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FLUTE: FLUORESCENT TECHNIQUE FOR TWO-PHASE-FLOW LIQUID-FRACTION MEASUREMENTS

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A new technique for the local, instantaneous (limited only by the speed of the data acquisition system; current rates possible in the 100 kHz range) measurement of liquid fraction in two-phase flows is described. The technique is based on a combined absorption/emission radiation mechanism tailored to a highly accurate linear response over the whole (0 to 1) liquid fraction range. In the particular application demonstrated here, ultraviolet-induced fluorescence (in the liquid phase) is used, but in principle other combinations of radiation are possible. The technique is essentially non-intrusive in that transmission of the incident ultraviolet radiation and the emitted fluorescence are effected by miniature optical fibers, and the measuring volume is at the intersection of their optical paths some distance away from the fibers. This distance and the size of the measuring volume can be tailored to the particular application. The technique provides a unique measuring capability, especially for highly dispersed liquid flows (high-end of churn turbulent and droplet flow regimes).

KEYWORDS Two-phase-flow Void fraction Measurement Fluorescence Instrumentation.

1. INTRODUCTION

The phase content is an essential parameter in two-phase flow, so that a whole field of investigation has been devoted to its measurement (Hewitt, 1978; Jones and Delhaye, 1976; Delhaye and Cognet, 1984). The work has been spawned, especially in the 1970's, by the need to understand in detail a very wide spectrum of transient, non-equilibrium, often multi-dimensional two-phase flows as they occur in a postulated Large Break Loss-of-Coolant Accident in a Light Water Reactor (Hon *et al.*, 1980). Early techniques aimed for global measurements (i.e. cross-sectional average), but interest in local measurements began as early as the late sixties and early seventies (i.e., Delhaye *et al.*, 1973). At this time, the available instrumentation is wide ranging in principles employed, and highly sophisticated in technique (e.g., double-sensor resistivity probes that measure not only local void fractions but also local interfacial area concentrations) (Wang and Kocamustafaogullari, 1990; Ishii and Revankar, 1991).

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The particular need that led to the investigation and technique reported herein was in measuring the water depletion from the mixing zone prior to a steam explosion. The experiment designed to explore the basic physics of this mixing process involves (Amarasooriya and Theofanous, 1987) a cloud of very hot (hundreds of degrees Celsius) particles poured into a liquid (water) volume, and a complex multiphase interaction as the very high steam production drives water out of the mixing region. The situation is highly transient, the flow patterns strongly two-dimensional, and the range of real interest is where the liquid fraction drops below $\sim 20\%$, i.e., the measurement is needed from liquid-continuous to a highly dispersed droplet flow. More generally, the technique is uniquely well suited for multi-dimensional, highly dispersed flows for which both radiation attenuation and the local contact probe techniques fail because of the respective attributes; that is, multi-dimensionality requires a local measurement which is not possible by attenuation methods (even tomography is limited by the conflicting requirement of spatial and temporal resolution), and local contact probes are not suitable for highly dispersed flows. For a good perspective on these matters, see Jones and Delhaye (1976).

II. THE MEASUREMENT PRINCIPLE

The basic idea is to induce and detect narrow band irradiation locally, such that in combination with attenuation principles a linear response, independent of the flow regime, can be obtained. As such, the technique relates to and combines elements of several previously known (not necessarily for two-phase) measurement techniques, including (a) activation methods such as “pulsed neutron activation”, “laser raman spectroscopy” (Regnier, 1973), “laser-induced fluorescence”, etc., (b) photon attenuation methods such as “ γ and x ray absorption”, etc., and (c) optical probe methods (measuring changes of the refractive index of the surrounding medium) such as the “U-shaped fiber optical sensor” of Danel and Delhaye, (1971), etc. In particular, laser-induced fluorescence has been used by McDaniel (1983a, 1983b) to measure velocity, density and pressure in iodine-seeded supersonic gas flows, by Owen (1976) to measure velocity and concentration (i.e., mixing) in a single-phase flow (two confined co-axial liquid jets), and more recently by Lai and Faeth (1987), again for single-phase, gaseous, buoyant wall plumes.

In this development we use Ultraviolet (UV)-induced fluorescence in combination with fiber optics and certain filter/mirror arrangements to achieve:

- (a) a good separation between the exciting and emitted radiation,
- (b) the proper attenuation behavior for a linear response, and
- (c) local, internal measurements in highly complex two-phase flows.

