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Measurement of Liquid Film Flow on Inclined Wall using Photochromic Dye Marking Method

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Chapter 1 Introduction

1.1 Review

Falling liquid films are widely used in several industrial applications such as food processing, seawater desalination, medical equipment and electrical component manufacturing industries, which mainly involve with heat and mass transfer process. The presence of interfacial waves occur in the falling films undeniable has become a major influence to the performance of heat and mass transfer process. Therefore, in response to demands of products with high efficiency heat and mass transfer process, a complete knowledge of falling liquid film behaviors that characterized by the velocities and local instantaneous thickness is of great important. Over than half a century ago, many researchers have devoted their efforts to develop the film flow measurement techniques. V.V Lel et al. [1] in their work have listed out various study of measurement techniques presented in the past, including the capacity method by Duckler and Berglin [2], the contact or needle-contact method by Ishigai et al. [3], the interferometric method by Unterberg and Edwards [4], the light absorption method by Portalski [5], and many more.

However, we found that most of the techniques in the previous work require complicated optical system or devices arrangement. Several others also have their limitations of applicability in certain flow field or needed to contact the flow field which might interrupt the flow structures. Based on this situation, we would like to propose a non-intrusive technique, called as photochromic dye marking method. It is just a color tracer technique and the crucial advantages lies on its non-intrusiveness and applicability in measuring very thin liquid films. It does not require particle seeding like the PIV measurement method which might not satisfactorily follow wavy films or applicable in very thin films.

The photochromic dye marking method was first developed by Popovich and Hummel [6] in 1967. Since then, its uses have been widely applied to develop variety of research fields, especially in the flow field measurements. For instances, considering the difficulty in precisely predicting heat and mass transfer during drop formation and release, G. Johnson and E. Marschall [7] implemented an investigation of flow mechanism for dispersed phase during drop formation and release in various flow fields using this technique as the visualization method. They stated the ability of this technique to provide precise velocity information. They used the petroleum solvent as the working fluid and conducted the experiments between the limits of pseudo-static

flow and the onset of jetting flow.

M. Kawaji [8] described several sample experiments conducted by his research team in order to prove the benefit of this technique in visualizing the two-phase flow. Sample experiments carried out include the wavy-stratified, free falling film, annular and slug flows. The investigation has reached better understanding of the flow structures and made supports and denial to some assumptions made earlier before. D. Homescu and P. Desevaux [9] applied this technique to determine the liquid free surface velocity on curved surfaces. The ability was shown on the silicone oil film flowing around finned tubes. The advantages and limitations of using this technique were also described in the paper.

H. Park et al. [10] used this technique to study a two-dimensional steady flow in a curved vessel. They measured the velocity fields and also estimated the vorticity, wall shear stress and pressure fields. In order to ensure the measurements accuracy of the method, the numerical flow field calculations were also performed. The comparison between both results was done and the validity of the laser tagging method was then strengthened from the good agreement of it. T. Kai et al. [11] proved the successful of using this technique to visualized particle movement in a two-dimensional fluidized catalyst bed. They enabled to measure the local velocity of particles around bubble and observed the bubble behavior without disturbing the flow pattern.

This technique was also applied by T. Sanada et al. [12] to visualize a bubble wake. They used silicone oil solution of photochromic dye to visualize the flow structure in the rear of a single rising bubble in a quiescent liquid. Note that appropriate photochromic dye must be assessed carefully in order to gain a successful application of this technique. In this matter, T.W. Fogwell and C.B. Hope [13] studied the properties and usefulness of photochromic dye in details. They presented the mechanisms and theory of chemical changes in the properties of many types of photochromic dye. Their investigation especially in describing the dye concentration is greatly beneficial in using photochromic dye properly for the flow visualization.

In addition to the above-mentioned examples, there are also several kinds of image processing method have been applied in the liquid flow investigations. S. Hosokawa, T. Fukunaga, and A. Tomiyama [14] improved a photobleaching molecular tagging velocimetry to evaluate turbulence energy budget in bubbly flows. They improved the method, for accurate evaluation of velocity gradients that reflected from the deformation of the shape of the tagged region in the fluid. The turbulence properties in single-phase and two-phase dilute-bubbly flows in a square duct were measured using the improved method. P.T. Tokumaru and P.E. Dimotakis [15] developed image

correlation velocimetry method, focusing on the correlation of two successive images of convected scalars, which was the dye markers carried by a fluid to measure the fluid motions. N. Santitissadeekorn and E.M. Bollt [16] presented a computational method to approximate the Frobenius-Perron operator from successive images. They reconstructed the velocity field from image sequences based on theory of infinitesimal generator of the Frobenius-Perron operator and related to famous optical flow problem.

In recently, we have successfully applied the photochromic dye marking method in visualizing detailed flow structures of creeping flow and liquid film jet [17]. Excimer laser (λ= 248 nm) and Nd: YAG laser (λ= 355 nm) were used to form the photochromic dye traces in the flow tests. From the movement of the dye traces, velocity vector field was measured. Vorticity and divergence of velocity vector field were also calculated.

1.2 Objective

The objective of present work is to apply a liquid film measurement technique, which is called as photochromic dye marking method. This technique utilizes a color change of the photochromic dye contained in the test film when ultraviolet (UV) light from Nd: YAG laser (λ= 355 nm) is irradiated. The movement of the dye trace formed by the UV irradiation is then captured by a high speed video camera and analyzed to obtain the surface velocity. The movement of wave passing by the dye trace is also analyzed and comparison between them is made to investigate the velocity change. The data regarding such information is nowadays lacking.

