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An Assessment of Experimental Techniques for the Measurement of Bubble Size in a Bubble Siurry Reactor as Applied to Indirect Coal Liquefaction

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Contract Number:

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Contractor:

Dr. S.C. Saxena 8321 Kilbourn

Skokie, Illinois 60076

Telephone:

(312) 679-4147

DOE Project Manager:

Mr. Dennis Smith

Telephone:

(412) 675-6165

DOE Contract Specialist:

Mr. Keith R. Miles (412) 675-5984

Telephone:

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An Assessment of Experimental Techniques for the Measurement of Bubble Size in a Bubble Slurry Reactor as Applied to Indirect Coal Liquefaction

S.C. Saxena

ABSTRACT

The proper design and control of an indirect coal liquefaction process plant requires an accurate knowledge of bubble sizes and size distribution. Of particular importance regarding proper design, is the understanding of the complicated dependence of bubble dynamics on bubble column geometry and the nature of the solid-liquid phases. The nature of the products and their relative proportions on the other hand is seriously influenced by the bubbling characteristics of the reactor because the mass transfer behavior and mixing are dependent on the bubble size and velocity distributions. This report deals with the review of the experimental techniques that have been employed to measure the bubble sizes in multiphase reactors. The methods which fall in three different catagories viz., the photographic, the optical probe and the electrical conductivity probe, are described in an historical sequence and in each case highlighting their unique features and design details. A general methodology of data analysis in each category is given together with their state-of-the-art technology assessment and finally their relative merits and demerits.

An Assessment of Experimental Techniques for the Measurement of Bubble Size in a Bubble Slurry Reactor as Applied to Indirect Coal Liquefaction

Introduction

Serious research efforts are underway to develop alternate energy sources in order to prevent petroleum supply disruptions from having damaging impact upon the economy of those countries dependent on external supplies of petroleum. The global energy crisis and the Arab oil embargo of the early seventies highlighted the vulnerability of the petroleum based industry, and sparked-off a variety of activities around the world related to the efficient conversion and utilization of coal. During the last fifteen years considerable work has been done to understand the science and technology of coal conversion. Monographs of Anderson and Tillman [1], and of Probstein and Hicks [2], discuss the methods and principles involved in the conversion of coal into synthetic fuels, gases and liquids. Much of the historical efforts are referred in these concise reviews. Another work of a similar nature is put together by Wen and Lee [3] and includes authoritative reviews on coal characterization, coal pyrolysis, coal combustion, coal gasification and coal liquefaction. The science and engineering principles involved in the direct liquefaction of coal are discussed at length in the treatise of Shah [4]. Another scheme for producing liquid petroleum products from coal via the synthesis of coal gasification products in a bubble slurry column is referred to as indirect coal liquefaction. Here the gaseous output of a coal gasification plant is bubbled through a liquid (heavy oil medium) column in which fine catalyst particles are kept suspended. The gases from the bubbles diffuse through the liquid and under favorable conditions will combine at the surface of an appropriate catalyst to produce the desired liquid petroleum products. Heat is generated during

the process of gas synthesis and this must be removed to keep the operating conditions favorable for desired kinetics, hence immersion coils are introduced to remove heat. The bubble diameter sensitively controls the mass transfer and its size changes due to a variety of factors as discussed later. Uniform dispersion of catalyst particles is essential for gas conversion. All these factors are generally interactive and the column hydrodynamics is a very complicated crucial feature of this indirect coal liquefaction techology.

The Fischer-Tropsch synthesis to produce transportation fuels via the indirect coal liquefaction route is successfully carried out in the slurry phase of the bubble column slurry reactor. In the so-called cocurrent bubble column reactor, liquid or slurry and gas are fed at the bottom of a cylindrical column, flow cocurrently through the column and are discharged at the top. The solid phase is a finely divided catalyst and is dispersed by ascending gas bubbles and/or by the upward flow of gas and liquid phases. In cocurrent flow, even large particles of high densities can be suspended. On the other hand if the solids are suspended only by rising gas bubbles, only small and light solids can be fluidized in the liquid phase.