The basic ideas of how these goals can be met are described below. Some details of the instrument in its present configuration (FLUTE.1) are given in the next section.

Fluorescence is a type of luminescence phenomenon created by the emission of energy as a molecule, activated by the absorption of a photon, returns to its ground state. Some of the absorbed energy is dissipated in collisions with neighboring molecules in the brief period prior to emission, thus the emitted radiation will be of lower energy and longer wavelength than the energy absorbed in the excitation (Guilbault, 1973; Udenfriend, 1962). The time for which the molecule stays in the excited state is extremely short (order of 10^{-9} s); for the fluorescein dye utilized in the present work, it is $\sim 5 \times 10^{-9}$ s. The peak excitation occurs at 491 nm and the peak in the fluorescence is at 512 nm. With the fiber optic arrangement of Figure 1, that is, a well-collimated beam of UV light and an orthogonally-placed detection path, we can create a measuring (or sampling) volume with a characteristic dimension as small as ~ 1.5 mm. Neglecting beam divergence and attenuation of the fluorescence in the solution (over the path L_2), the power (P) received by the detecting fiber can be expressed as

$$P = I_0 A_0 \phi e^{-L_1 a c \beta} (1 - e^{-\Delta a c \beta}) \mathcal{F} \quad (1)$$

where I_0 is the intensity of the UV exiting the emitting fiber, A_0 is the cross-sectional area of each fiber, c is the concentration of the fluorescein dye in solution (grams of dye per gram of solution—here 10^{-6} to 10^{-4}), ϕ is the quantum efficiency (defined as the number of quanta emitted for each quantum absorbed; for the dye used here, $\phi = 0.92$), a is the absorption coefficient of the solution for the UV radiation, β is the liquid fraction taken as constant along the short path length, and \mathcal{F} is a factor for the geometric optics involved. The basic idea of the current application is to make $a c L_1 \beta \ll 1$, i.e., by appropriate

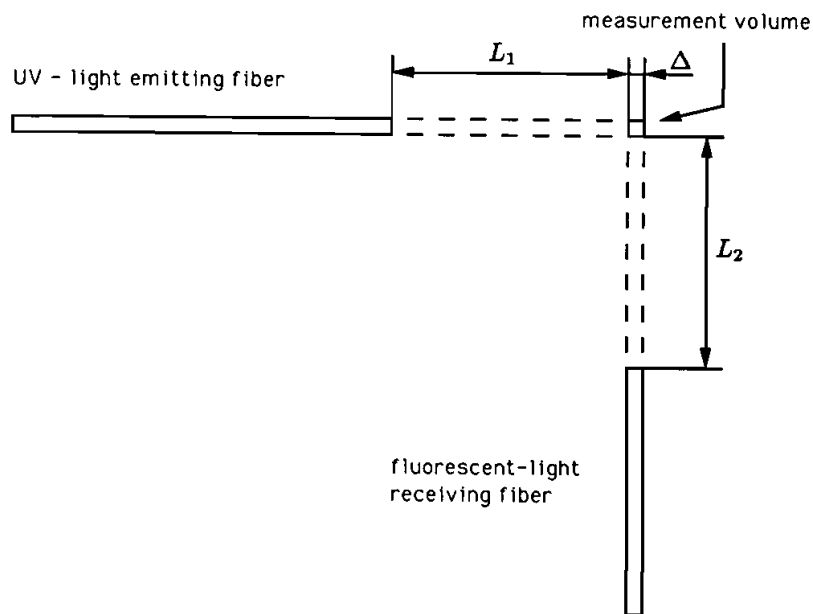


FIGURE 1 Schematic of the arrangement of optical fibers in FLUTE.

selection of c and L_1 , such that $e^{-acL_1\beta} \sim 1 - acL_1\beta$ and $e^{-ac\Delta\beta} \sim 1 - ac\Delta\beta$. Thus, Eq. (1) can be linearized to

$$P = I_0 A_0 \phi ac \Delta\beta \mathcal{F} \quad (2)$$

and with all other factors remaining constant the signal received is made proportional to the liquid fraction β .