Apart from the measurement as mentioned above, this paper also aimed to produce the average waves velocity and film thickness results data. The wave velocity is measured by cross correlating two signals of light intensity from two laser beams spaced in a known distance; parallel with the film flow direction. Taking advantages from one of the laser beams sourced from a diode laser light (λ= 407 nm), the thickness is determined based on the intensity of absorption light passing through the test film contained a fluorescent dye, coumarin-153. This technique is a non-intrusive and easy technique to be conducted, as it only requires simple optical arrangement and calibration process. The data from this study are also compared with previous published results [1], [18], [19].

The experiment is conducted at different inclination angle from the horizontal direction (θ= 30°, 60°, 90°). The effects of Reynolds number (Re= 50.8, 81.3, 108.4, 138.9) and film flow characteristics at various positions on the inclined wall defined as distance from liquid inlet (x= 60 mm, 100 mm, 140 mm, 180 mm) are investigated.

Chapter 2 Experimental apparatus and methodology

2.1 Experimental apparatus

Figure 1 illustrates the overview of experimental apparatus used in the present study. The photograph of experimental apparatus setup is shown in Fig. 2. It is comprises of 3 main sections which are test section of liquid film flow, optical arrangement for laser irradiation and data analysis system. The schematic view of the optical arrangement is illustrated in Fig. 3.

2.1.1 Inclined wall

The photograph of inclined wall setup is shown in Fig. 4. The inclined wall is made of acrylic plate composed with the reservoir where the test liquid was first accumulated here before flowing down the wall. The wall surface to allow full development of liquid film flow is in 300 mm length and 120 mm width. The whole inclined wall is mounted on aluminum frames installed with free-angle brackets which allow the inclination angle to be changed from vertical up to horizontal direction. A screw rod mechanism is also installed to the frames of inclined wall to easily change the measuring position on the wall.

2.1.2 Kerosene oil

In this study, we employed the kerosene oil dissolved with photochromic dye and coumarin-153 as the test liquid. It is circulated to flow on the inclined wall by the pump shown in Fig. 5 and stored in the liquid storage basin as shown in Fig. 6. The property of kerosene oil is shown in Table 1.

2.1.3 Photochromic dye

The most important material in this study is the chemical compound which is called as photochromic dye. There is a lot of chemical compound provide photochronism properties in their usage. However, only a small number of them are applicable to the fluid flow analysis, such as spyropiran group.

In this study, we used the photochromic dye of 1'3'3-trimethylindolino-6'-nitrobenzopyryospiran from spyropiran group as shown in

Fig. 7. Figure 8 shows the molecular configuration in the dye upon irradiated with ultraviolet (UV) light. The absorption spectrum is shown in Fig. 9. Before undergoing the UV light irradiation, kerosene oil solution containing the photochromic dye appear in a colorless state due to the high absorption band which merely occurs in the UV-ray area not in the visible ray. At this time, irradiation with UV light will result a molecular configuration of the dye and yield the change in the absorption spectrum. Thus, the absorption band in visible ray area would highly arise and cause the irradiated section in the solvent turn into a color state.

Generally, we use photochromic dye with concentration of 0.1~0.2% by weight. However, appropriate concentration for the best experimental result is depends on the strength of the UV laser light. In other words, if the concentration of photochromic dye used is low, UV laser light will pass through the working fluid easily and prolong the lifetime of the dye coloration. However, the visualization will become harder as the color appear faded and unclear. On the other hand, light energy absorption of photochromic dye in high concentration will turn the incident light of the UV laser into heat energy and thus affect the overall property of the flow field. Therefore, considering appropriate concentration of photochromic dye used is vitally important to achieve the best experimental results.

2.1.4 Coumarin-153

Coumarin-153 is a fluorescence dye usually used to determine liquid film thickness as it has a great potential to attenuating light intensity. It is a well known dye, probably one of the most often used in fundamental studies of the environment influence on fluorescence properties of a molecule. It is a fairly simple molecule, strongly emissive, which can be used as perfect model for much more complicated systems. Figure 10 shows the coumarin-153 used in this study. The molecular structure is shown in Fig. 11.

Note that it is essential to choose a fluorescence dye which is able to absorb specific wavelength in the region outside the photochromic dye reaction. Due to this reason, we choose the coumarin-153 whose absorption band around 400~420 nm as the fluorescence dye used in the present study. As shown in Fig.12, photochromic dye mainly does not fall into the absorption band of coumarin-153.

2.1.5 Laser

(i) Nd: YAG laser

Figure 13 shows Nd: YAG laser used in this study. The prospect for this laser is shown in Table 2. The light emitted from this laser is 1064 nm wavelength. As illustrated in Fig. 14, the ultraviolet (UV) light is extracted from visible light using combination of lens and mirror, and then focused to the measuring spot on inclined wall with appropriate size.

Nonlinear crystals of beta barium borate (BBO) crystals are placed on the laser route in order to form the 355 nm wavelength UV light. In detailed, the first BBO crystal is used to extract the second harmonic light, 532 nm wavelength from the fundamental light, 1064 nm wavelength. The second BBO crystal then enables the formation of third harmonic light, 355 nm wavelength which extracted from the 532 nm wavelength and 1064 nm wavelength. A harmonic separator is used to reflect the unwanted 1064 nm and 532 nm wavelengths. The actual image of the optical arrangement is shown in Fig. 15.