Mashelkar [5], and Heijnen and Van't Riet [6] have reviewed the available information on hydrodynamic, heat transfer, mass transfer and mixing in bubble columns. The importance of the knowledge of bubble size in such columns is pointed out since it has a direct relationship with the transfer coefficient as well as with the transfer area. Interfacial area is inversely dependent on mean bubble size, and the mass transfer constant is directly dependent on bubble size, Miller [7]. The bubble diameter depends upon several factors [5]. In general, it is found to be a strong function of the orifice diameter and a weak function of the gas velocity in the orifice at low gas superficial velocities smaller than 0.5 cm/s. At moderate gas

velocities, smaller than 10 cm/s but greater than 0.5 cm/s, the bubble diameter is a stronger function of the gas velocity in the orifice. For gas velocities greater than 10 cm/s, both the orifice diameter as well as the velocity have a smaller effect on the bubble size. Whenever electrolytes are present in water, the ions at the gas-liquid interface generate surface tension and electrostatic potential resulting into relatively smaller bubbles. It is generally believed that the bubble diameter depends upon the electrolyte concentration as well as the type of electrolytes used. Heijnen and Van't Riet [6] have summarized the available information on the dependence of bubble diameter on various parameters in relation to the nature of gas spargers. In the latter category, they have discussed an orifice on perforated disc, a porous disc, and nozzles. Findings are also reported on the bubble size at a distance from the sparger, and in coalescing and non-coalescing media.

In the indirect scheme of coal liquefaction, the synthesis gas is bubbled through a slurry of iron catalyst suspended in molten wax. According to Calderbank et al. [8], the hydrogen diffusion in the liquid phase and the rate of Fischer-Tropsch catalytic reaction determines the overall rate of synthesis. During the process of synthesis finely divided carbon may be formed according to the Boudouard reaction viz., $2CO \rightarrow CO_2 + C$. These fibrous particles are constantly sheared from the catalyst surface and are broken up by the stirring action of gas bubbles and thereby resulting in a fine solid suspension. This increases the viscosity of the liquid which causes enhancement in bubble coalescence resulting in larger gas bubbles. The resulting reduced gas-liquid interfacial area decreases the potential rate of reactant mass transfer from the gas bubbles to the surface of the catalyst. This effect becomes more pronounced as the height of the reactor

increases. Satterfield et al. [9] on the other hand are of the opinion that the increase in the viscosity of the liquid is not caused by the aggregates of carbon particles coalescing in the liquid medium to form agglomerates, and their concentrations are rather too low to cause any appreciable increase in viscosity. According to them [9] this viscosity increase primarily occurs because of the accumulation of high molecular weight hydrocarbons in the liquid. In any case, it is apparent that the viscosity of the liquid phase and surface tension of the gas-liquid interface sensitively controls the bubble dynamics in the column.

Another illustration of the importance of the knowledge of bubble diameter in a bubble column is evident from the interpretation of experimental data dealing with the indirect liquefaction of coal involving the Fischer-Tropsch synthesis in slurry reactors. On the basis of experimental data available in the literature in conjunction with well defined kinetic and bubble column models, Satterfield and Huff [10] have inferred that the rate hydrogen and carbon monoxide synthesis on the iron catalyst in the bubble column is limited by gas-liquid mass transfer. On the other hand Deckwer et al. [11] based on their analysis of investigations with eight different catalysts in the slurry phase of the bubble columns in conjunction with more recent hydrodynamic properties data i.e., gas hold-up, bubble diameter, and liquid side mass transfer coefficient, have concluded contrary to the inference of Satterfield and Huff [10] that the Fischer-Tropsch synthesis process in the slurry bubble column reactors is governed by reaction resistance, mass transfer limitations are considered to be negligible for the known catalysts and the recommended operating conditions. The model employed in their analysis is essentially similar to that of Satterfield and Huff [10] except Deckwer et al. [11] have allowed for the effect of gas

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volume contraction on gas rise velocity and assumed a different model for liquid phase mixing. Attempts to explain and resolve this controversy have only highlighted the importance of knowing with enough certainty the hydrodynamical characteristics of the reactor and in particular of bubble sizes under actual conditions prevailing in the Fischer-Tropsch process. A detailed discussion of this controversy including several works which have appeared in recent years concerning this matter is presented by Saxena et al. [12] and need not be repeated here.

