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AN OPTICAL STUDY OF INTERFACIAL TURBULENCE IN A LIQUID–LIQUID SYSTEM

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Abstract—In order to make clear the relation between the mass-transfer rate and the mechanism of interfacial turbulence in a liquid–liquid system a fundamental, qualitative study of interfacial turbulence was carried out. The mass transfer of propionic acid in the di-n-butylphthalate–water system was observed in a well devised test cell, and the concentration profile of propionic acid near the interface measured by microinterferometric and Mach–Zender interferometric methods. When these concentration profiles had a typical pattern, we defined them as the interfacial turbulence (turbulence) and observed them qualitatively.

The following results were obtained from these experiments:

(1) The existence of stable and oscillatory turbulence in the interfacial turbulence.
(2) Information about the process of how some small concentration discrepancies, after making the interface, develop to the interfacial turbulence.
(3) Information about the process how interfacial turbulence changes with time.

Fluid flow around the interfacial turbulence was investigated by following the movement of small dispersed particles in the system and the relation between the interfacial turbulence and the fluid flow is discussed.

INTRODUCTION

MASS transfer between the heterogeneous phases in the liquid–liquid system has been studied theoretically and experimentally. Recently some unusual fluid movement called interfacial turbulence was found experimentally near the interface of liquid–liquid systems and the effect of interfacial turbulence on the mass transfer evolved as a problem of interest.

The first investigators of this phenomenon were Lewis and Pratt [1]. They found some unusual fluid disturbances at the surface of a droplet when they measured the interfacial tension of liquid by the drop weight method. They concluded that this phenomenon came from the local difference of interfacial tension accompanied by local temperature difference. In the extraction of acetic acid from a solvent Sherwood and Wei [2] obtained several times larger mass-transfer coefficients than those calculated by the penetration theory and the reason for these large values was explained by the surface activity. On the other hand, Sternling and Scriven [3] analysed the mechanism of interfacial turbulence by introducing a two-dimensional model based on hydrodynamic and diffusion theory. Afterwards, Orell and Westwater [4] observed the interface of two liquids by schlieren method; the light was passed normal to the interface and revealed the same convective flow as in the Sternling and Scriven model. Haydon and Davis [5, 6] consider that oscillation of pendant drops in a liquid–liquid system comes from the increase of surface pressure arising from the local concentration difference and the values of surface pressure calculated theoretically are in agreement with experimental results.

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Recently Bakker *et al.* [7] and Ellis *et al.* [8] measured the size and depth of cellar convection and the amplitude of rippling originating from interfacial turbulence. Thomas *et al.* [9] found the generating condition of interfacial turbulence and the time necessary for the formation of cellular convection. Ruckenstein [10] made the same flow pattern as the interfacial turbulence by forced convection in the cell, simulated the phenomenon of interfacial turbulence and found that the thickness and length of the roll cell are almost the same value as those of the actual turbulence. He mentioned that this method is very useful for the analysis of wave motion and turbulence motion at the interface.

As mentioned above, studies of interfacial turbulence have recently begun to detail the phenomena, but these phenomena are so complicated that many kinds of studies are necessary to get the quantitative relation between the mass-transfer rate and the intensity of interfacial turbulence. In this study, the phenomena of interfacial turbulence are investigated by observing the concentration profile near the interface of two liquids in a well devised cell by an optical method and much useful knowledge of interfacial turbulence is obtained.

EXPERIMENTAL APPARATUS AND PROCEDURE

In this study microinterferometric and Mach–Zender interferometric methods were used.

(1) *Microinterferometric method*

A schematic diagram of this apparatus is shown in Fig. 1. It consists of an optical system and the photographic equipment. The photographic equipment consists of a microscope and camera for taking microscope pictures of 8 mm movies. Lenses with small aberration are used for the object and the eye lenses in the microscope. The object and the eye lenses are of two and seven magnifications respectively. The optical system consists of super high pressure mercury lamp as the light source, an interference filter and a test cell. The two semi-transparent mirrors of the test cell, with 60–70 per cent fractional transparency, are fixed to the frame in such a way that the plating surfaces are face to face with each other and the wedge-shaped space between the two semi-transparent mirrors can be adjusted to the experimental condition.

