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STUDY OF DROP OSCILLATION AND ROTATION IN IMMISCIBLE LIQUID SYSTEMS

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1. INTRODUCTION.

The dynamics of free liquid drops has long been a subject of interest, both for the sake of basic scientific understanding as well as for various applications in material science and the chemical industry. The rigorous treatment of the problem of dynamics of liquid drops in a gravitational field is complicated by the fact that, for most droplets of practical size, surface-tension forces and gravity are two competing factors influencing the dynamics of such oscillations or rotations. This is the limitation inherent to all laboratory work. Experiments carried out in the low gravity of space flight may not suffer from such interference, but other complications arise from the remote operations (Wang 1979; Jacobi et al. 1979 [1][2]). In immiscible liquid systems, the effects of gravity can be made negligible, although other difficulties arise owing to the mass loading from the outer host liquid and from boundary layer dissipation.

2. DROP OSCILLATIONS IN IMMISCIBLE SYSTEMS.

In this section, we shall be interested in the measurement of the first few resonance frequencies and the damping constant in the small-amplitude region as both the drop size and the viscosity are varied. Viscosities varying between 1.3 and 130 cSt will be studied, and the drop diameter will range between 0.5 and 1.5 cm. The experimental method involves acoustic levitation and radiation-pressure-force modulation (Marston and Apfel 1979 [3]). In addition to quantitative infor-

mation about the various resonance modes of the drop, a qualitative treatment of the characteristics of the internal fluid particle flow field has also been obtained. Comparison with available theoretical predictions was generally favorable when appropriate precautions had been taken to satisfy the theoretical assumptions. It then appears, at least for the steady-state or long-time behavior, that the linear theory yields satisfactory answers to the problem at hand. No detailed study of a possible transient regime has been carried out since this work.

A liquid drop can be trapped at a stable position in an acoustic standing wave existing in a resonant cavity filled with liquid. The static equilibrium shape of the drop can be controlled by varying the magnitude of the acoustic radiation force, which is, to second order, proportional to the square of the magnitude of the first-order acoustic pressure. In this case the cavity is of rectangular geometry, and the axis of symmetry is taken as its vertical axis. Depending upon the standing acoustic wave, or the pressure intensity, a drop may be given the shape of a prolate spheroid, or oblate spheroid, or a nearly perfect sphere.

Shape oscillations are induced through a low-frequency modulation of the acoustic radiation force. The excitation can be that corresponding to either a periodic elongation of the drop at the poles, or a periodic compression of the drop at the poles. The restoring force is, in the ideal case, provided only by the interfacial tension which tends to drive the drop back to the equilibrium shape. For low-amplitude vibrations both modes will yield the same results for the resonance frequency, although this shall not be the case for large displacements (Trinh and Wang 1980) [4].

Various liquids have been used for both drop and host media. First the combination of a phenetole drop suspended in a 1:2 by volume mixture of methanol and distilled water was used. Next, various viscosity grades of Dow Corning silicone oil (5 - 200 cSt) were mixed with carbon tetrachloride to form drops which were suspended in distilled water. All drop liquids were colored with oil-

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soluble dyes.

The acoustic frequencies were either 22 kHz ($\lambda \approx 6.75$ cm in water) or 66 kHz ($\lambda \approx 2.25$ cm in water). For a water host the kR parameter had values between 0.23 and 0.69 at 22 kHz, and between 0.69 and 2.1 at 66 kHz. Here k denotes the magnitude of the acoustic wave vector ($2\pi/\lambda$) and R is the radius of the drop.

The drop oscillation amplitude is monitored using an optical detection technique: a slit parallel or perpendicular to the axis of symmetry is uniformly illuminated, the shadow of the drop is then centered across the slit, blocking some of the light. As the drop oscillates, the light intensity detected by a photo-transistor varies periodically in phase. Up to reasonably large amplitude the response has been found to be approximately proportional to the drops deformation. If the drop axis rotates, however, the detector will no longer yield a true maximum amplitude variation.

Driven shape oscillations of liquid drops under reasonably well-controlled conditions have allowed the determination of the resonance frequencies of the first few modes. The configurations of the oscillating drops resemble closely those predicted. The time dependence of the oscillation amplitude is very close to sinusoidal for free vibrations, but the time spent in the various configurations characteristics of each mode depends strongly on the acoustic drive for driven oscillations. For example, in the case of the fundamental axisymmetric mode, a freely oscillating drop spends only a very slightly longer time in the prolate configuration, while a driven drop might be found in the prolate shape during a longer, equal, or shorter time, depending upon the driving mode.

For small-amplitude oscillations, a drop suspended in a host liquid behaves in a way very similar to the usual damped harmonic linear oscillator in many respects: the response to a sinusoidal excitation is almost purely sinusoidal, the frequency of maximum response is characterized by an approximately 90° phase shift between

the drive and the response, the decay rate is roughly linear with the drop viscosity, and the resonance curve for the displacement has the familiar shape.