Note that $\Delta \ll L_1$ and the conditions for linearity above is achieved by keeping the product $ac\beta L_1$ small. Maximum sensitivity can be obtained by selecting a concentration, c_0 , such that, with the liquid fraction at the upper end of the range of interest (for the particular application), say β_{\max} , the signal obtained is at the upper end of the linear response. Clearly, as β_{\max} increases c_0 must decrease proportionally. At the limit of covering the whole liquid fraction range, $\beta_{\max} = 1$, c_0 has to be quite small; however, with a good UV source, a good signal-to-noise result can be obtained for β 's down to a few percent. The advantage of operating at this limit is that the calibration procedure is greatly simplified, i.e., only one reading (with appropriate consideration of background noise, of course) at $\beta = 1$ defines the calibration line. For the same concentration then, c_0 , the fluorescence can be directly related to liquid fraction through the same calibration line.

Alternatively, assuming that the absorption coefficient does not vary with concentration and that its effective value in two-phase flow is not strongly affected by reflections and refractions (see discussion in Section IV), Eq. (1) can be used directly as providing P as a function of $c\beta$. In this approach a whole calibration curve is obtained by measurements at different values of c , extending well into the non-linear region, in 100% liquid. This is the approach employed in this paper.

III. DEMONSTRATION OF THE TECHNIQUE

In the present experimental set up, the UV source is a 200 W mercury lamp (Oriel Corp. Model 66042). A good separation between excitation and emitted radiations can be obtained by limiting the UV to under 400 nm and measuring the fluorescence in a narrow band around 510 nm. This is accomplished by passing the source light (after collimation) through a dichroic mirror that reflects selectively in the 300–450 nm (and only 10% of the spectrum above), while it transmits and absorbs all other wavelengths. A colored glass filter is used next to eliminate completely the 400 to 700 nm portion of the spectrum (it passes 200–400 nm and above 700 nm) before it enters the transmitting fiber. The collecting fiber leads to a narrow band width interference filter with a peak transmittance at 510 nm and a 50% value in 10 nm around the peak. This filtered signal is read directly into a digital Radiometer/Photometer system (EG&G model 550-1) for steady-state measurements or into a system of photomultiplier tubes, digital voltmeters, and an Hewlett Packard high-speed data acquisition system for transient measurements. Only steady-state measurements are reported here, with the photometer RC time constant set at 400 ms.

The measuring volume can be controlled by adjusting the beam divergence and the distances from the control volume. The best collimation (minimum divergence) can be obtained by placing the input end of the emitting fiber directly into the collimated beam of the UV source. Various degree of divergence can be obtained by focusing the source beam into the fiber inlet. A corresponding (to the focusing) intensification of the transmitted light can then be obtained. For each application, the selection of these parameters (L_1, L_2 , incident beam intensity and divergence) can be made to optimize the results.

We will limit ourselves here to the demonstration of the feasibility of the technique for two-phase dispersed flows in the low liquid fraction range. For this purpose we obtain a basic calibration curve giving the variation of measured signal with dye concentration at 100% liquid, then demonstrate the consistency obtained per Eq. (1) by measuring in a dispersed flow of known dye concentration and liquid fraction; that is, the same reading for the same $c\beta$. The calibration curve at 100% liquid was obtained by starting somewhat above the upper end of the linear region (for the spacing of $L_1 = L_2 = 8$ mm utilized) and carrying out successive dilutions, down to a few percent of the original concentration. The known liquid fractions were obtained by using precision-made perforated discs in the manner described below.

The perforated discs were made from acrylic material (~1 cm thick) drilled on a square array covering a circular area of diameter ~23 mm. The center-to-center spacing is called D , and the hole diameter d ; a disc is then referred by the particular d/D values in thousandths of an inch as, for example, 30/110 meaning holes of 30 thousandths on a 110 thousandth square array. A steady flow through a disc was created under a constant liquid head h , which could be varied in the range of 30 to 70 cm. Taking into account the acceleration of the flow, under gravity, to a measuring distance z below the disc, the liquid fraction can be simply expressed as

$$\beta = \beta_{\text{ref}} \sqrt{\frac{h}{h + z/\alpha_v^2}} \tag{3}$$

where α_v is a loss coefficient for the flow through the hole (measured experimentally) and β_{ref} is the liquid fraction at the disc exit given simply by

$$\beta_{\text{ref}} = \frac{\pi d^2}{4 D^2} \tag{4}$$

with d being the actual diameter of the holes. Thus, by varying the liquid head, h , and the measuring distance, z , a range of two-phase flow cross-sections with "known" liquid fraction can be created. Further, some variation of the internal flow characteristics could be obtained by utilizing 5 discs as follows: 29/80, 29/100, 35/80, 35/110, 51/130. The hole diameters specified are nominal values, some variation being present due to "wiggling of the drill". In calculating β_{ref} a rough average value was used within the range obtained from physical inspection of each disc.