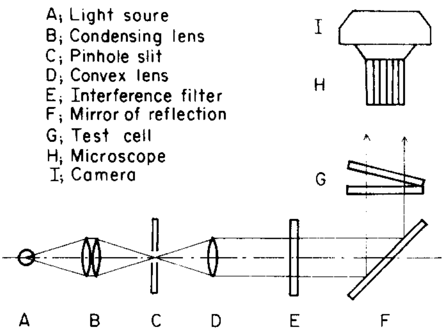
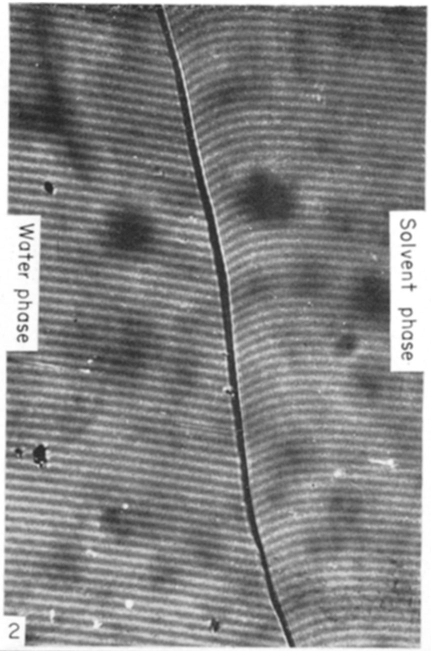


FIG. 1. Schematic diagram of microinterferometric method.

In order to observe the interfacial turbulence, the test cell is adjusted to make Fizeau fringes at suitable intervals before injecting the sample liquid. An interface between two liquids is made by carefully injecting two different liquids into the cell from two different locations.

When mass transfer occurs in the system, the concentration profile near the interface is changed and the fringes are deviated by the change of refraction ratio accompanied by concentration change. In the case of an inactive interface, mass-transfer rate is determined by the molecular diffusion, and concentration of the solute is only a function of distance from the interface. Then the deviation of Fizeau fringes is constant as shown in Fig. 2. In the active interface, the deviation of Fizeau fringes near the interface is very different from those near the inactive interface, as shown in Fig. 3. These phenomena deal with interfacial turbulence in this experiment. The dotted lines in Fig. 3 show the curve plotting the starting point of the Fizeau fringes deviation and we define them as bulk equal concentration curves.



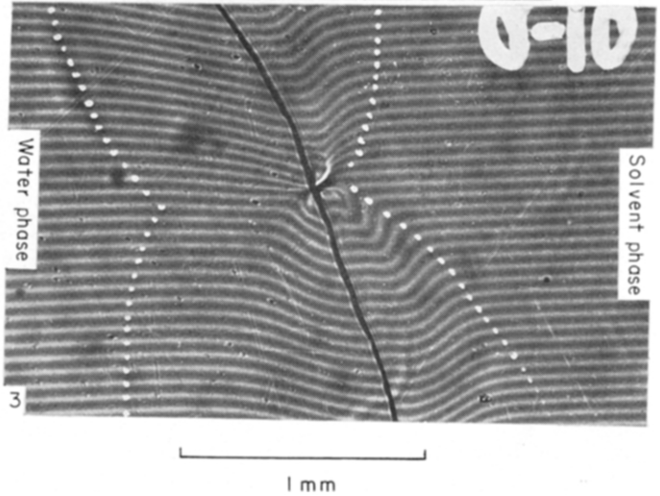
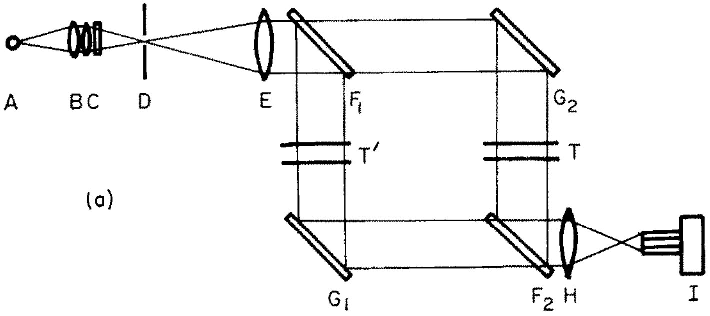


FIG. 2. Fizeau fringes near inactive interface observed by microinterferometric method.