It is thus clear that many hydrodynamical parameters need to be considered at the same time in developing a realistic model for a slurry bubble column if accurate kinetic information is to be generated or a reliable base is to be established for proper design and scale-up of the slurry bubble column. For example, an appropriate model for mixing of the liquid phase [13], different diffusivities of the feed synthesis gas components in the liquid phase [14], changing viscosity of the liquid phase as a result of either carbon production [8] and/or different high molecular weight hydrocarbons [9], gas volume contraction as a result of reaction [11] etc., must be considered in formulating a general hydrodynamic model of the bubble column in conjunction with an appropriate kinetic model. In such a treatment of kinetic and hydrodynamic benavior of bubble columns, interfacial area enters as a sensitive parameter which in turn requires for its determination the gas hold-up in the column and the Sauter mean bubble size under actual operating conditions. While the gas hold-up measurement is rather straightforward, the determination of bubble diameter is involved particularly for an actual slurry bubble column under operation. A fact which further complicates the extrapolation of this information as gathered for one specific system to another different system is its sensitive dependence on the design of gas sparger, column

geometry particularly cross-sectional area and aspect ratio, presence of internals such as measuring and metering probes, size and configuration of cooling coils, etc. The temperature, chemically reactive nature of the column liquid phase, optical opaqueness of the medium, presence of surfactants and flocculents in the column fluid, electrical properties of the medium etc., further compounds the difficulty of measuring the bubble diameter and applying a suitable technique.

In this article, we discuss the various commonly used experimental techniques for the measurement of bubble diameter and elaborate the exploitation of each of the techniques by different workers with particular emphasis to its scope as regards to the range of conditions for which it is adaptable and what refinements, if any, will enhance its applicability for bubble columns operating under somewhat extreme conditions in a hostile environment. We also compare these techniques against each other in the background of their successful potential for adaption in slurry bubble columns employed for indirect liquefaction of coal.

Experimental Techniques For Bubble Size Measurement

A number of techniques have been developed over the years to measure bubble size in liquids and liquid-solid dispersions. One of the simplest techniques is used by Van Krevelen and Hoftijzer [15] in which the gas bubbles are formed at a single orifice opening upwards, rise through a column of pure liquid containing no internals or obstacles and its dimensions are large enough so that the vessel walls do not influence the bubble rise. The liquid is motion free except for that which is induced by bubble motion. The bubbles so formed rise either separately as discrete bubbles or in series forming chain-bubbling. For the former case the bubble diameter is found to be independent of the gas flow rate

and proportional to the cube root of the orifice diameter, while for the latter case the bubble diameter is independent of orifice diameter but increases with increasing gas flow rate. For determining the average bubble diameter, a known number of bubbles are trapped in a calibrated vessel and the total volume determined. Experiments revealed that the gas hold-up per unit column height is independent of liquid column diameter and its height. This experimental device though attractive for its simplicity is limited in its scope and cannot be successfully adopted for slurry bubble columns. The general methods which have been employed with varying degrees of success are:

- The photographic methods,
- (ii) The optical probe methods, and
- (iii) The electrical conductivity probe methods.

Some of the reviews dealing with this topic in general and of varying scope are due to Delhaye and Jones [16,17], Azzopardi [18], Shah et al. [19], and Buchholz and Coworkers [20-23]. The various investigations belonging to each of these categories will now be discussed in a historical perspective.