(ii) Diode laser

Figure 16 shows the diode laser used in this study. The prospect for this laser is shown in Table 3 and the spectroscopic result is shown in Fig. 17. In this study, the original diode laser beam is divided into two parallel beams using a beam splitter and a mirror. The optical flat used for the liquid film thickness calibration process is also placed on the beams path. The optical arrangement for the diode laser irradiation is shown in Fig. 18. The two beams passing through the liquid film is conducted to the optical fibers placed on the back side of the inclined wall. The optical fibers are hold by a metallic holder as shown in Fig. 19. A blue interference filter as shown in Fig. 20 is used to prevent unwanted wavelength from the fluorescence light of test liquid. The prospect of the optical fibers and the measure guideline is shown respectively in Table 4 and Fig. 21.

2.2 Experimental methodology

The definition of experimental parameters applied in the present study is illustrated as Fig. 22. The experiment is conducted in different wall inclination angle defined as θ, in 30°, 60° and 90° with respect to the horizontal position. Measurement is done at various positions on the test section of inclined wall, represented by distance from liquid inlet defined as x. Liquid film is allowed to flow on the test section in width defined as b. The flow rate defined as Q is controlled by a flow meter. The liquid film velocity defined as V_l is measured using the photocromic dye marking method, while the wave velocity defined as V_w is measured using image analysis method and cross correlation method. The wave velocity V_w together with the wave frequency defined as f are then used to determine the wavelength defined as λ. The liquid film thickness defined as δ is measured using light absorption method and the amplitude defined as A is determined from the difference between a minimum and maximum value of film thickness.

The Reynolds number, Re is defined as below equation:

$$Re = \frac{\rho Q}{\eta b} \tag{1}$$

Where,

ρ : Density
Q : Flow rate
η : Viscosity
b : Test section width

The experimental condition for the experiment is written as below:

1. Different position on inclined wall (x= 60 mm, 100 mm, 140 mm, 180 mm) with constant Reynolds number (Re= 81.3)
2. Different Reynolds number (Re= 50.8, 81.3, 108.4, 138.9) with constant position on inclined wall (x= 140 mm)

In this study, kerosene oil dissolved with 0.12wt % of photochromic dye and 0.5 g/l of coumarin-153 is used as the test liquid. The experiment methodology is outlined as below:

① The test fluid is circulated by the electrical pump to enter the reservoir area in the inclined wall. Then, it flows down the wall due to the gravitation and comes back to the liquid storage basin.

② The liquid flow rate is controlled by a valve and the passing amount can be read by the flow meter. Another nylon tube is connected to the basin to drain excess liquid during the flow rate reduction, and also to prevent the temperature rise of the liquid test caused by the pump rotation.

③ Upon the liquid film flows down the wall, a pulse of diode laser beam (λ= 407 nm) is irradiated to the liquid film flow for the average wave velocity and film thickness measurement. Half of the original beam passes through a beam splitter placed on the beam route, while another half divided by the beam splitter is reflected to a mirror and conducted to the liquid film flow. The two beams with a known spacing between them (S= 12 mm) thus appear on the liquid film flow with parallel direction of the film flow. Considering the effect of liquid film thickness results from the optical flat used in the calibration process, the optical flat is also placed on the route of the two beams.

④ On the back side of the inclined wall, a pair of optical fiber (namely A and B) hold by a metallic holder is attached with their surfaces covering the regions of the total incident light of the two beams. A blue interference filter is placed between the inclined wall and the optical fibers in order to merely conduct light intensity passing through the liquid film to the optical fibers, and reflect the unwanted light intensity such as the light intensity of the fluorescence test liquid.

⑤ The light intensity conducted by the optical fibers is then converted into voltage signals by two photomultipliers which are connected to the opposite tip of the optical fibers. The electrical energy for the photomultipliers is provided by a high power voltage supply shown in Fig 23. The average wave velocity is measured by cross correlating two signals of light intensity from the two beams using the fast Fourier transforms (FFT) analyzer shown in Fig. 24.

⑥ Taking advantages from one of the laser beam irradiation, the average film thickness is measured based on the intensity of absorption light passing through the test liquid contained the fluorescent dye, coumarin-153.

⑦ On the other hand, pulses of ultraviolet (UV) laser light from Nd: YAG laser is irradiated to the liquid film flow in order to form a dark trace of photochromic dye for liquid film and wave velocity measurement. Beta barium borate (BBO) crystals are used to obtain the third harmonic light of 355 nm wavelength.

⑧ By using combination of lens and mirror, the UV light is lead to the liquid film flow preferably in between the incident light of parallel diode laser beams. A convex lens placed on the laser path is used to focus the UV laser light in order to gain a concentrated dye trace for good analysis.

⑨ Simultaneously, the flow patterns and the movement of wave and dye trace appeared in the liquid film flow is recorded by a high speed video camera shown in Fig. 25 (Spec of the camera is shown in Table 5). In order to determine the surface velocity, the motion picture is then transferred to a still image data and analyzed on the computer by digitizing the coordinates.

⑩ A halogen lamp is also used to provide lighting for best flow visualization.

2.2.1 Liquid film and wave velocity measurement (Image analysis method)

Sample images of the liquid film flow obtained in the experiment are shown in Fig 26. As indicated in Fig. (a), the dark trace represents the photochromic dye trace marked by the UV irradiation, while the strip lines represent the surface waves occurred in the liquid film flow. Brightness and contrast of the image is altered to get a higher contrast appearance between the liquid film, waves and dye trace, as shown in (b). Then, the coordinate of dye trace and wave in x-direction is digitized.

We could assume the dye trace velocity as the liquid film velocity as the dye is believed to move along with the liquid film flow. Furthermore, the dye trace we analyzed in the present study is chosen based on the image that a dye trace appeared in front of a wave strip. Fig. 27 shows the movements of dye trace from the beginning when it is formed in the film flow until passed by a wave. From this, the change of movement for both liquid film and wave could be observed and thus velocity difference between them could be obtained.

