



Short Communication

A simple derivation of the critical condition for the ultrasonic atomization of polymer solutions



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ABSTRACT

A simple model is proposed for the ultrasonic atomization of polymer solutions. In this model, the atomization process is approximated as an equilibrium process. It is shown that the minimum attainable droplet size is determined by two parameters, the (Rayleigh) acoustic pressure acting on the surface of the liquid, and the surface tension of the liquid. Increasing the viscosity of the liquid suppresses the formation of small-sized droplets because of increased attenuation of the sound wave and thus decreased acoustic pressure. Lowering the surface tension of the liquid (e.g., by spreading a surfactant film on the liquid surface) has the opposite effect of enhancing the formation of smaller droplets. Also, there exists a maximum limit for the droplet size, because when the produced droplet is too large, the aspiration flow is unable to carry the droplet against sedimentation. These predictions are supported by experimental observations.

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1. Introduction

Ultrasonic atomization (or spray) is a versatile technique for producing liquid micro droplets for various uses, particularly for particulate drug formulations [1,2]. The ultrasonic spray method is also recognized as a powerful tool to prepare nanostructured materials [3]. The problem of estimating acoustic radiation pressures at free liquid surfaces has been a subject of theoretical study for some 50 years [4–8]. Many empirical studies have been reported on how process conditions (such as excitation frequency and amplitude, and liquid flow rate and geometry) and liquid properties (e.g., viscosity, surface tension) impact the numbers and sizes of droplets produced for processes involving continuous flows of liquids [9–14]. Recently, our laboratory has demonstrated a very simple batch-type ultrasonic atomization process in which liquid droplets produced are collected using a bench-top aspirator [15,16]. Using this procedure, we were able to produce micron-scale particles of a variety of polymeric materials (such as poly(vinyl alcohol), poly(ethylene oxide), poly(vinyl pyrrolidone), and their composites with carbon nanotubes). In these previous studies, we showed that the number and size of ultrasonically generated drops are governed by two dimensionless parameters: the

so-called Ohnesorge number, $Oh (\equiv \eta_o / (\rho R \gamma))^{1/2}$ where η_o is the zero-shear-rate viscosity of the liquid, ρ is the density of the liquid, γ is the surface tension of the liquid, and R is the radius of the capillary formed by extension), and the Deborah number, $De (\equiv \lambda / t_R)$ where λ is the relaxation time of the liquid, and t_R is the Rayleigh time ($\equiv \rho R^3 / \gamma$)^{1/2}. When $De > 1$ (i.e., when capillary wave breakup is delayed by a long relaxation time relative to the Rayleigh time scale), larger droplets were observed; the elongated stream of liquid breaks up into droplets due to the surface tension-driven Rayleigh instability. The droplet number drops rapidly with increasing Oh (i.e., as the viscous effect becomes dominant), and eventually the process fails to produce any droplets when Oh exceeds a certain value. These observations confirm the current understanding of the ultrasonic atomization mechanism. In this paper, we propose an alternative (and much more simplistic) way of understanding the key aspects of the process. We are not aware of any similar previous attempts in the literature.

2. Theoretical models

We propose a simple theoretical model that can be used to rationalize the sizes of ultrasonically atomized liquid droplets. A detailed analysis of this problem would require the use of interfacial fluid mechanical approaches and simulations [5–7]. However, in the present work, we will only use a simple thermodynamic argument. Referring to the situation described in Fig. 1, let us

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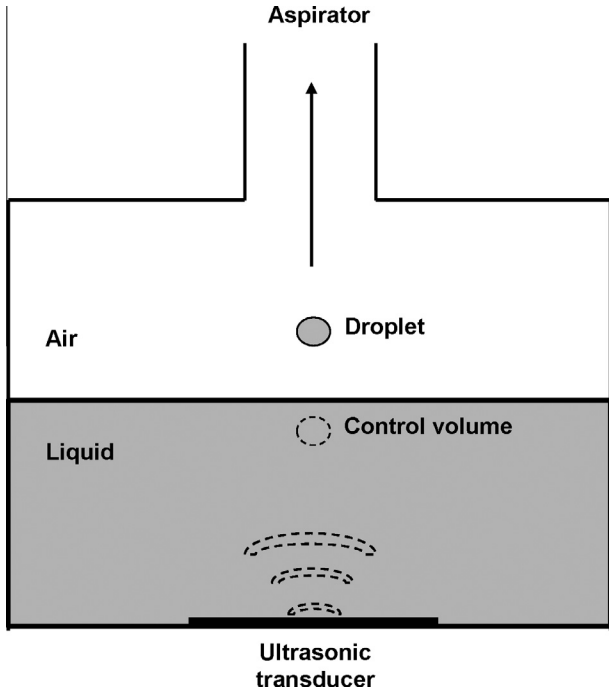


Fig. 1. Schematic illustration of the ultrasonic atomization setup used in this study. The dimensions of the main components are as follows: diameter of the vessel = 6 cm, height of the liquid volume = 2 cm, diameter of the ultrasonic transducer = 2 cm.

specifically consider a droplet of radius R formed from a liquid under acoustic pressure. We define the liquid in the droplet as the control volume. The Gibbs free energy change associated with transferring the liquid in the control volume from the bulk state to the droplet form (ΔG_{drop}) can be expressed as

$$\begin{aligned} \Delta G_{drop} &= G_{drop}^l(T, P) - G_{bulk}^l(T, P_{aco}) \\ &= [G_{bulk}^l(T, P) + 4\pi R^2 \gamma] - G_{bulk}^l(T, P_{aco}) \end{aligned} \quad (1)$$