(i) The Photographic Methods

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The photographic technique is the simplest and a large number of workers have adopted this method to establish the bubble size in liquid columns. The analysis of the film to compute bubble diameter is quite a laborious and time consuming task though in recent years significant improvement and automation have occurred thereby adding to the ease and rapidity of the process. In conjunction with an electronic signal, the optical technique has recently been studied [24] to investigate the complicated process of bubble coalescence in transparent liquids. A

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majority of the investigations have been conducted in not too optically dense dispersions of two phase systems i.e., gas and liquid. In most cases the bubble diameter is defined as the surface-volume mean diameter known as the Sauter diameter, $\mathbf{d}_{\mathbf{S}}$. It, therefore, represents the diameter of a bubble for which the volume-to-surface ratio is identical with that of the entire dispersion. Assuming the bubbles to be spherical of equal diameter, we have

$$d_s = \frac{6H}{A} = \frac{\Sigma nd^3}{\Sigma nd^2}$$

Here H and A are the total gas holdup and total interfacial area respectively, n is the number of bubbles and d is the diameter of bubbles. Some workers [25] have regarded each bubble to be an oblate spheroid and measured its major and minor axes. The volume equivalent average diameter of bubbles as determined from

$$d_{p} = \begin{Bmatrix} n \\ \sum_{i=1}^{n} 6V_{pi} / n_{\pi} \end{Bmatrix}^{1/3}$$

is preferred by some investigators in their computations. y_{pi} is the volume of i-th bubble, n is the number of bubbles, and d_p is the volume equivalent spherical bubble diameter. The bubbles in a two-dimensional bubble column are generally regarded to be oblate ellipsoids having the volume-surface mean diameters equivalent to spherical bubbles. Details of some of the investigations are given in the following.

Zieminski et al. [26] employed a 50cm high plexiglas column of rectangular cross-section of 5cm by 10cm fitted with a porous plate to produce a swarm of bubbles whose diameters were measured photographically. A polaroid camera equipped with a close-up lens was employed for taking the pictures. The camera had an attached electronic flash adapter to

which was connected a stroboscope. The latter produced a flash of 3µs when the shutter triggered. Polaroid films, type 46-L or 146-L, were used for photography and these were projected on the screen for bubble size measurement. To establish the actual dimensions of the bubble, a rod of known diameter was properly placed in the column at an angle to the axis of the camera ensuring that a part of it would be in focus on the film. From the total number of bubbles photographed, some twenty were selected for measurement of their major and minor axes. The bubble diameters were then computed from the knowledge of the actual diameters of the rod and its measurement of the projection on the screen. To ascertain the accuracy of the technique, glass spheres of known diameters were photographed which revealed its magnitude to be \pm 2.3 per cent.

Marrucci and Nicodemo [27] photographed swarms of bubbled formed in a lucite column 100cm high and 8cm square cross-section fitted with . distributors of various types. In particular, bronze porous plates of varying porosities 8, 20 and 70 mm, and a perforated plate having 0.3 mm diameter holes arranged with a lcm square pitch were used. Photographs of the bubbling bed were taken at a height of 45cm above the distributor plate and enlargements were made corresponding to a magnification of six. Zones of the photograph containing at least fifty bubbles were analyzed. The mean diameter was calculated by averaging the bubble volumes and nonsperical bubbles were assumed to be oblate spheroids. No details of the camera, film etc., are given but it is mentioned that the square section of the column reduced the distortions in the photographs.

Ashley and Haselden [28] took photographs of foam using an "open-air spark" technique with a flash duration of less than a μs . The air-water rig comprised of a rectangular plate having the dimensions of 61cm by 30.5cm and perforations of 3.2mm diameter on a 12.7mm triangular

foam as viewed from vertically above the plate were taken using a "speedflash" unit giving a flash duration of about 5µs. The newly-formed bubbles could be seen in the photographs rising through the clear liquid zone near the floor of the plate. No further details of the photographic or analyzing equipment are given.