The liquid film velocity, V_l is obtained from the displacement, ΔX_l over consecutive frames during the time interval, Δt. The wave velocity is also measured as the same method. Note that these velocity profiles in the present study are only obtained

for x-component direction, and the data is taken for 60° and 90° of inclination angle only. The equation is written as below:

$$V_l = \frac{\Delta X_l}{\Delta t} \tag{2}$$

Where,

ΔX_l : Liquid film displacement over consecutive frames
Δt : Time interval
V_l : Liquid film velocity

2.2.2 Average waves velocity measurement (Cross correlation method)

As a wave propagates downstream, it passes over each of the diode laser beams which are lead with parallel direction of the film flow. If the distance between the two beams is sufficiently small, the wave characteristics changes very little over the distance. Thus, waves induces nearly identical voltage signals in each of the photomultipliers.

The voltage signals from the photomultipliers depend on the total incident light on the surfaces of the two optical fibers, while the total incident lights depend on the liquid film thickness and wave structure. Therefore, it is essentially needed to ensure the surfaces of the two optical fibers cover the incident light perfectly. The temporal distance of the two identical signals can be found by cross correlating the two signals using fast Fourier transform (FFT) analyzer. A sample image of the cross correlated voltage signal obtained by the FFT analyzer is shown in Fig. 28.

The waves velocity can be determined from the known spacing of the two beams (measuring position, S= 12 mm) along with the time maximum, $\Delta\tau_{max}$ at which the cross correlation is achieved. Note that waves velocity profiles for this measurement system are obtained from the average of 16 events. The calculation theory is written as below:

$$V_w = \frac{S}{\Delta\tau_{max}} \tag{3}$$

Where,

S: Distance between two diode laser beams
$\Delta\tau_{max}$: Time maximum at which cross correlation is achieved
V_w: Waves velocity

Moreover, comparison has been made between the wave velocity obtained by the cross correlation and image analysis method. Fig. 29 and 30 show the wave velocity of different inclination angle respectively at different Reynolds number and position on inclined wall. It is plotted with the cross correlation method versus the image analysis method. As can be observed from the figures, the data points seem to fit with the line, showing a good agreement between results obtained by both methods. We also found that wave velocity at all angle increases with the increasing of Reynolds number and distance from liquid inlet.

2.2.3 Average waves frequency and wavelength measurement

Figure 31 shows the sample image of voltage signal B from light intensity of optical fiber B for waves frequency measurement. The waves frequency is obtained from the time of events, n resulted within a specific time period, τ. Note that the waves frequency data in this study is taken from the average of 3 times events in same experimental condition. The waves frequency, f and waves velocity, V_w obtained in the above Eq. (2) are then can be used to determine the wavelength, λ. The calculation theory is written as below:

$$f = \frac{n}{\tau} \tag{4}$$

$$\lambda = \frac{V_w}{f} \tag{5}$$

Where,

n : Times of events
τ : Length of the time period
f : Waves frequency
V_w : Waves velocity
λ : Wavelength

2.2.4 Average liquid film thickness measurement

Liquid film thickness in this study is measured based the light absorption method. The absorption is described by the well-known theory of Lambert-Beer law. As shown in Fig. 32, the amount of light penetrating a solution is known as transmittance, expressed as the ratio between the intensity of the transmitted light, I and the initial light intensity of the light beam, I_0:

$$T = \frac{I}{I_0} \tag{6}$$

Where,

T: Transmittance
I: Intensity of the transmitted light
I_0: Intensity of the initial light beam

Even though the relationship between transmittance and absorbance would appear to be a simple inverse relationship, the true relationship between these two variables is inverse and logarithmic:

$$T\% = 100T = 100 \times \frac{I}{I_0} \tag{7}$$

$$A = \log(\frac{1}{T}) = -\log T \tag{8}$$

Where,

A: Absorbance
$T\%$: Transmittance percentage

The absorbance A of a dissolved substance is a linear function of its concentration, the so-called Lambert-Beer Law. The length of the light path (thickness of the cell) and the extinction coefficient (a substance specific constant) determine the slope of the linear plot.

$$A = \varepsilon cd \tag{9}$$

Where,

c: Concentration
d: Thickness of the cell

ε: Extinction coefficient

In this study, a calibration process is needed to determine the linear function of coumarin-153 absorbance. The set up of the calibration process is illustrated in Fig. 33 and the photograph is shown in Fig. 34. It is carried out by sandwiching a known thickness plate with an optical flat to the inclined wall together. The thicker space the more test liquid could be filled into the sandwiched device (probe), and thus causes the higher light intensity absorbed by the coumarin-153 contained in the test liquid. The voltage signal A and B, respectively from the light intensity of laser beam A and B is recorded along various positions from the bottom part of the optical flat. The voltage output is expected to decrease with the increasing of positions from the bottom part due to the thickness increasing.

As the value of output voltage would change even with a very little change of the light incident position on the optical fibers, the calibration process in this study is made for every inclination angle, θ= 30°, 60° and 90°. Fig. 35 shows the calibration results of voltage signal B for all inclination angles. The film thickness, δ is shown to be linearly related to the voltage output, E. Thus, the expected film thickness linear dependence on the absorbance is confirmed.

