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Bubble formation in complex fluids using an orifice in throat arrangement

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ABSTRACT

During the co-flow of a gas and a surrounding liquid film, the inner thread of gas breaks due to Rayleigh instability, and produces a series of bubbles in the embedding liquid. This article describes a co-flow arrangement that generates bubbles to form a hydrogel scaffold. The flow arrangement utilizes the "orifice in throat" configuration for a second squeeze on the bubble that resulted in further split into the bubbles of smaller size. Aqueous solutions of two biocompatible polymers were considered as the continuous phases. These are alginate and chitosan. The mechanism of bubble formation was studied under a microscope. The bubbles were collected on a petridish in a thin layer of liquid. The images demonstrate the self-alignment of bubbles in a monolayer, without coalescence, or shrinkage. An edge detection algorithm was utilized to compute the bubble size. The cumulative frequency of less than type as a function of the bubble diameter was fitted to Gompertz function. The derivative of this function provided the bubble size distribution. Multiple functions were considered for a single curve to explore possibilities other than unimodal distributions. The bubble growth and disengagement processes, and consequent dispersion in bubble size were compared for the two gel systems. The effect of heterogeneity in the gel formulation on the bubble generation process associated bifurcations was reviewed. Sprinkling of the cross-linker solution on the scaffold resulted in the formation of a robust free-resting gel film, without any alteration of the mutual alignment of bubbles. The vacuum-dried gel film was further studied under a scanning electron microscope.

1. Introduction

Emulsification of one fluid phase into another immiscible phase can be accomplished using a fluidic device. The advances in the construction of fluidic devices with small feature size enable formation of emulsions, where the dispersion of the phases can be controlled to micro scale. The dispersion process, by itself is instability driven. When the continuous fluid is as complex as a swelled biopolymer from a natural source, and the dispersed phase is an inert gas that has significantly different density and viscosity, it is anticipated that the bubble formation process may go through chaotic oscillation or bifurcation from steady states. This combination of complex liquid and the gas phase is encountered while making porous gel film for controlled release and /or tissue harvesting applications.

A gel film that provides a three dimensional support for the formation of a matrix, and also delivers the biological agents is a subject of extensive investigation. Use of natural and synthetic polymers, and hydrogels are known, where the biocompatibility and the biodegradability are important requirements. A gel layer has the potential to hold a significant volume of a biological agent that can diffuse into the host tissue over a period of time. Also the gel layer, loaded with a matrix forming cell can act as a scaffold, over which the tissue regeneration takes place. In these applications, it is important that a substantial porosity is induced in the gel layer. Additionally, the porosity has to be uniformly distributed so that the pore to pore distance remains uniform. This calls for a highly ordered pore structure. Emulsion freeze drying, fiber bonding, solvent casting or particulate leaching, gas foaming, thermal phase separation, electrospinning, and use of supercritical CO_2 are some of the methods to induce the voids in a gel layer [1]. Direct introduction of bubbles using a fluidic arrangement is an alternative method that allows better control of the void size and the porosity. Under most circumstances, the bubbles generated by this method are monodisperse. The bubbles rapidly self-assemble, and provide an ordered structure. Also, the gel is not exposed to any chemical or thermal treatment by this method. The method is inexpensive, in comparison with the solid free form fabrication techniques.

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This article describes a co-flow arrangement to generate voids in two types of gel systems. These are alginate and chitosan. Alginate is a naturally occurring polysaccharide, sourced from brown algae that grow in warm areas. Chitosan is the deacetylated form of Chitin, the second most abundantly available polysaccharide in the nature, other than the cellulose. Chitin is commonly sourced from the shells of crabs, prawns, lobsters, shrimps and exoskeleton of insects. Both of these gel systems are known for their biocompatibility, and have potential use in drug delivery and tissue regeneration [2–11].

Bubble generation in a single nozzle with a co-flowing liquid has been investigated by several researchers [12,13]. Use of pulled microcapillaries to generate a gel scaffold of alginate has been reported [14]. In a general co-flow arrangement, the pulled capillaries are arranged one inside the other. The inner gas thread is dragged by the co-flowing liquid until the gas stream snaps to form a bubble. There are other variants to this arrangement [15–18]. In a flow focusing arrangement, the liquid may be injected in a cross-flow manner to impart a direct squeeze on the dispersed phase [18,19]. Another extension to this scheme has been reported very recently [20], where the bubble formation was orchestrated by flowing the two phases through a series of flow focusing joints one after the other. Fu et al. [21] conducted experiments with polyacrylamide solution to emphasize the importance of rheology in bubble generation.

In this article, a co-flow device is described, where a second constriction beyond the tip of the inner capillary ensures further splitting of the bubbles. In traditional co-flow arrangements, the bubbles were found to be an order of magnitude larger than the feature size of the device. Introduction of a second squeeze enables achieving a smaller bubble size, on the order of the feature size of the device. A major objective of this research was to explore the bubble generation process in this "orifice in throat" configuration. The disengagement of bubbles from such device requires the collapse of the neck that holds the bubble to the tip of the nozzle. Since the collapse is instability driven, a bifurcation from monodisperse cluster to higher levels of dispersion may occur. Other than the velocities, the type and consistency of the fluid may influence such bifurcations.