Akita and Yoshida [29] photographed bubbles in three transparent acrylic resin columns of 250cm height and having square cross-sections 7.7cm \times 7.7cm, 15cm \times 15cm, and 30cm \times 30cm. They showed that a square column gave the same performance as a round column with a diameter equal to the side of the square. Perforated plate and porous plate gas spargers were used with the 15cm square column with the intention to produce relatively uniform bubble size distribution at lower gas rates. Single orifice gas spargers were used with all the three columns to simulate more practical cases. For photographing the bubbles, a single reflex camera and an electronic flash light were placed on the opposite sides of the column. To obtain uniform illumination several sheets of paper were introduced between the column and the flash light. The camera was focussed in a plane approximately midway between the column wall and its axis and at a height of 150cm above the distributor plate. A scale located in this plane served to calibrate the pictures and thereby to establish the absolute size of the bubbles. The bubbles were not spherical but could be approximated by an oblate spheroid, and maximum and minimum dimensions of individual bubbles were scaled on photographic prints. For a single run, several hundred to a few thousand bubbles were photographed in three to ten pictures

Deckwer and coworkers [30-33] have used cylindrical bubble columns of approximate length of 720cm and 440cm to study bubble sizes in

different liquids. The former column had a diameter of 20cm and a cross of nozzles (56) with holes of 1mm diameter as the gas sparger while the latter had a diameter of 15cm and distributor plate was a glass sintered plate with a mean pore diameter of 150µm. The gas-liquid dispersion was photographed at different heights above the gas distributor plate. The bubble size distribution was obtained from the enlarged photographs by scaling about 500 to 1000 bubbles. A particle size analyzer, Zeiss TGA, was employed which classified the bubbles in 48 sorts of different diameter. In another investigation [33] the bubble column employed was 9.5cm in diameter and 135cm in height. Three types of gas spargers; a sintered plate of mean pore diameter of 75µm, 2 single hole orifice of 0.09cm, and a perforated plate having 19 holes of 1.08mm diameter, were used. The photographs were taken 65cm above the gas sparger and about 650 bubbles were analyzed to determine the Sauter mean diameter. A high-grade steel wire was installed near the inside wall as a calibration device. Only bubbles near the wire were employed to size the bubbles and hence determination is almost free from distortions. On the other hand, the photographically determined bubble sizes apply only to bubbles located in the column region near the wall.

Sagert and Quinn [34] have measured coalescence times for bubbles of hydrogen sulfide and carbon dioxide on adjacent nozzles in water at pressures up to 3.5MPa. High speed cinematography is used to follow the coalescence events and interesting details are given by the authors and these are reproduced here. A 600W quartz iodine lamp was used as light source and its intensity could be varied. Bubbles were filmed by a Red Lake model K20S4E Hycam camera equipped with a 75mm extension tube provided with a 100mm Elgeet Cinetel lens opened to f5.6. The

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bubbles of radii close to 1.5mm filled the frame with the bubble images. The full 16mm frame was used at a shutter speed of 1/2.5 of the framing speed which were varied in the range 500 to 5000 frames per second. To provide adequate light for good exposure of the film, an X-L light meter of Photographic Analysis Co. was used to determine the lamp voltage. The film speeds were established from a calibrated timing light, a Red Lake Millimite TLG-3, which produced dots along the side of the film at 10ms intervals. For majority of the experiments, Kodak 4X negative films, 400ASA-tungsten, were used. For lower speeds 200ft rolls were used but at higher speeds 400 ft. rolls were found necessary to film several events. This technique has been successfully employed by the authors [35] to measure the lifetimes of thin aqueous films formed between nitrogen bubbles in dilute solutions.

Bhavaraju et al. [36] have investigated the bubbling characteristics of air in a 0.15m square plexiglas column of 1m height from vertical nozzles of three different diameters. Liquids used in the experiments were water and aqueous solutions of carbopol. To obtain bubble sizes in the columns, photographs were taken in two regions, one within 0.1m from the nozzle and another at about 0.8m from the nozzle. No further details of the photographic equipment are given by the authors.