Figure 36 shows the sample image of voltage signal B from light intensity of optical fiber B for film thickness measurement. Based on the linear function of absorbance obtained from the calibration result depicted in Fig. 35, the film thickness can be determined. As the FFT analyzer used in this study is an old device with no data collecting system direct to the computer, we used a simple software named 'Simple Digitizer' in order to digitize the voltage output image and plot into graph. The sample image of film thickness result after digitizing is shown in Fig. 37. Same as the average wave velocity measurement, the liquid film thickness is also obtained from average of 16 events. The amplitude is also determined from the difference between a minimum and maximum value of film thickness.

Furthermore, the measured data in this study is averaged and compared to the well-known Nusselt's theory of laminar flow which is given as below equation:

$$\delta_{Nu} = \sqrt[3]{\frac{3\nu^2 Re}{g \sin(\theta)}} \qquad (10)$$

Where,

ν: Kinematic viscosity of kerosene oil
Re: Reynolds number
g: Gravity acceleration
θ: Inclination angle
δ_{Nu}: Nusselt's theoretical liquid film thickness

In order to make comparison with previous studies in the past, the data is presented in terms of dimensionless mean film thickness as expressed by below equation:

$$\delta^*_m = \delta_m\left(\frac{g\sin\theta}{\nu^2}\right)^{(1/3)} \tag{11}$$

The comparison is made with the data of previous studies reported by V.V. Lel et al. [1], T.D. Karapantsios et al. [18], and G. Kosky [19]. The correlation is respectively written as below equations:

$$\delta^*_{m,Ka} = 0.451Re^{(0.538)} \tag{12}$$

$$\delta^*_{m,V} = 1 + 0.615Re^{(0.47)} \tag{13}$$

$$\delta^*_{m,Ko} = 0.05121Re^{(0.875)} \tag{14}$$

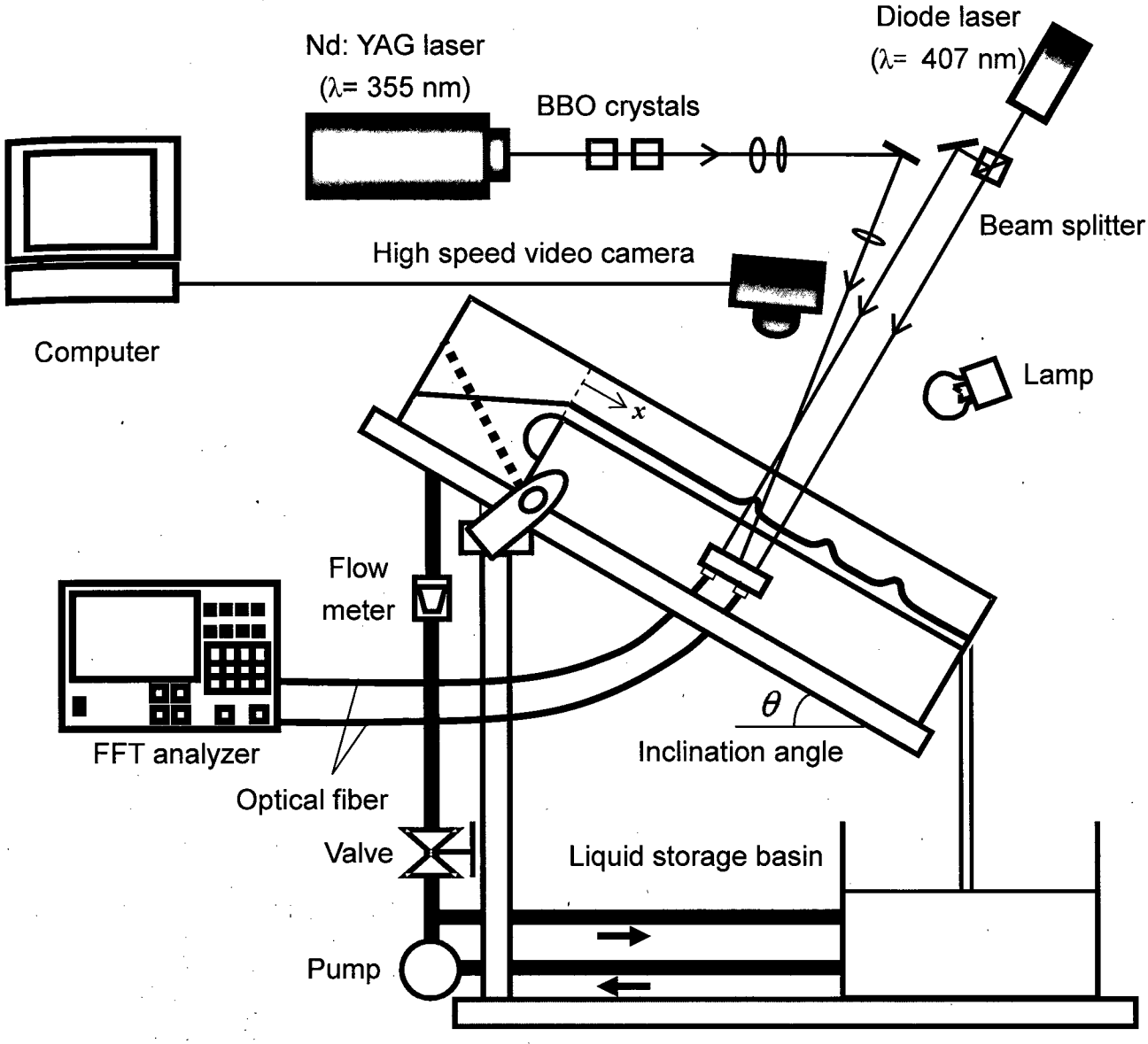

Fig. 1 Overview of experimental apparatus

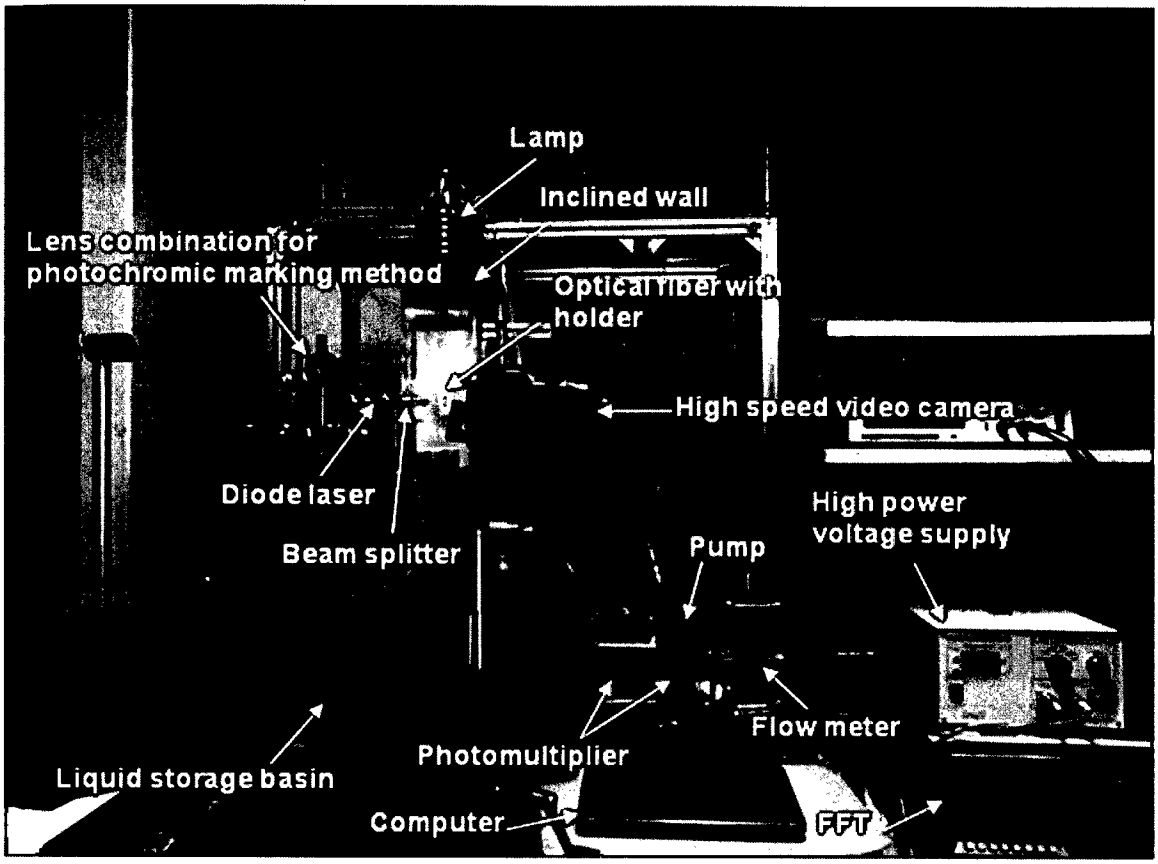

Fig. 2 Photograph of experimental apparatus setup