Here we assume that the atomization process occurs at a constant temperature (T), and the acoustic stress at the air–water interface (P_{aco}) that causes the droplet to form immediately relaxes to a value, P ($< P_{aco}$), for the liquid inside the atomized droplet. Under the isothermal condition, the fundamental free energy equation becomes $dG (=VdP - SdT) = VdP$ (where V is the volume, and S is the entropy), and since a liquid is typically effectively incompressible, $\Delta G = V\Delta P$. Therefore, Eq. (1) can be rewritten as

$$\Delta G_{drop} = V_{drop}(P - P_{aco}) + 4\pi R^2 \gamma = \frac{4}{3}\pi R^3(P - P_{aco}) + 4\pi R^2 \gamma \quad (2)$$

where V_{drop} is the volume of the droplet. It is obvious that the first term on the right-hand side of the equation (i.e., the bulk term) favors the formation of the droplet, whereas the second term (the surface term) suppresses it. Also, the bulk term varies as R^3 , whereas the surface term varies as R^2 ; this means that at small R the surface term dominates, while for large R the opposite is true. Therefore, the acoustic stress would not be able to break up the liquid at the air–water interface into too small-sized droplets – smaller than a certain critical size; the surface tension imposes a minimum attainable droplet size. We would like to point out that Eq. (2) is analogous to the expression of the classical homogeneous nucleation theory for the free energy change of forming a liquid drop in surrounding vapor; the difference is that Eq. (2) concerns the detachment of a droplet from a body of liquid, whereas the classical nucleation theory describes the nucleation of a liquid phase

from a supersaturated vapor; for this reason, the first terms of the two free energy equations are different [17]. Similarly to the derivation of the Kelvin equation for homogeneous droplet nucleation [17], this critical minimum droplet radius (designated as R_{min}) can be calculated by differentiating Eq. (2) and equating to zero (i.e., $d\Delta G_{drop}/dR = 0$ at $R = R_{min}$). This procedure gives

$$R_{min} = \frac{2\gamma}{P_{aco} - P} \quad (3)$$

The equilibrium pressure of the liquid inside the droplet (P) is unknown. To an approximation, one can assume that the pressure inside the droplet is not much different from the vapor pressure of the liquid at the given temperature; that is, $P \cong P_{sat}(T)$. In reality, P is expected to be greater than $P_{sat}(T)$. Therefore, this assumption likely provides a minimum likelihood estimate for R_{min} .

We would like to note that in form Eq. (3) is identical to the Young–Laplace equation that relates pressure differences to curvature across a surface, $\Delta P = 2\gamma/R$ [17]. This analogy allows us to explain how ultrasonic surface disturbances give rise to atomization. When the acoustic wave reaches the liquid’s surface, the acoustic pressure produces capillary waves [4,9]. If the sizes of the wave protrusions ($\sim R$) are large that the Laplace pressure ($2\gamma/R$) is less than the acoustic radiation pressure ($\Delta P \cong P_{aco} - P_{sat}(T)$), the wave peaks break off into droplets. Therefore, P_{aco} and γ primarily determine the sizes of the resulting droplets. Eq. (3) specifically explains how the parameters P_{aco} and γ affect the sizes of the droplets formed by ultrasonic atomization of a liquid; if one wants to reduce the sizes of the droplets, one would need to increase the acoustic stress transmitted to the surface of the liquid (e.g., by decreasing the viscosity of the liquid) or decrease the surface tension of the liquid (e.g., by coating the surface of the liquid with surfactants). We performed experimental tests to qualitatively confirm these trends (discussed in Section 4).

We note that Eq. (3) predicts that R_{min} scales linearly to γ (given that P_{aco} is independent of γ). This prediction is different from the previously proposed relation, $R = 0.17(8\pi\gamma/\rho f^2)^{1/3}$ where ρ is the liquid density, and f is the frequency of the capillary waves on the liquid surface [9]. The exact scaling relationship between R and γ remains to be tested experimentally. It is also interesting to note that Eq. (3) shows how R depends on P_{aco} (the “amplitude” of the ultrasonic waves) and thus on the energy intensity (“loudness”) of the ultrasonic waves ($I \sim P_{aco}^2$), whereas the Robert Lang’s equation above defines R as a function of the frequency (“pitch”) of the waves.

We expect that there also exists a maximum limit for the size of the droplet (R_{max}). This upper limit is imposed by the fact that if the droplet is too large, the aspiration force applied to the droplet may not be sufficient to overcome the sedimentation force and pull the droplet toward the filter where the dried polymer particle will be collected. This R_{max} quantity can be evaluated by comparison of the downward sedimentation versus upward aspiration forces. The magnitude of the sedimentation force can be calculated as

$$F_{sed} = \frac{4\pi R^3}{3}(\rho_{drop} - \rho_{air})g \cong \frac{4\pi R^3}{3}\rho_{drop}g \quad (4)$$

where ρ_{drop} is the density of the liquid inside the droplet ($=1000 \text{ kg/m}^3$), ρ_{air} is the density of the surrounding air ($\approx 1 \text{ kg/m}^3$), and g is the gravitational acceleration ($=9.8 \text{ m/s}^2$). The magnitude of the aspiration force can be calculated as

$$F_{asp} = f v_{vap} = (6\pi\eta_{air}R) \left(\frac{\dot{M}_{vap} v_{sat}^v}{A} \right) \quad (5)$$

where f is the viscous drag coefficient ($=6\pi\eta_{air}R$ for a spherical droplet in air according to Stokes’ law), v_{vap} is the water vapor flow velocity, η_{air} is the viscosity of the air ($=1.827 \times 10^{-5} \text{ Pa s}$), \dot{M}_{vap} is

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