



Validated modeling of bubble growth, impingement and retraction to predict cell-opening in thermoplastic foaming



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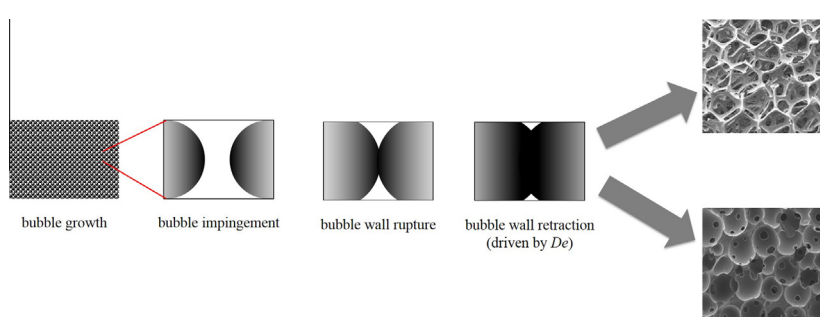
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HIGHLIGHTS

- The sequence of operations involved in gas foaming process is modeled.
- The dynamics of bubble growth and impingement is used to predict rupture and retraction.
- A new retraction criterion is proposed for bubble wall opening.
- The model is validated with poly(ϵ -caprolactone) foamed with CO₂.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 20 July 2015

Received in revised form 5 October 2015

Accepted 6 November 2015

Available online 17 November 2015

Keywords:

Foaming
Open-cell
Modeling
Elastic recovery

ABSTRACT

In this work a design tool to control cell-opening in gas foaming of thermoplastic polymers is developed. The sequence of events following bubble nucleation, namely, bubble growth and impingement, is modeled to gain a comprehensive, perspective view on the mechanisms of bubble wall rupture and on the conditions for achieving a fully open-cell morphology. In particular, unlike the previously published literature, the polymer elastic recovery is recognized as an important factor for wall retraction, which is typically considered as solely driven by surface tension. The new approach is experimentally validated on poly(ϵ -caprolactone) (PCL), foamed with CO₂, as a model polymer/gas system.

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1. Introduction

Due to their mass and energy transport, acoustic absorption, catalytic, impact, and cushioning properties, open-cell polymeric foams are used in a multitude of different applications, including transportation, construction, packaging, food, extraction and separation [1], as well as in leisure and sport. They are also used

in tissue engineering, as culture substrates for living cells [2], and as templating structures for ceramic and metal foams.

The *gas foaming technology*, which makes use of a physical blowing agent (e.g., carbon dioxide or nitrogen) to form bubbles in a softened polymer, is the most used process for the making of open-cell foams, mainly because of its high productivity. The sequence of operations involved in the gas foaming technology, specifically for the case of open-cell foams, is: (i) blowing gas solubilization at high pressure (to achieve a polymer/gas solution); (ii) bubble nucleation induced by an instantaneous pressure quench; (iii) bubble growth; (iv) bubble impingement (where the growing bubbles start

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“feeling” each other, and the polymeric layer separating them progressively thins); and (v) bubble wall rupture [1].

In the last decades, a large number of scientific papers has dealt with the modeling of the above steps, both in order to understand the underlying physico-chemical phenomena and to optimize process conditions. Today, step (i) is quite well understood, in particular how to form the polymer/gas solutions and how much the presence of the solubilized gas affects the thermal [3], mass transfer [4], sorption [5], rheological [6–10], interfacial and volumetric properties [11] of the polymer. More recently, the specific interactions occurring between the polymer and the gas molecules have been considered, opening the way to the design of novel blowing agents [12,13].

Moving to step (ii) above, gas supersaturation is induced by imposing a rapid pressure drop on the polymer/gas solution. The subsequent bubble nucleation is typically modeled by the classical nucleation theory, where the original formulation, developed for water vapor droplets, is adapted to foaming to take into account gas solubility and the high molecular weight of the expanding material [14–16].

Bubble growth (step (iii)) is modeled by mass and momentum (and, in a few cases, also energy) balance equations. Recently, taking advantage of the improved calculating tools, the standard, purely viscous constitutive equations have been substituted by more realistic equations for viscoelastic fluids [17–20].

In some cases, in order to gain some predictive capability on the final foam structure, the nucleation and growth steps are coupled by the so-called *influence volume* approach [21], which assumes that, in the volume of the polymer/gas solution involved in the growth of a bubble, no other bubbles may nucleate as a consequence of the depleted gas concentration [22]. As the bubble keeps on growing, the subsequent step of bubble impingement has to be taken into account to consider bubble/bubble interactions. Typically, a few growing bubbles are considered and numerical models are used to investigate the evolution of the bubble wall thickness, state of stress and deformation [23–29]. Eventually, the bubble walls may rupture, due to the presence of structural inhomogeneities in the polymer matrix, either introduced from outside (e.g., solid particles) or generated inside the polymer (e.g., crystallization) [30–32]. In both cases, it is assumed that due to the presence of such inhomogeneities, a non-uniform deformation takes place, determining a high level of stress at the polymer-heterogeneity interface that, in turn, may lead to wall rupture.