An important difference arises from the presence of the outer support liquid which not only adds additional inertia, but also plays an important role in the dissipation mechanism, as predicted by the theory (Marston 1980) [5]. For low viscosity (less than 20 cSt), and drops volumes between 0.5 and 2 cm³ the dissipation mechanism is dominated by the energy loss through the boundary layer at the drop surface. As the viscosity of the inner fluid grows larger, however, the theoretical predictions appear to overestimate the dissipation rate. This has been tentatively attributed to the residual momentum of the boundary-layer fluid. Study of the internal fluid particle flow has revealed a quasi-potential flow field with no noticeable circulation, thereby confirming Lamb's predictions and the validity of the theoretical assumptions.

The acoustical-levitation technique allows a non-invasive experimental study of the dynamics of oscillating drops if some precautions are taken to minimize the influence of the acoustic fields. A detailed quantitative analysis is needed in order to determine exactly the extent of the influence of acoustic forces, but is reasonable to assume that, for very small levitating forces and oscillation amplitudes, their disturbing effect upon the dynamics of the drop vibrations is quite small.

3. DROP ROTATION IN AN IMMISCIBLE SYSTEM.

This section describes the investigations of the dynamics of a rotating liquid mass under the influence of surface tension.

A large (~ 15-cc) viscous liquid drop is formed around a disc and shaft in a tank containing a much less viscous mixture having the same density as the drop. This supporting liquid and the drop are immiscible. If the shaft and disc were not present, the drop would float freely in the surrounding medium and assume the shape

of a sphere. With the drop attached and initially centered about the disc, the shaft and disc are set into rotation almost impulsively, reaching a final steady angular velocity within one-half or two revolutions. The drop deforms under rotation and develops into a variety of shapes depending on the shaft velocity. The process of spin-up, development, and decay (or fracture) to some final shape was common to all runs.

In this system, gravity is diminished at the expense of introducing a supporting liquid which is viscous and which may be entrained by the motion of the drop, thereby allowing angular momentum to be transferred from the drop. Nevertheless, comparison of this experiment's results to the theory of free rotating liquid drops is prompted by the fact that several novel families of drop shapes have been observed.

It is important to recognize that existing theory deals mainly with equilibrium shapes and their stability, while the drop in this experiment is undergoing a far more complicated process. The shape of a liquid drop spun on a shaft and supported by another liquid is very much a dynamical problem. A proper understanding of the results will only come with a dynamical analysis which succeeds in explaining the growth and decay with time of the various drop shapes.

Shapes of a rotating spheroid, have been observed and recorded in this experiment [6]. These include the flattening of slowly rotating drops and the generation of toroidal and lobed shapes at higher rotation rates. Using data recorded on movie film, the development and decay of the rotating shapes were studied for the first time. The neutrally buoyant tracer droplets allowed us to study the dynamics of the behavior, the secondary flow generated by the rotation, the interaction between the drop and the host liquid, and the coupling between the shaft and disc and the drop.

The non axisymmetric shapes of a rotating drop in an immiscible system have been studied. Five basic families of shapes (axisymmetric, two-lobed, three-lobed,

four-lobed, and toroidal) have been observed. The sequence (axisymmetric $\rightarrow$ two-lobed $\rightarrow$ three-lobed $\rightarrow$ four-lobed $\rightarrow$ toroidal) seems to be linked to increasing spin-up velocity. For the axisymmetric case, direct comparisons of experiments with the theory of a free rotating drop were surprisingly good the equatorial area differs from theory by only 30%. Furthermore, the non-axisymmetric shapes are in good qualitative agreement with the theory, although the theory does not address the presence of an outer fluid.

4. OSCILLATION OF A ROTATING DROP IN AN IMMISCIBLE SYSTEM.

A particular subject of interest in this section is the effect of the solid-body rotation of a liquid sphere on its normal mode resonance frequencies for shape oscillations.

Measurements extending up to rotational velocity of 720 rpm have been performed. A typical result would be a shift in the quadrupole ($\ell = 2$) mode frequency ($\Delta\omega_2/\omega_2$) of 1.8 for a 0.4 cm^3 drop. Both denser and less dense drops show a monotonic increase in the fundamental resonance frequency (ω_2) with rotational velocity.

Results of a parametric study of the frequency shift involving variations in the drop radius, density, and interfacial tension will be presented. Generally, the relative frequency shift has been found to be proportional to the square of the rotational velocity and inversely proportional to the interfacial tension. Two striking results are a smaller relative frequency shift for higher resonant modes and an enhanced energy dissipation as the rotation rate is increased.

Comparison with the predictions of a theoretical treatment based on a first order expansion of the fluid dynamical equations has yielded a close agreement for rotation rates up to 700 rpm [7]. A noticeable discrepancy appears above 500 rpm for larger drop sizes (0.4 cm^3 volume), but the agreement remains for the smaller volumes (0.2 cm^3).

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The disagreement occurring at the larger drop radii may be attributed to the shape distorting effect of the levitating acoustic field which induces an oblate equilibrium shape of the drop. Such a non-spherical static shape has been found to result in a higher effective restoring force (i.e., interfacial tension) during shape oscillations. Another explanation for this discrepancy may be that inertial waves in the cylinder are being excited by the larger oscillating drops. In these cases, the condition $\omega_{\text{oscillation}} < 2 \Omega_{\text{rotation}}$ is satisfied, where $\omega_{\text{oscillation}}$ is the drop oscillation frequency and Ω_{rotation} is the rotation velocity.

Further experimental studies are being conducted to measure the dissipation constant for rotation drops, and to detect the presence of inertial waves.

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