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Numerical simulation of polypropylene foaming process assisted by carbon dioxide: Bubble growth dynamics and stability

Yan Li [b], Zhen Yao [b,*], Zhen-hua Chen [c,d], Kun Cao [a,b,**], Shao-long Qiu [b], Fang-jun Zhu [b], Changchun Zeng [c,d,***], Zhi-ming Huang [b]

[a] State Key Laboratory of Chemical Engineering, Zhejiang University, Hangzhou 310027, China
[b] Institute of Polymerization and Polymer Engineering, Department of Chemical and Biological Engineering, Zhejiang University, Hangzhou 310027, China
[c] High Performance Materials Institute, Florida State University, Tallahassee, FL 32310, USA
[d] Department of Industrial and Manufacturing Engineering, FAMU-FSU College of Engineering, Tallahassee, FL 32310, USA

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ABSTRACT

A mathematical model was established to simulate the bubble growth process during foaming of polypropylene (PP) by carbon dioxide, taking into account of a wide range of physical and rheological properties (solubility, diffusivity, surface tension, long-chain branching, zero shear viscosity, relaxation time, strain hardening), as well as processing conditions. By employing the Considère construction the possibility of growth instability and bubble rupture at later stage of bubble growth was predicted. The simulation revealed that the improvement of foamability of polypropylene by introducing long-chain branching was due to the well-defined viscoelastic characteristics of the melt. Rheological factors that impede bubble growth are beneficial in stabilizing the bubble growth. Stability during bubble growth is further facilitated by moderate strain hardening characteristics and elastics properties of the polymers. The diffusivity and solubility characteristics also have profound impact on the bubble growth stability, while the influence of the surface tension is negligible.

1. Introduction

With the progress of research work on high melt strength polypropylene (HMSPP) to address the foamability issue, there is increasing amount of interest in using polypropylene (PP) as a foams material. Melt strength may be improved by using polymers with broad or bimodal molecular weight distributions. However, this approach has been shown not effective for PP (Sugimoto et al., 2001). The main and efficient method to prepare HMSPP is to incorporate long-chain branch (LCB) by chemical modification (Auhl et al., 2004; Borsig et al., 2008; Cao et al., in press; Mabrouk et al., 2009; Yao et al., 2009; Ye et al., 2004). Many researchers have carried out experimental investigation on the effect of LCB on foamability of PP. Nam et al. (2005) modified PP by introducing the LCB onto the backbone and studied the effects on the performance of foaming process. They found that the LCB structure of PP was the main factor affecting the foam density. Stange and Munstedt (2006) investigated blends of a linear PP (L-PP) and a long-chain branched polypropylene (LCBPP). They found that a small amount of LCBPP into L-PP could significantly improve the foaming process. Spitael and Macosko (2004) studied the strain hardening in PP and its role in extrusion foaming. They reported that the strain hardening in LCBPP could retard cell coalescence. Recently, Zhai et al. (2008) investigated the foaming behavior of linear and branched PP. They pointed out that the LCBPP exhibited well-defined close cell structure comparing with L-PP due to the increased melt strength. These experimental studies demonstrate that the presence of LCB can improve the foamability of PP and the strain hardening plays an important role in stabilizing the cellular structure during the foaming process.

Furthermore, a great deal of numerical studies was also devoted to better understand the PP foaming process. Otsuki and Kanai (2005) carried out numerical simulations for five kinds of L-PP. They used the Phan-Thien Tanner (PTT) constitutive equation to describe the rheological characteristics of the polymers. The nonlinear parameter, on which the strain hardening property strongly depends, was varied to analyze the effects of the linear and nonlinear viscoelastic characteristics on bubble growth during isothermal extrusion foaming. It was found that the linear viscoelastic characteristics were more influential and

* Corresponding author.
** Corresponding author at: State Key Laboratory of Chemical Engineering, Zhejiang University, Hangzhou 310027, China.
*** Corresponding author at: Department of Industrial and Manufacturing Engineering, FAMU-FSU College of Engineering, Tallahassee, FL 32310, USA.
E-mail addresses: yaozhen@zju.edu.cn (Z. Yao), kcao@che.zju.edu.cn (K. Cao), zeng@eng.fsu.edu (C. Zeng).

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the strain hardening had little effect on bubble growth rate. Den Doelder et al. (2002) investigated the nonisothermal foaming process for a L-PP and a LCBPP. Contrary to Otsuki and Kanai (2005), they concluded that the strain hardening should be one of the most important factors that stabilized the bubble growth.

All previous numerical studies on bubble growth were based on the assumption that the bubbles could grow steadily until thermodynamic equilibrium was achieved between the bubbles and the melt (Mao et al., 2006). That was to say, the melt extension was always stable in bubble growth.

In this paper, extensional flow instability of the melt in bubble growth and bubble rupture were simulated. Rheological data of five different model PPs were first used in the simulation to investigate the influences of the macromolecular architecture (branch ratio) on the bubble growth and rupture in the foaming process. This was followed by extensive analysis of individual rheological parameters, and some additional physical properties of the polymer–gas system and processing conditions, and their effect of bubble growth dynamics and stability.