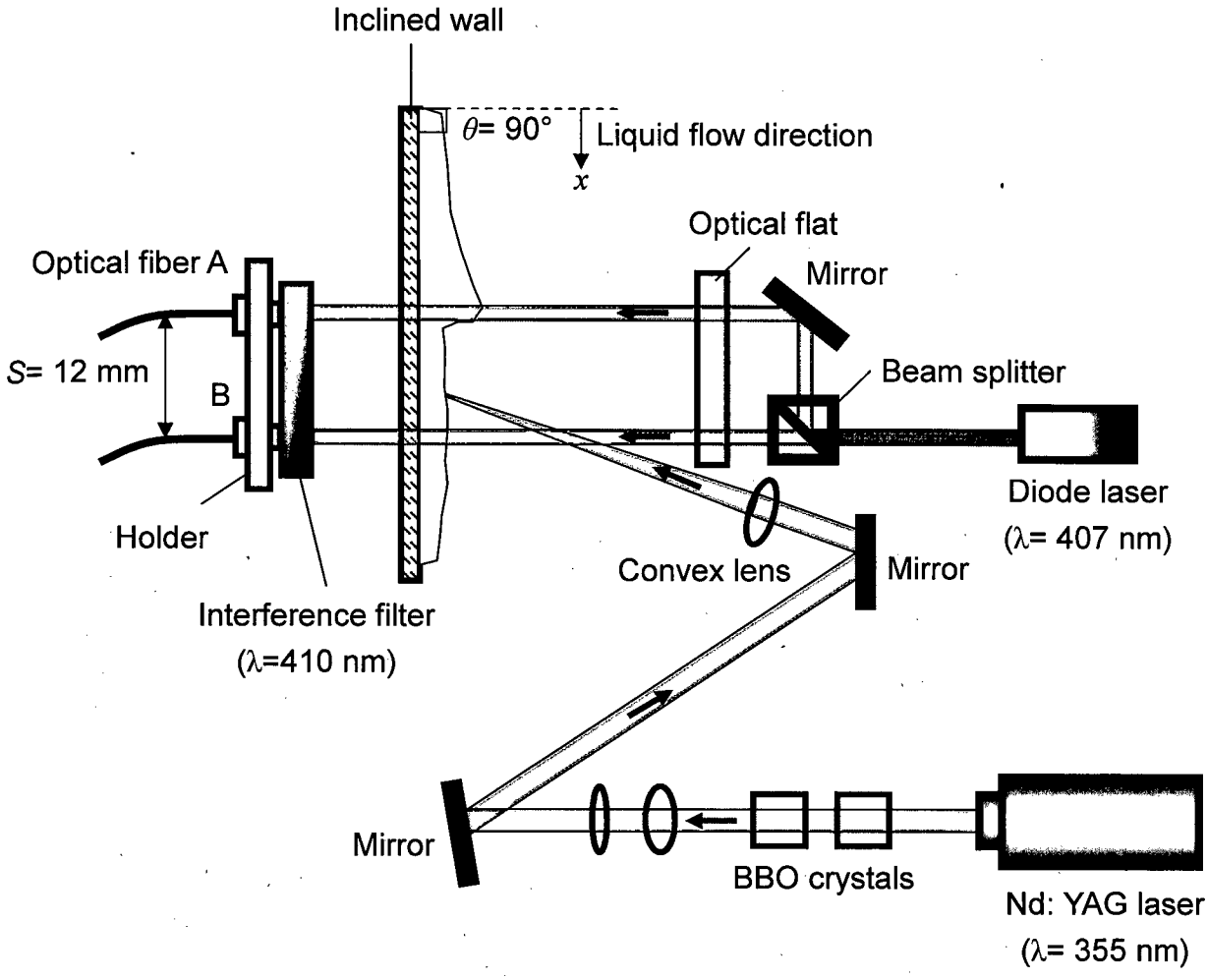

Fig. 3 Schematic view of optical arrangement

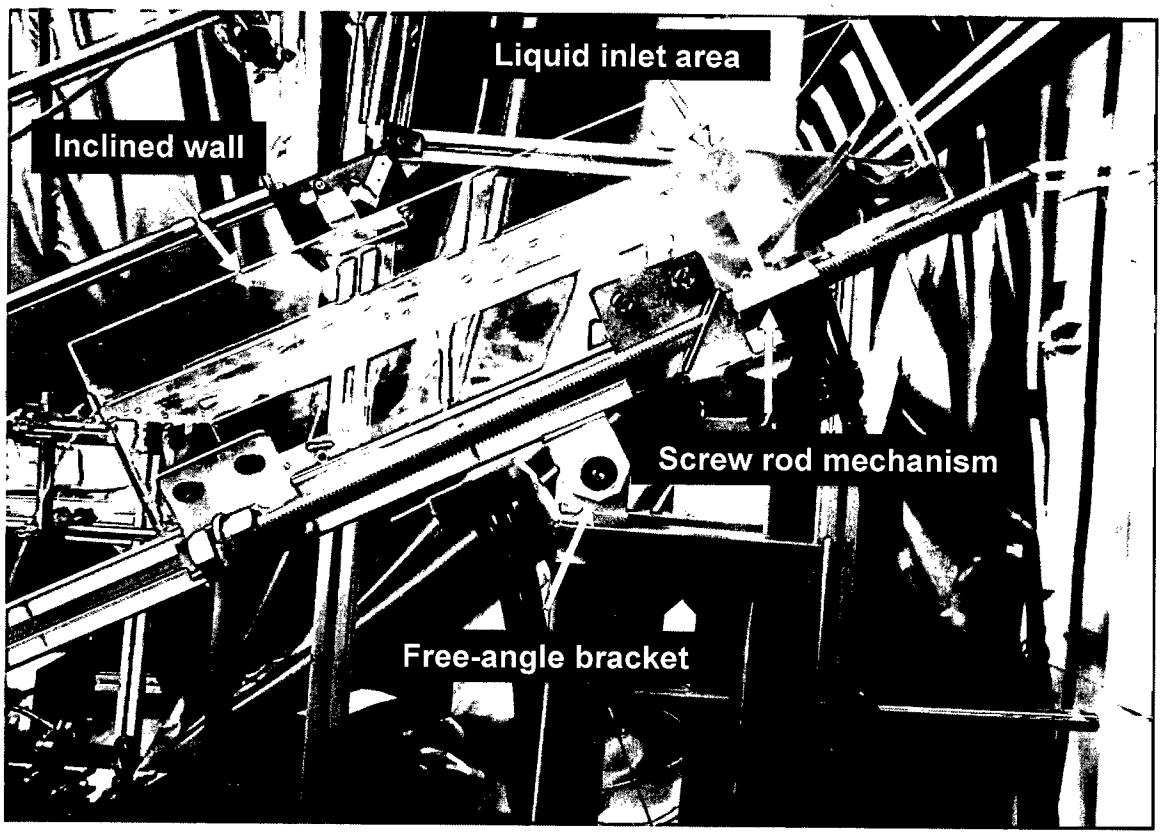

Fig. 4 Inclined wall setup

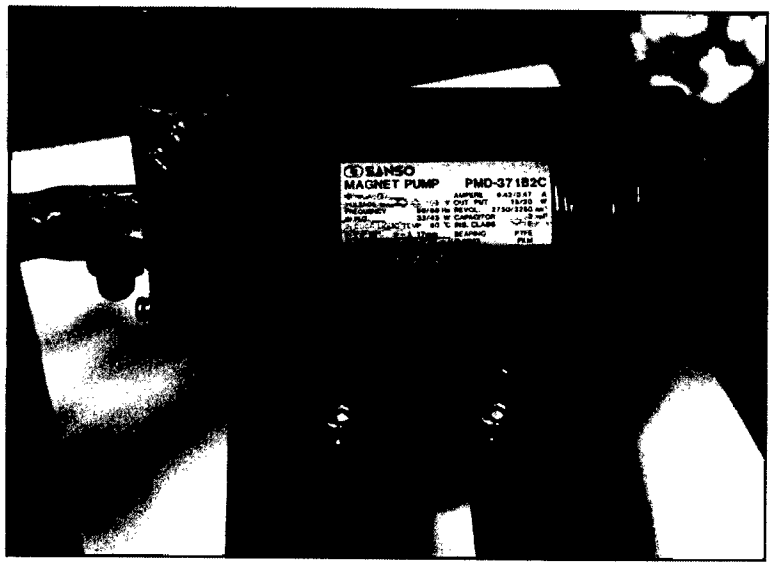

Fig. 5 Pump

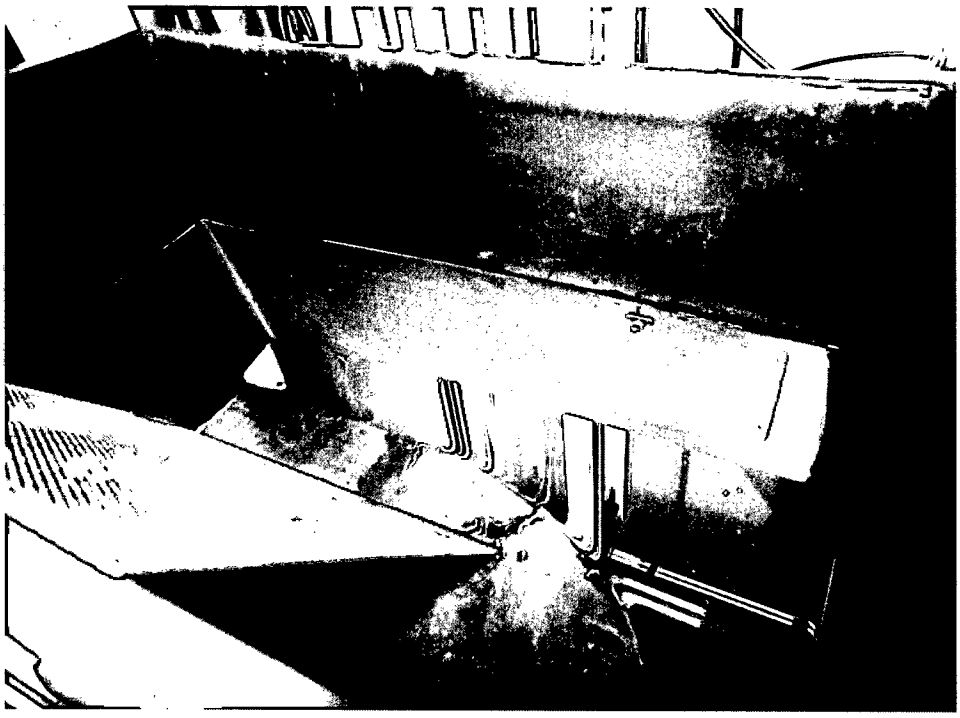

Fig. 6 Liquid storage basin with test liquid

Table 1 Kerosene oil property