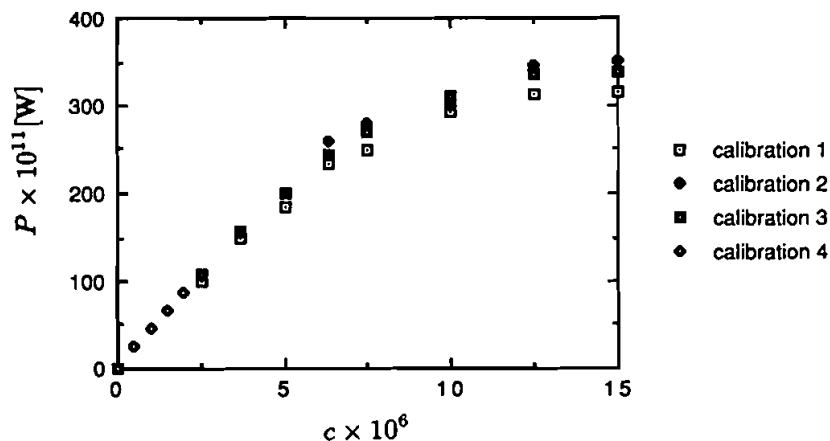


FIGURE 2 Four calibrations obtained at 100% liquid, as described in the text. The points in the range $0 < c < 2.5 \cdot 10^{-6}$ were obtained only for calibration 4.

The calibration curve obtained at 100% liquid in four separate trials is shown in Figure 2. Each trial consisted of a series of measurements obtained with liquid samples from a series of standard solutions previously prepared for this purpose, over the time span of 48 hours. The upper end of the linear region appears at a concentration of $\sim 5 \cdot 10^{-6}$ (relative units, mass of dye to mass of water). Some systematic variation is evident in the non-linear region, whose exact source has not been identified.

The data obtained with the five discs are shown, in relation to the calibration line, in Figure 3. In all these runs the same dye concentration of 10^{-5} was utilized. The data were generated with 5 different liquid driving heads, at a fixed measuring distance ($z = 19$ cm) and a fixed measuring position (center) within the

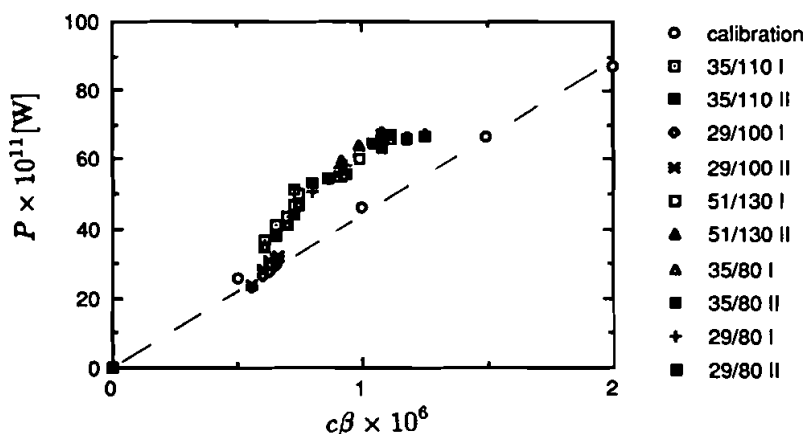


FIGURE 3 Fluorescence data as a function of $c\beta$, obtained with five different discs (with a concentration of 10^{-5}) in relation to the calibration line.

flow. For each disc two independent trials were made—each trial involved a complete reassembly of the experimental set-up. The positioning of the measurement volume was made visually, such that it was well within the flow while avoiding any locally inhomogeneous regions. The difference between these data and the calibration line gives a first idea of the current “accuracy” of the technique, with the following important qualifications:

- (a) Because of small imperfections in hole positioning, size, and alignment there may be a significant variation of *actual local* (in the measuring volume) liquid fraction and the average value calculated from Eqs. (3) and (4). Thus the measurements could be even better than appear in Figure 3—disc 29/100 which lies exactly on the calibration line appeared to produce the most uniform flow.
- (b) Because of reflections and refractions at the gas/liquid interfaces (see also below) there may be inherent errors in the measurement, depending on the actual frequency and geometry of these interfaces in, and perhaps even around, the measuring volume. In this interpretation, it appears that, at least for the kinds of flows considered here, such errors are limited, and the technique is still rather promising.
- (c) The experimental spread in the non-linear portion of the calibration curve (Fig. 2), and the use of c_0 in this range (10^{-5}) for the two-phase measurement, could likewise impact the linear range as in Figure 3.

At this time, our effort is to install the transient measurement capability, construct better discs and an accurate traversing device, such that an error estimate can be accurately pinned down in terms of well known overall quantities (e.g. the total liquid flow rate).

IV. DISCUSSION

The presence of gas/liquid interfaces within the travel paths of the emitted and the detection beams introduce some complications not considered explicitly in the formulation of Eq. (1). Several interpretations/approaches are possible, and the purpose of this section is to provide some initial considerations on this topic. In addition, since the high-speed data acquisition system will allow essentially instantaneous “readings” at rates up to 100 kHz, some interesting questions arise on how to optimize the rate in relation to a particular transient under investigation (i.e., time constant of transient, characteristic length scales in the flow, flow speed). Some initial considerations on this topic are presented also in the second part of this discussion.

A *The Role of Gas/Liquid Interfaces*

The refractive index of water in the 400 to 500 nm range of interest is ~ 1.35 (Hale and Querry, 1973), which implies a normal reflectivity of only 1.7%. As the angle of incidence increases the reflectivity increases also, but it becomes

significant only beyond ~ 70 degrees, thus averaged over all angles the so-called hemispherical reflectivity is still only a few percent (Siegel and Howell, 1981). Clearly, if only a few interfaces are involved the loss due to reflections is negligible. Depending on the two-phase flow regime, i.e., the liquid fraction and the internal length scales, the fiber arrangement can be made such that this condition is met. Note that in addition to fiber distance and beam collimation this "arrangement" can also involve bundles of fibers. The absorptivity in distilled water of both UV and fluorescent radiation is also negligible. Thus refraction remains the only intangible, for now, source of potentially significant error. We believe that it is responsible for a significant fraction, if not all, of the deviation from the ideal behavior (calibration line) observed in Figure 3. As noted already, it is encouraging that even in the presence of a significant number of drops this error appears to be limited; however, more study is needed to fully characterize this effect, such as to optimize the measurement. On the other hand, a limiting condition can be identified as one with a close enough proximity of the fibers that minimize the refraction error to the required degree. The quantitative consideration of refraction has been under way, and it is shortly to appear (Yan *et al.*, 1992).

B Measurement Statistics

Because of the short response time and the capability of making measurements in a small volume, FLUTE is quite flexible and can be adapted to measure liquid fractions in rapidly varying, in both space and time, two-phase flows. In a specific experiment, the measuring volume and data acquisition rate must be selected to ensure that the correct statistics can be estimated from the "sample" record. To illustrate this point, let us consider an idealized situation as follows: a spherical measuring volume of radius R_0 within an infinite regular square array of spherical drops of radius R_l . Depending on the location of the center of the measuring volume, the "measured" liquid fraction will fluctuate between a theoretical maximum and minimum as illustrated in Figure 4. The number of "measurements" required such that five consecutive "average" liquid fractions (over the total number of "measurements") agrees to within 1% of the true value is shown in Figure 5. The results shown in Figures 4 and 5 provide an initial perspective on the interplay between β , R_0/R_l , the number of measurements required, and the potential error band involved. As expected, the measurement requirements increase as the liquid fraction and the relative (to the drop) size of the measuring volume decrease. Further consideration of this topic is to be given in Yan *et al.* (1992).