2. Bubble growth model

The simulation of bubble growth and stability is based on the well-known cell model (Arefmanesh and Advani, 1991; Leung et al., 2006; Shafi et al., 1996; Venerus et al., 1998; Venerus, 2001; Venerus, 2003; Tuladhar and Mackley, 2004). The cell model approximates the situation in polymer foaming by considering the growth of a single bubble with a shell of polymer–gas solution. A schematic is shown in Fig. 1. The following assumptions are used in our model:

(1) The system is isothermal.
(2) The bubble is spherically symmetric.
(3) The polymer melt is incompressible.
(4) The inertial forces and gravity are neglected.
(5) The gas concentration in the polymer–gas solution and the pressure inside the gas bubble obeys Henry's law.

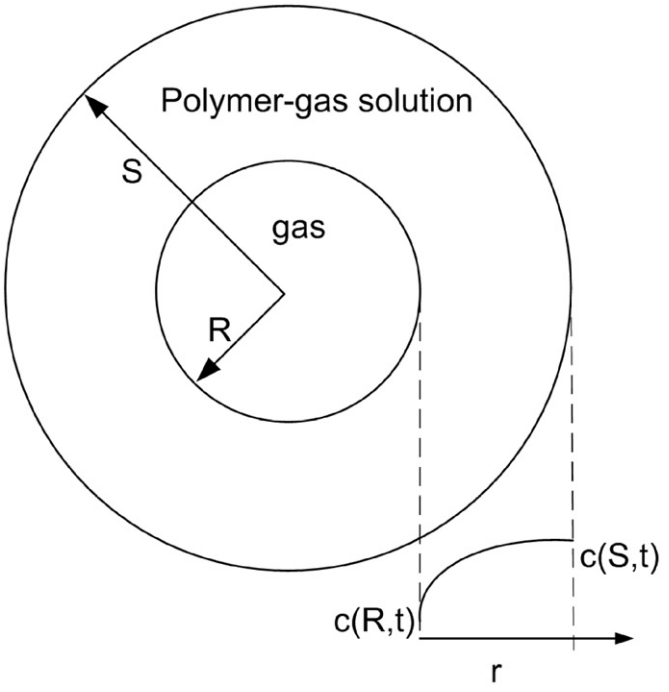

Fig. 1. Schematic of the cell model.

(6) The effect of plasticization is ignored
(7) There is no loss of gas to the surroundings

2.1. Mathematical formulations (governing equations)

Using the cell model, the bubble growth can be calculated by simultaneous solving the mass and momentum transfer between the bubbles and the surrounding polymer–gas shell, typically in a spherical coordinate.

From Arefmanesh and Advani (1991) and Amon and Denson (1984) the kinematics of spherical bubble growth are ideally described by a purely radial velocity field, which is obtained from the continuity equation as

$$v_r = \frac{\dot{R}R^2}{r^2} - \frac{1}{3}\left(\frac{d\ \ln\ \rho}{dt}\right)\left(r - \frac{R^3}{r^2}\right) \tag{1}$$

where r is the radial position, v_r is the radial velocity of the fluid at r position, $\dot{R}$ is the radial velocity at the interface between the gas and the melt. Assuming incompressible flow the above equation reduces to

$$v_r = \frac{\dot{R}R^2}{r^2} \tag{2}$$

The deformation rate tensor derived from the above equation (Amon and Denson, 1984) indicates that it is a biaxial extension flow:

$$\underline{\underline{d}} = \begin{pmatrix} -2 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \frac{\dot{R}R^2}{r^3} \tag{3}$$

Using this equation together with the assumption of negligible inertia and gravitational force (assumption (4)), the *momentum equation* is written as

$$P_g - P_a - \frac{2\gamma}{R} + 2\int_R^s \frac{\tau_{rr} - \tau_{\theta\theta}}{r}\ dr = 0 \tag{4}$$

where γ is the surface tension, P_g and P_a are the bubble and system pressure, respectively, τ_{rr} and $\tau_{\theta\theta}$ are the stress components.

A more accurate calculation of the gas phase pressure is conducted to take into account the non-ideal behavior of the CO_2 gas under elevated pressure during foaming. Thus the Peng–Robinson (PR) cubic equation of state is employed in polynomial form:

$$Z^3 - (1-B)Z^2 + (A - 2B - 3B^2)Z - (AB - B^2 - B^3) = 0 \tag{5}$$

where

$$Z = \frac{PV}{\Re T} \tag{6}$$

$$A = \frac{aP_g}{\Re^2 T^2} \tag{7}$$

$$B = \frac{bP_g}{\Re T} \tag{8}$$

$$a = \frac{0.45724\Re^2 T_c^2 \alpha}{P_c} \tag{9}$$

$$b = \frac{0.07780\,\Re T_c}{P_c} \tag{10}$$

$$\alpha = [1 + k(1 - T_r^{0.5})]^2 \tag{11}$$

$$T_r = \frac{T}{T_c} \tag{12}$$

$$k = 0.3746 + 1.54226\,\omega - 0.26992\,\omega^2 \tag{13}$$