The two gel systems were considered. An aqueous solution of sodium alginate with dissolved pluronic F127 as the surfactant made the liquid phase in one gel system. In the other case, chitosan, dissolved in 0.2 M acetic acid solution in presence of Lutensol AT 25 made the liquid phase. The latter system was found to be more heterogeneous, and the contrast makes the two systems as a good set of candidates for this study. To draw parallel from other flow focusing devices, the bubble size distributions from T-joint of similar feature size are reported here. The formation of bubbles was studied under a digital microscope. The self-alignment of bubbles in a petridish was also studied under the microscope. The size distributions of bubbles for different gel systems at varying liquid to gas flow ratio are reported. The changes in the dimensions of the gel film, bubble size and the mutual alignment of the bubbles were analyzed, as the film underwent crosslinking and vacuum drying.

2. Methods

4% sodium alginate (Sigma Aldrich) solution in distilled water was prepared by using a magnetic stirrer at 350 rpm for 12 h, and subsequently a mechanical mixer at 3000 rpm for 3 h. The pH of the solution was found to be 7.55. 4% pluronic F-127 (Sigma Aldrich) solution in distilled water was used as a surfactant. The pH of the pluronic solution was found to be 6.58. The solution was stirred for one hour using a magnetic stirrer at 350 rpm. The solution was then kept in a refrigerator at 4 °C for 24 h to ensure complete dissolution. The alginate and the pluronic solutions were mixed in even proportions using a magnetic stirrer at 100 rpm for 10 min.

The chitosan solution was prepared by dissolving chitosan beads (Sigma Aldrich) in 0.2 M acetic acid solution with the use of a mechanical stirrer. The stirring at 2500 rpm was conducted for 16 h. The viscous chitosan solution was filtered through Whatman filter paper of Grade 4. Subsequently the Lutensol AT 25 (a linear alcohol ethoxylate from BASF) was added to the solution. The pH of 1.5 wt% of chitosan solution was found to be 4.50 prior to the addition of surfactant. The addition of Lutensol AT 25 (1 wt%) resulted in increase of pH to 5.5.

The viscosity of the two polymer solutions were measured in the Brookfield viscometer and Anton Paar rheometer using the cone and plate arrangement, and the parallel-plate geometry respectively. Surface tension and contact angle of chitosan solutions were measured in a Goniometer (Rantac, Germany). Prior to the flow experiments, a settling time of one to two days was provided to the polymer solution at the post-mixing stage.

A co-flow device was fabricated using pulled glass capillaries (Fig. 1a). The diameters of the inner and outer capillaries are 0.5 mm and 0.75 mm respectively. The outer capillary was extended by 1.0 mm beyond the tip of the inner capillary. At the tip, the outer capillary was squeezed further to an inner diameter of 0.5 mm, resulting in the "orifice in a throat" configuration. On the upstream side of the tip, there were straight sections in both inner and outer capillaries to ensure fully developed flow. The gas was flowed through the inner capillary, while the aqueous polymeric solution flowed through the outer capillary. Constant flow rate of aqueous solution was maintained by displacing the solution from a transfer cylinder with the paraffin oil from a syringe pump from Harvard Apparatus, U.S.A. The flow of nitrogen was obtained from a gas cylinder through a mass flow controller from Alicat Scientific, U.S.A. The gas flow rate was varied from 0.1 mL/min to 1 mL/min, where as the liquid flow rate was set at a value within the range of 4.0–8.0 mL/min. For comparison, the bubbles were formed using a T-joint (Fig. 1b) with the diameter of the tube as 0.5 mm. In this joint, the gas phase met the liquid phase at an angle of 90°.

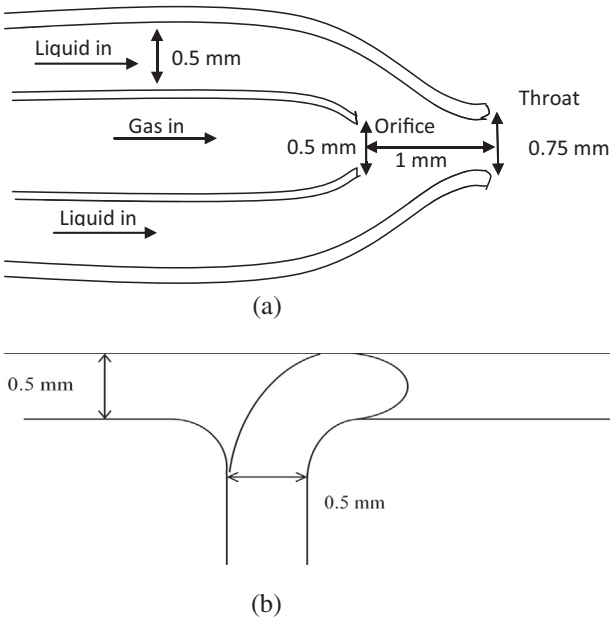

Fig. 1. Schematic drawing of (a) orifice-in-throat arrangement and (b) T-joint.