V. CONCLUSION

The principle and feasibility of a fluorescence-based method for the measurement of liquid fraction in two-phase flow has been demonstrated. The technique appears promising especially for filling an important need for highly transient,

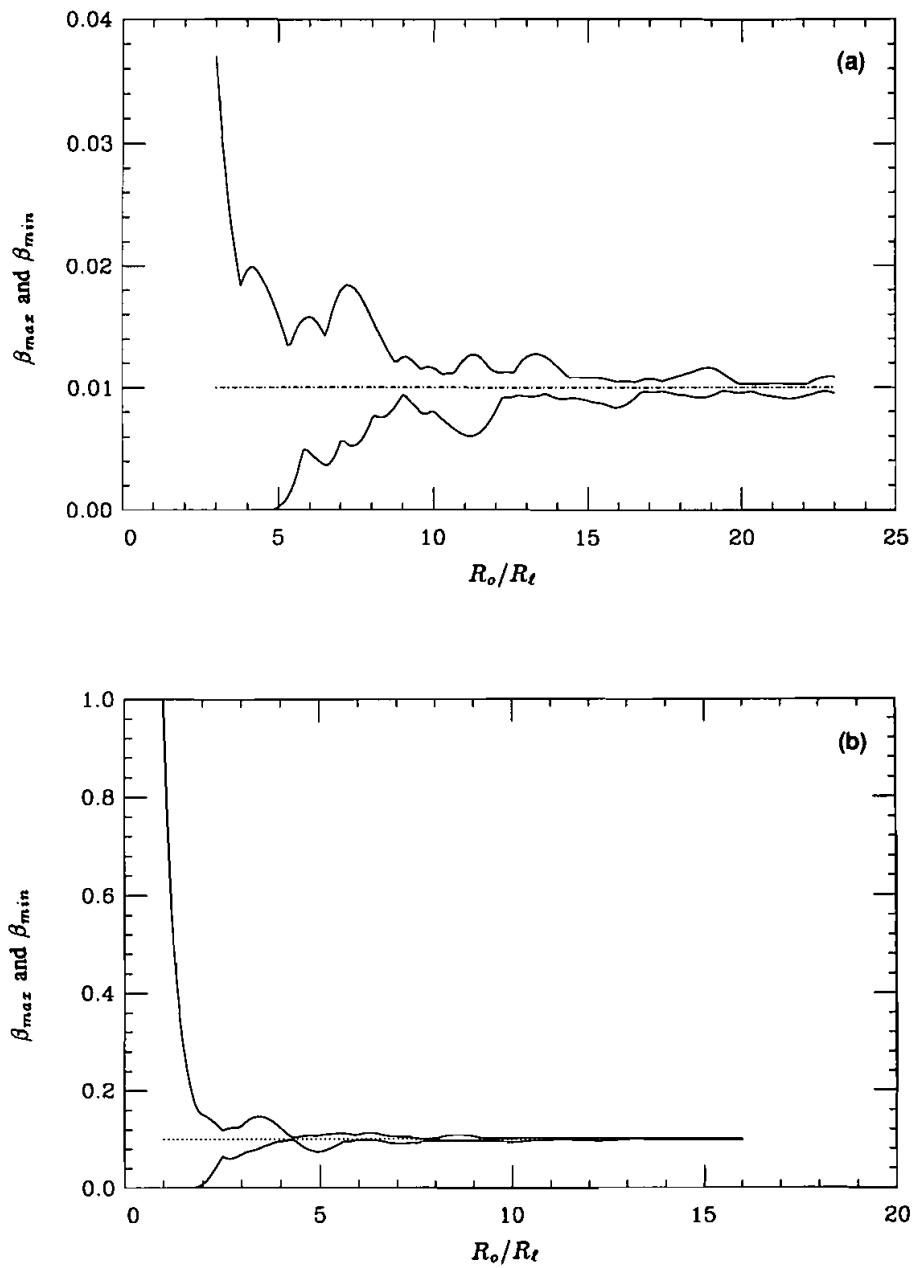


FIGURE 4 The dependence of theoretical maximum and minimum values of liquid fractions on the relative size of the measurement volume (to the drop)—see text. (a) $\beta = 0.01$, (b) $\beta = 0.10$ and (c) $\beta = 0.52$.