Yamashita et al. [37] measured the bubbles generated in a two-dimensional bubble c lumn made of transparent acrylic resin plates, 100cm high and having a rectangular cross-section of 30cm width and 3cm thickness, by adopting a photographic technique. A performated plate distributor made of brass, 0.7mm thickness, with twenty-nine holes of 0.5mm diameter arranged at 1.0cm pitch on the longer central line of the plate. Bubbles were photographed at a position situated from 70 to

80cm above the gas distributor and within about 5cm horizontally from the central axis of the column. No details of the camera or related equipment are given. For a single run, one hundred to two hundred bubbles were measured from projections which were so adjusted that the bubble sizes were enlarged by about 3 to 4 times of the actual sizes. Most bubbles were not spherical but could be approximated by oblate ellipsoids and on this basis the volume and surface area of each bubble were calculated and hence the volume surface mean diameter equivalent to sphere. Bubbles with the major axis smaller than 1.0mm were not counted since their contribution to gas holdup and interfacial area could be assumed to be negligibly small. Two mean diameters were computed, d' and d, defined as follows:

 $d' = \sum_{i=1}^{n} d_i^3 / \sum_{i=1}^{n} d_i^2$,

and

$$\overline{d} = \sum_{i=1}^{n} d_i / n$$

Here d_i is the arithmetic mean of the maximum diameter and minimum diameter of the i-th bubble, and n is the number of bubbles counted. The measured volume-surface mean diameters were found to be in good agreement with the computed diameter values of d' and \bar{d} .

Kunugita et al. [38] photographed the motion of a solid particle and the dynamic behavior of bubbles (especially bubble coalescence) in a clear acrylic plastic column of diameter 5.0cm and height 100cm and equipped with a porous plate distributor of 60µm pores. To prevent distortion, the bubble column was surrounded by a clear acrylic plastic box of square cross-section and the space between the box and the column was filled with the same liquid (water) as the column. Six mirrors were placed around the bubble column and adjusted to take two mutually perpendicular views of the column simultaneously. The camera

employed was a Nikon F Motor Drive with telephotolens (Nikkor-Q Auto 1, 3.5) of 135mm focal length. The films used were Fuji Neopan SS (ASA100). Otake et al. [39] have utilized this technique to study gas holdup in bubble columns in both cocurrent and countercurrent gas-liquid flow systems. The experiments were conducted in a cylindrical glass column, 50cm in diameter and 1.50m in height, and for two types of stainless steel multinozzles distributor plates and for two types of single nozzles. Bubble size was computed by the arithmetic average of the maximum and minimum dimensions of each rising bubble on at least 150 photographed bubbles.

Klug and Vogelpohl [40] have studied the formation of bubbles; at a single-hole plate and at a sieve plate with large hole spacing. The column consisted of a perspex cylinder having an overall height of 1.5m and inner diameter of 0.172m. The holes in both distributor plates were 1mm in diameter. A plane viewing window was provided to eliminate optical distortions which would otherwise occur at the curved reactor wall. Bubble size was determined by high-speed photography with a frequency of 350 frames per second. Each developed film was manually evaluated on a transparent screen. The films were projected on the latter by means of an appropriate optical system. With the use of an analog-digital processor, the bubble data were recorded and transmitted directly to a desk computer. Sada et al. [41] measured the bubble size in a column with a single nozzle for gas-molten salt systems. A transparent pyrex glass bubble column, 7.3cm inside diameter and 95cm in height, provided with four polished glass windows at heights of 7, 17,32 and 52cm from the bottom plate of the column to photograph the bubble size, was used. As gas spargers were used three single nozzles of inside diameters 1.5, 2.7 and 5.7mm. Enlarged prints of the bubble photographs were made and

several hundred to a few thousand bubbles were scaled in 10 to 15 prints for a single run. The individual bubble was sized by an arithmetic mean of the maxinum and minimum dimensions scaled on the prints. The volume-surface mean diameter of the swarm bubbles in the column was calculated from an equation similar to that given above for d'.