Density, ρ	**Dynamic viscosity, η**	**Kinematic viscosity, ν**
800 kgm^{-3}	1.64×10^{-3} kg $m^{-1}s^{-1}$	2.05×10^{-6} m^2s^{-1}

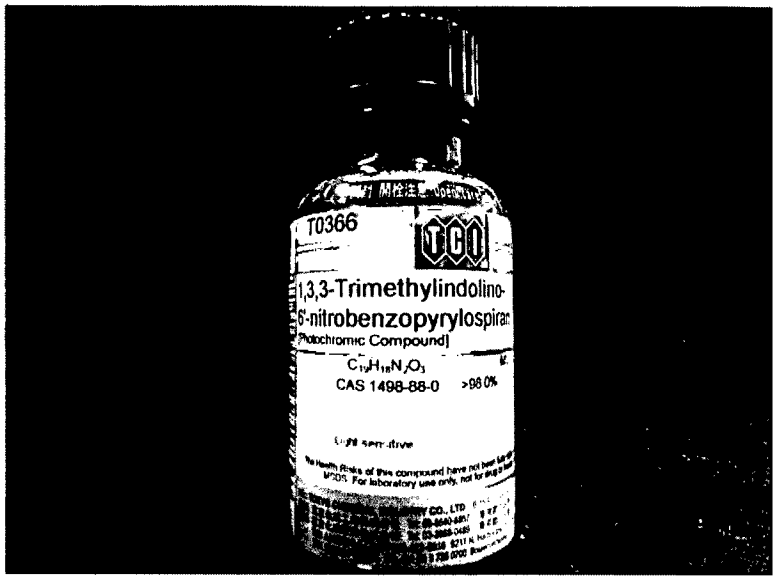

Fig. 7 Photochromic dye

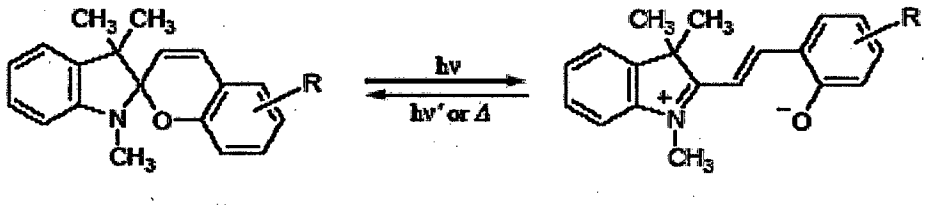

Fig. 8 Molecular configuration of photochromic dye upon UV laser irradiation