



Theoretical and experimental investigation of physical mechanisms responsible for polymer nanofiber formation in solution blowing



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ARTICLE INFO

Article history:

Received 17 September 2014

Received in revised form

30 October 2014

Accepted 5 November 2014

Available online 15 November 2014

Keywords:

Solution blowing

Nanofiber

Numerical modeling

ABSTRACT

This work describes a comprehensive numerical model of solution blowing process of multiple three-dimensional polymer jets issued from a die nosepiece into a high-speed air flow and deposited onto a moving screen. The model solves the quasi-one-dimensional equations of the mechanics of free liquid jets with the jet axis configuration being three-dimensional. It accounts for the polymer solution viscoelasticity, jet interaction with the surrounding high-speed air flow, and solvent evaporation and jet solidification. The results include the polymer jet configurations in flight as well the detailed information on the pattern in which the oncoming polymer jets are deposited on the moving screen (the so-called lay-down), and its characteristics, in particular, the fiber-size distributions obtained under different conditions. The work also describes experiments on solution blowing and comparison of the numerical and experimental data.

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1. Introduction

Solution blowing is kindred to meltblowing. In solution blowing polymer solution is issued as a slow jet into a co-flowing sub- or supersonic gas jet which stretches the polymer jet directly, as well as via a vigorous bending instability, thus leading to formation of polymer nanofibers. In meltblowing molten polymer jet undergoes similar transformations in co-flowing subsonic gas jet, which results in formation of polymer microfibers. For several decades meltblowing, which is more mature than solution blowing, gained immense popularity in nonwoven industry, which used several thermoplastic polymers to form nonwoven mats of polymer microfibers in the size range of 1–100 μm [1–4]. In distinction to meltblowing which results in microfibers, solution blowing results in nanofibers. It does it with the production rate superior to electrospinning and with much less restrictions on the electrical parameters of polymer solutions [5–7]. Solution blowing has been used to form polymer nanofiber mats for different applications, such as nanotubes [6,8], microfiltration [9], and biomedical

applications [10,11]. Successful applications of solution blowing in forming nanofibers from various biopolymers were recently demonstrated [11–14].

Recent theoretical/numerical and experimental results shed light on the physical mechanisms responsible for microfiber formation in meltblowing and provided their detailed description [15–17]. The situation with solution blowing is different: until now it has been lacking a theoretical description. Since solution blowing is related to meltblowing, and in general, to free liquid jets moving relative to the surrounding gas with high speed, it is worth of mentioning some of these works as an appropriate context for the present work [7,18]. Highly viscous liquid jets moving with a high speed relative to the surrounding gas experience lateral distributed force which tends to increase bending perturbations, as was revealed in the seminal work [19]. A general theory of thin liquid jets moving in air applicable to both Newtonian and non-Newtonian liquids, including viscoelastic polymer solutions and melts, was given in Refs. [18,20]. It was already applied to describe electrospinning [21,22] and meltblowing [15–17]. This approach is extended in the present work to incorporate solution blowing.

The present article discusses the theoretical aspects in Section 2. Section 3 is devoted to the description of the experimental setup. The results are presented and discussed in Section 4, and the conclusions are drawn in Section 5.

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2. Theoretical

2.1. Straight part of the jet

The experimental data discussed below show that polymer solution jets in solution blowing possess a straight part where the cross-sectional diameter of the jet is still large enough to prevent significant bending perturbations. The jet is straight, pulled in the axial direction by the surrounding high-speed gas flow and attenuating in response. This happens on the background of solvent evaporation. Following Ref. 22, the mass of an unperturbed element of unit length in the straight part of the jet decreases according to the following equation

$$\frac{d(fV)}{dx} = -h_m [C_{s,eq}(T) - C_{s,\infty}] 2\pi a \quad (1)$$

Here x is the axial coordinate reckoned along the straight jet axis, f is the area of a jet cross-section which is assumed to be circular, with a being its radius, V is the absolute axial jet velocity, h_m is the mass transfer coefficient, $C_{s,eq}(T)$ and $C_{s,\infty}$ are the solvent vapor volume fractions at the jet surface and far away from it, respectively, T is temperature which is the same for polymer solution and the surrounding air in the case of solution blowing, which is assumed to be an isothermal process.

According to Ref. [22], the mass transfer coefficient is given by the following expression

$$h_m = \frac{D_a}{2a} 0.495 \text{Re}^{1/3} \text{Sc}^{1/2} \quad (2)$$

where $\text{Re} = V2a/\nu_a$ and $\text{Sc} = \nu_a/D_a$ are the Reynolds and Schmidt numbers, respectively, with ν_a being the kinematic viscosity of air and D_a being the solvent vapor diffusion coefficient in air.