In the above, so far we have discussed the works which have employed the photographic technique for bubble columns involving only two-phases viz., gas and liquid. We will now review those efforts which pertain to three phases viz., gas bubbling in a liquid column with suspended solids. Stewart and Davidson [42] photographed air bubbles in water fluidized glass Ballotini, iron shot and lead shot with a mean diameter of 0.46mm. A two-dimensional fluidized bed contained between two perspex plates 6.35mm apart was employed. The bed was strongly illuminated from the front to show the particles. The photographs were taken with an exposure of 1/500 s, some of them were taken with the camera mounted on a platform which could be moved up with the bubble.

Page and Harrison [42] photographed the size distribution of bubbles produced in a cylindrical glass column 22.8cm in diameter containing nearly spherical close size range 500µm sand particles fluidized by water and bubbles of air. Two types of gas distributor were employed to generate large and small bubbles. The former were produced from a 0.635cm diameter single orifice pointing vertically upwards while the latter were produced from a distributor with ten 0.154cm diameter holes arranged in a straight line across the diameter of the column. A wire gauze was placed over the top of the column to minimize the loss of sand particles. The upper part of the column was encased in a watertight perspex box 60cm high and with a 27cm square base. Optical distortion could be eliminated by matching

the refractive indices of the liquids inside and outside the column with the material of construction of the column. However, they found that the distortion of the bubbles due to differences of refractive index was negligible and was probably less than the distortion of the bubbles caused by irregularities in column glass which can occur during manufacture. Behind the column bed, a white screen was uniformly illuminated by a quartz-iodine floodlamp and the sides of the box which encased the column were darkened by black paper. This lighting gave good definition of the bubble edge, and it also introduced a useful three-dimensional effect because bubbles near the camera were illuminated by light incident from a smaller angle than those away from the camera. In this way near bubbles appeared very dark at the edges, with a small central light region where light passed straight through the bubble without deviation. Bubbles far from the camerawere illuminated from a larger angle, appeared much lighter, and the dark rim around the bubble was much thinner. These differences were found particularly helpful in the analysis of the film.

Cine-photographs were taken of the bubbles employing a Mitchell HS16-F2 camera using a 7-1/2° shutter at eight frames per second and a 25mm Dallmayer lens. A very large number of bubbles, up to as many as 3500 in a single run, were recorded and special care was taken to avoid measuring the same bubble twice. The 16-mm film was projected onto a film analyzer which produced an image on a horizontal ground-glass screen. To obtain the bubble diameter and the bubble spatial distribution, a special equipment referred to as the x-logger in one dimension was used.

Henricksen and Ostergaard [44] photographed single bubbles rising in a two-dimensional Perspex column, 112cm high and 39.5cm wide, which contained a liquid and glass beads of diameters 0.2mm, 1mm and 3mm.

The separation between the two plates was 8mm at the sides of the column and 9mm at the middle. The liquid distributor was 3mm thick sintered bronze plate with a mean pore diameter of 25µm. Single gas bubbles were injected through a 0.55cm internal diameter tube located 55cm above the distributor. At this location the fluidization had become smooth and uniform.

Sixteen mm movie films of the fully developed bubbles were taken with a Bolex camera fitted with a 38.1mm, f/2 lens. The camera speed was either 24 or 32 frames per second. A white PVC powder was suspended in the liquid to show the bubble wake. On the back of the column, a black cardboard was placed and the column was illuminated from the front with two photofloods. The column was illuminated with direct light from behind while photographing bubbles in fluidized beds of particles. The analysis of the single frames of the films was conducted by projecting it on a measuring table. These authors [45] also examined the state of bubbles in liquids and three-phase fluidized beds as solid particles fell through them. Single steel spheres (5mm diameter) and glass spheres (3 or 6mm diameters) were dropped above the bubbles and fell through them, and this meeting was photographed with a movie camera at about 200 frames per second from two perpendicular directions in the same horizontal plane. In none of the cases examined did the bubbles disintegrate as the spheres fell through them.