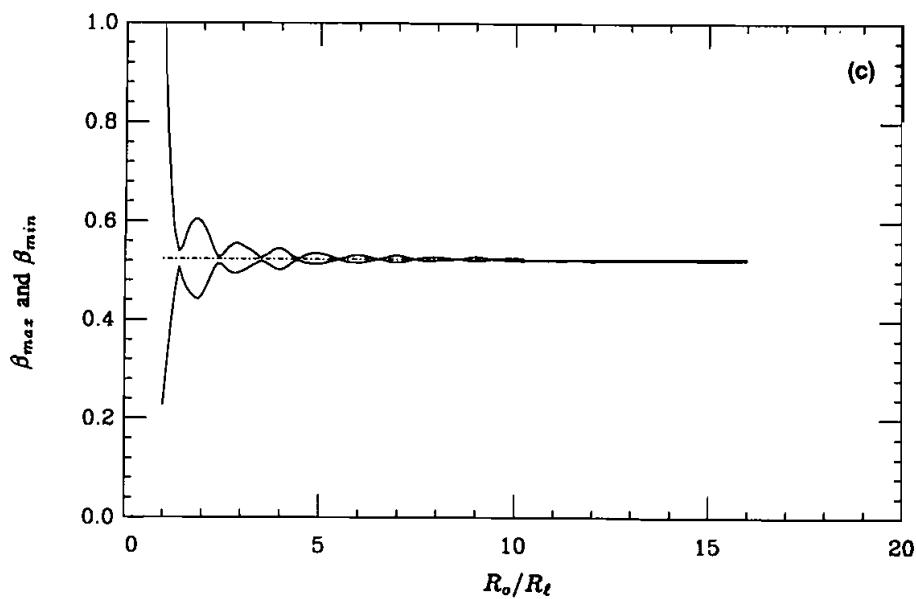


FIGURE 4—(Continued).

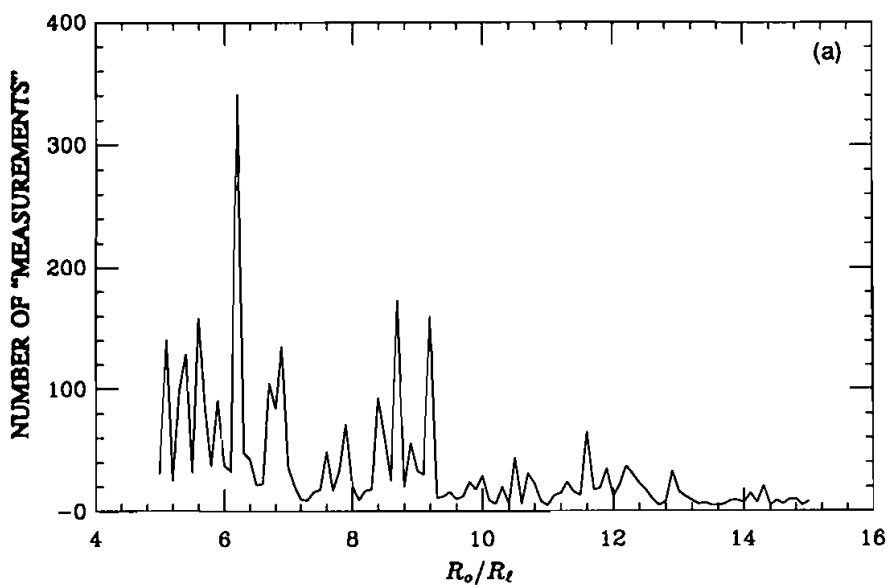


FIGURE 5 The required number of "measurements" to converge to the true value of liquid fraction within 1% and remain within the tolerance in 5 consecutive "readings"—see text. (a) $\beta = 0.02$, (b) $\beta = 0.10$ and (c) $\beta = 0.15$.

