition and temperature. Since the filament composition varies from dope composition to pure polymer and the filament temperature from wet-bulb temperature to air temperature, it is desired to make the measurements on the constants which cover these ranges of composition and temperature. The important constant concerning the movement of solvent vapor is the mass diffusivity in solvent vapor and air system.

It is noteworthy that if the concentration of solvent is made equal to zero, the equations for dry spinning entirely coincide with those for melt spinning which were previously derived.<sup>1</sup> Furthermore, by applying the concept stated in this paper it may become possible to derive equations for wet spinning which includes more than two species of material and accompanies chemical reactions.

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