Miyahara et al. [46] measured the bubble diameters in acrylic-resin columns of internal diameters of 5, 10 and 15cm and equipped with brass perforated plates with holes arranged either in the form of equilateral triangles or a square. To eliminate optical distortion during photography of the bubbles, the columns were enclosed in a transparent acrylic-resin

box filled with the test liquid. The photographs of the bubbles were taken at intervals of approximately 5cm along the column, the light being moved up and down with the camera. The photographic negatives were enlarged, and major and minor axes of each bubble, which was assumed to be an ellipsoid, were measured for as many as 40-100 bubbles for any given condition. The volumetric mean bubble diameter, $\mathbf{d}_{\mathbf{p}},$ was computed from the expression given earlier and this number was considered as reliable for statistical purposes. In another investigation, Miyahara et al. [47] employed a right-angled parallelepiped of transparent-acrylic, $10 \times 10 \times 60$ cm column, and photographed the bubbles with the aid of a Hycam 100 model FT high-speed camera, 600-800 frames per second. The enlarged negatives were analyzed. These authors [48] have also employed this photographic technique to examine a chain of bubbles rising in a transparent acrylic resin column with an internal diameter of 10cm which was partly enclosed in a transparent acrylic resin viewing vessel filled with the test liquid to avoid optical distortion.

Recent technological advancements have added a new dimension to the applicability of this photographic technique in as much as sophisticated high speed cameras are available as also high speed automated film analysing equipment. One such facility is reported by Saxena et al. [49] and utilized by them in photographing the bubble motion in a two-phase gas-solid system in a two-dimensional bed [50]. It is comprised of a Nova high speed camera, Soligor spot sensor and a photo-optical data analyser. The Nova high-speed camera is a continuously moving film type motion picture camera employing a rotating prism as an optical compensator and shutter. The camera is capable of filming 10 to 10,000 pictures per second (pps) in the 16mm format and 20 to 20,000 pps in the 8mm format.

The basic camera body has a 100 ft film capacity but is provided with accessories to accommodate an auxiliary film magazine of 400 ft capacity. Two timing lights which impart a density on the edge of the film are mounted in the basic camera body. These timing lights are used with a timing light generator which has a self contained power supply and provides counts of 10,000 and 1000 cps square wave directly to the camera's built-in timing lights. The camera is equipped with several high quality lenses viz., Pentax 1: 1.4/50, Zoomar 1: 2.8/90 and 49mm closeup lenses No. 1, 2, and 4. The camera is provided with two 140 V AC-DC series wound motors to operate it at frame rates from 200 pps to 10,000 pps. These are energized by a 140V DC power supply. While filming, the camera is mounted on a Samson Quickset tripod. The test section of the bed column to be photographed is properly illuminated with two mounted flood lights each having four bulbs controlled by four switches. A Soligor spot sensor II meter with $\mathbf{I}^{\mathbf{O}}$ angle of incidence is employed to establish appropriate combination of film speed and lens opening for a specific photographic requirement.

The film analysis equipment comprises of a LW224A photo-optical data analyser, LW110C photo-optical digitizer and LW1224 electronic graphics calculator connected to a Teletype 43 terminal and tape-printer which is interfaced with an IBM 370/3081 computer. The photo-optical data analyzer can accommodate an unusually wide range of 16mm film viewing modes, from stop motion, frame by frame, to eighteen or to normal twenty-four frames per second. The digitizing system is a very simple and convenient way for the visual analysis of films. Here, together with the graphics calculator, it is capable of continuous measurement of length and area which are outputted to the tape-printer or to the terminal for on-line computer processing.

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