The comprehensive picture of the state of the art of foaming modeling is reported in Fig. 1, as a scheme of the aforementioned sequence of operations, together with the most important scientific contributions that have dealt with the modeling of one or more of these operations.

Although the strategies for bubble wall rupture should lead, by definition, to open-cell foams, a close observation of the foam morphology, and, in particular, of the polymeric layer dividing two neighboring cells, usually shows that bubble walls are still present, even if broken (Fig. 2b). In this case, then, some of the properties may not conform to fully open-cell foams, in which the polymer is solely confined to the struts. Fig. 2 shows a clarifying example of a closed-cell foam (a), an open-cell foam with broken walls (b) and a fully open-cell foam, with no walls at all and with the polymer solely confined to cell struts (c).

In order to have a fully open-cell morphology, where no material occupies the bubble walls, it is not sufficient to produce a fracture within the wall, but a *bubble wall retraction* is needed. In metallic foams as well as in thermosetting polyurethane foams and soap bubbles, low viscosities and high surface energies are straightforward conditions for wall retraction, which can be very fast and easy [45,46]. In thermoplastic polymers, conversely, viscous forces can be much stronger than interfacial forces,

hindering bubble wall retraction, thereby leading more easily to open-cell morphologies such as the one reported in Fig. 2b [31].

In this work, in order to achieve fully open-cell foams, we conduct a comprehensive analysis, by means of experimentally-validated modeling and numerical simulations, of the sequence of events leading to the formation of the foam and to the development and rupture of the bubble walls. In particular, we investigate in details the role of the polymer elastic recovery as an additional, crucial factor in the bubble wall retraction mechanism. Finally, the developed model allows the design of the material and the process to drive the foam to a fully open-cell morphology. Both the use of the elastic recovery as an additional (actually dominant, at least in thermoplastic polymers) retraction mechanism and the resulting design tool for final foam morphology control are the main novelties of the present contribution.

The approach was validated by using a homemade apparatus with a visualization window that was designed for microcellular foaming at different processing conditions (i.e. temperature and pressure). The thermoplastic polymer used during the experiments was a poly(ϵ -caprolactone) (PCL) foamed with CO₂. The experimental results were compared with the theoretical analysis, as it is shown in the result section.

2. Theoretical background and modeling

The present approach analyzes the foaming process, from bubble growth to bubble impingement and bubble wall opening, and is divided into four operations, as described in Fig. 3: single bubble growth (SBG), impingement (IM), bubbles wall rupture (RU), and bubbles wall retraction (RE). It is worth noticing, here, that we did not include the bubble nucleation stage in the model. The latter assumes that a given nuclei density is fixed and describes the growth, interaction and coalescence phenomena. In the following, we will describe in detail the different steps and their mutual interactions.

2.1. SBG

The single bubble growth (SBG) model describes the growth dynamics of an isolated bubble, driven by the presence of supersaturated gas within the polymeric matrix. Single bubble growth in a viscoelastic system is modeled through a detailed 3D mathematical description, by adapting a model, initially proposed by Everitt et al. [25], to the PCL/CO₂ case at hand. All relevant properties (rheological, sorption, volumetric and interfacial) have been either taken from the literature [11] or directly measured in the present work, as described below.

We consider a single spherical gas bubble with initial radius R_0 surrounded by a spherical shell of a viscoelastic liquid containing a given quantity of dissolved gas. The initial bubble volume is $V_0 = 4/3\pi R_0^3$ and the initial gas pressure in the bubble is p_{g0} . Bubble growth is driven by the difference between the actual gas pressure inside the bubble, p_g , and the external ambient pressure, p_a . We assume isothermal conditions, incompressibility of the viscoelastic fluid and negligible inertia.

Under the above conditions, the bubble growth dynamics are governed by the momentum balance equation for the liquid shell and by the diffusion equation of the gas from the liquid to the bubble. Because of the spherical symmetry of the system, we choose a spherical coordinate system with the origin coinciding with the bubble center. Furthermore, due to the liquid volume conservation, we transform the radial coordinate r into a Lagrangian volume coordinate x such that $r^3 = R^3 + x$ [24], where R is the time-dependent bubble radius. Therefore, $x=0$ is the (Lagrangian) position of the bubble-liquid interface and $4/3\pi x$ is the liquid volume between a generic radial position inside the fluid shell and the

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