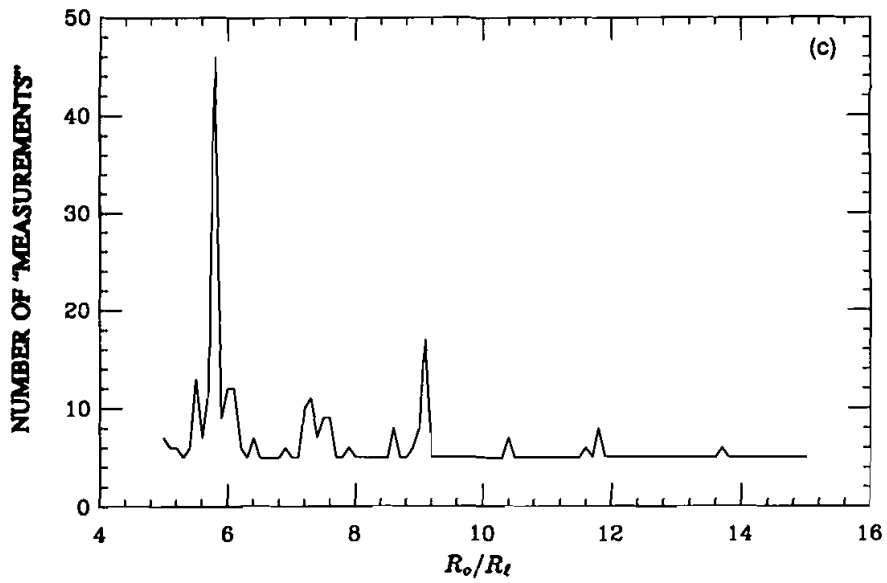
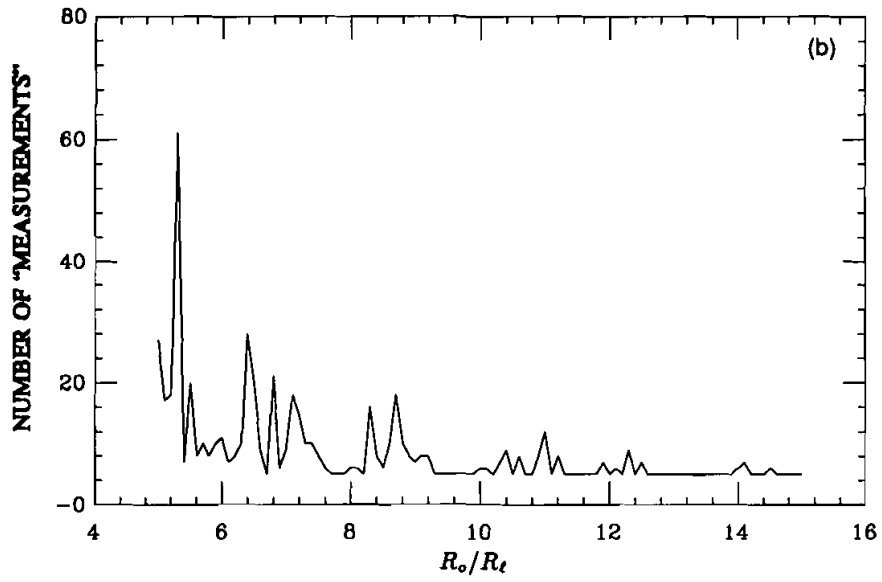


FIGURE 5—(Continued).

spatially highly non-uniform three-dimensional dispersed flows. Work is continuing to define the limitations of the technique (i.e., the role of refractions) and the statistical aspects necessary for applications in rapidly varying, non-uniform flows.

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The topic (two-phase flow instrumentation) and motivation (steam explosions) of this paper are only two of the many research areas in which George Bankoff has made his mark. It is a pleasure and honor to dedicate this contribution to him on this special occasion.

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NOMENCLATURE

| | |
|---------------|---|
| A_0 | cross-sectional area of fibers |
| a | absorption coefficient of ultra-violet radiation |
| c | concentration of solution (grams of dye/grams of solution) |
| c_0 | concentration of solution used in liquid fraction measurement |
| D | spacing between holes |
| d | diameter of holes in disc |
| \mathcal{F} | geometric optic factor |
| h | water level in glass tube |
| I_0 | exciting-light intensity |
| L_1 | distance from end of UV-light carrying fiber to axis of detecting fiber |
| L_2 | distance from end of detecting fiber to axis of UV-light carrying fiber |
| P | power received by fiber |
| R_0 | radius of detecting volume in numerical simulation |
| R_t | radius of droplets in numerical simulation |
| z | distance from disc exit to fiber location |

Greek

| | |
|---------------|--|
| α_v | loss coefficient in the disc |
| β | liquid fraction |
| β_{ref} | reference liquid fraction (disc parameter) |
| Δ | diameter of fiber |
| ϕ | quantum yield of dye |

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