The momentum balance in the straight part of the jet reads [7,18]

$$\rho \frac{d(fV^2)}{dx} = \frac{d(\sigma_{xx}f)}{dx} + q_\tau \quad (3)$$

where ρ is the polymer solution density, σ_{xx} is the longitudinal stress in the jet, and q_τ is the aerodynamic drag pulling the jet in the axial direction, which is given by the following expression [23,24]

$$q_\tau = c\pi a \rho_a (U_a - V)^2 \left[\frac{2(U_a - V)a}{\nu_a} \right]^{-0.81} \quad (4)$$

In Eq. (4) c is an empirical constant discussed below, ρ_a is the air density, and U_a is the absolute velocity of air in the axial direction. It should be emphasized that the empirical Kase–Matsuo Eq. (4) incorporates the dependence of the aerodynamic drag on the Reynolds number based on the relative velocity, $\text{Re}_{\text{relative}} = 2(U_a - V)a/\nu_a$, as $\text{Re}_{\text{relative}}^{-0.81}$, which is characteristic of the turbulent boundary layer (in distinction from the laminar boundary layer where the dependence $\text{Re}_{\text{relative}}^{-0.5}$ would be expected [25–27]).

In addition, according to Refs. [7] and [18], the longitudinal stress σ_{xx} is equal to the difference of the axial and radial deviatoric stresses, τ_{xx} and τ_{yy} , respectively, i.e. $\sigma_{xx} = \tau_{xx} - \tau_{yy}$. In flows with strong uniaxial elongation, such as in solution blowing, the radial component τ_{yy} is negligibly small compared to the axial one τ_{xx} , and $\sigma_{xx} \approx \tau_{xx}$ [16]. In the uniaxial elongational flows, such as the electrospinning and meltblown jets, a plausible description of the rheological behavior of polymer solutions and melts is given by the upper-convected Maxwell model (UCM) [7,15–18,21,22]

$$V \frac{d\tau_{xx}}{dx} = 2 \frac{dV}{dx} + 2 \frac{\mu}{\theta} \frac{dV}{dx} - \frac{\tau_{xx}}{\theta} \quad (5)$$

where μ and θ are the viscosity and the relaxation time of polymer solution, respectively.

Eqs. (1), (3) and (5) can be reduced to the following system of two differential equations for two unknowns V and τ_{xx}

$$\frac{dV}{dx} = \frac{\rho\kappa V - (\tau_{xx}/V)(\kappa + f/\theta) + q_\tau}{\rho_0 f_0 V_0 - \rho\kappa x - (f/V)(\tau_{xx} + 3\mu/\theta)} \quad (6)$$

$$\frac{d\tau_{xx}}{dx} = \frac{1}{V} \left[2 \frac{dV}{dx} (\tau_{xx} + \mu/\theta) - \tau_{xx}/\theta \right] \quad (7)$$

In these equations the jet cross-section f is given by

$$f = \frac{1}{V} (f_0 V_0 - \kappa x) \quad (8)$$

where, as in Eq. (6) and hereinafter, subscript 0 denotes the values in the initial cross-section which are given. Also, the constant κ is given by

$$\kappa = \frac{D_a}{2a} 0.495 \text{Re}^{1/3} \text{Sc}^{1/2} [C_{s,eq}(T) - C_{s,\infty}] 2\pi \quad (9)$$

In addition, due to solvent evaporation the viscosity μ and relaxation time θ vary with the polymer volume fraction C_p along the jet as [7,22]

$$\mu = \mu_0 10^{B(C_p^m - C_{p0}^m)}, \quad \theta = \theta_0 \frac{C_p}{C_{p0}} \quad (10)$$

where B and m , μ_0 and θ_0 are the material parameters.

It should be emphasized that the loss of mass from the jet is associated only to the solvent evaporation, and thus similarly to Eq. (1) the solvent volume in a unit jet length $M_s = C_s f V$ is subjected to the following equation

$$\frac{d(C_s f V)}{dx} = -h_m [C_{s,eq}(T) - C_{s,\infty}] 2\pi a \quad (11)$$

where C_s is the volume ratio of solvent.

Then, integrating Eq. (11), we find C_s as

$$C_s = \frac{C_{s0} f_0 V_0 - \kappa x}{f V} \quad (12)$$

Therefore, the polymer volume fraction $C_p = 1 - C_s$ is found using Eqs. (12) and (8) as

$$C_p = \frac{C_{p0}}{1 - \kappa x / (f_0 V_0)} \quad (13)$$

The latter equation provides us with the values of C_p needed to find the viscosity and relaxation time using Eq. (10).

The system of Eqs. (6) and (7) is solved numerically using the Kutta–Merson method with the boundary conditions

$$x = 0: \quad V = V_0, \quad \tau_{xx} = \tau_{xx0} \quad (14)$$

where the values of the polymer feeding velocity V_0 and the longitudinal stress τ_{xx0} at the nozzle exit are determined by the flow in the nozzle and are, in principle known.

2.2. Perturbed part of the jet

The general quasi-one-dimensional theory of free liquid jets moving in air provides us with two inter-related types of approaches to problems dealing with jet bending [7,18]. If the jet is

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