FIG. 3. Interfacial turbulence and bulk equal concentration curve observed by microinterferometric method.



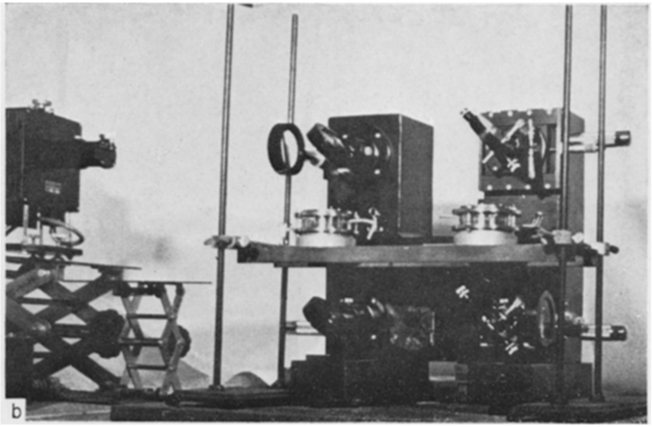
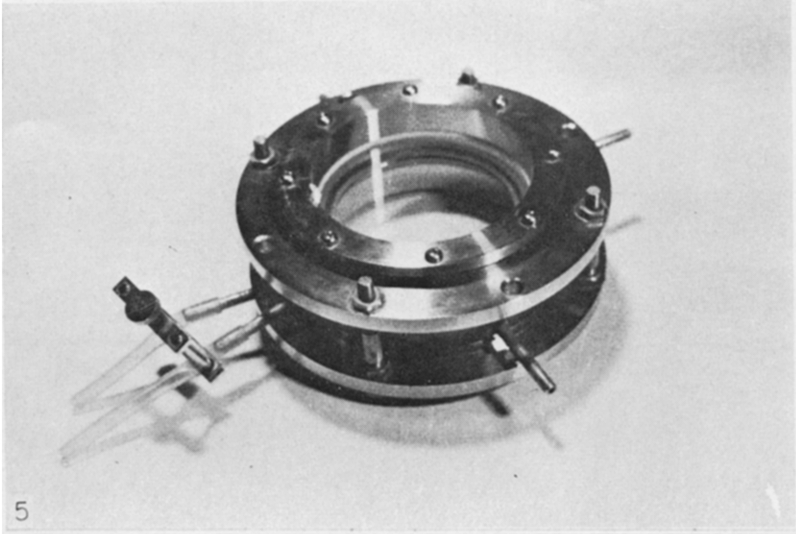


FIG. 4a. Schematic diagram of the Mach–Zender interferometric method.

FIG. 4b. The main part of the Mach–Zender interferometric method.



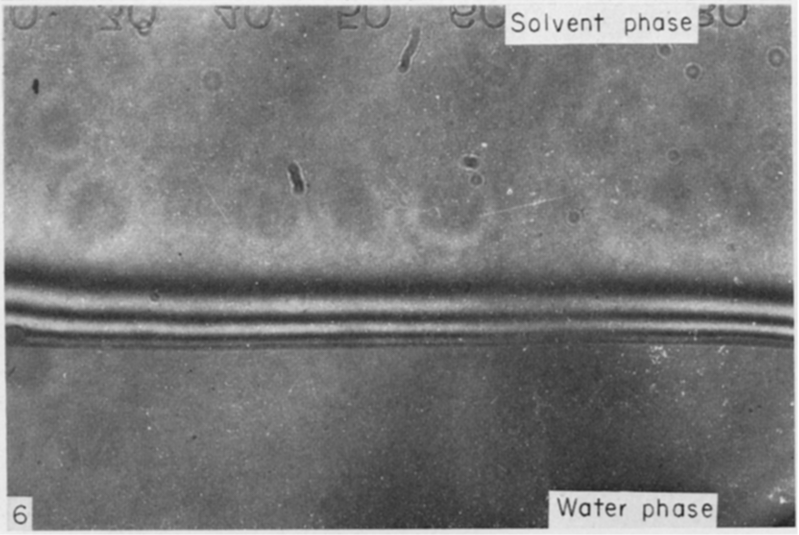


FIG. 5. Test cell used in the Mach–Zender interferometric method.
FIG. 6. Interface fringes near inactive interface observed by the Mach–Zender method.

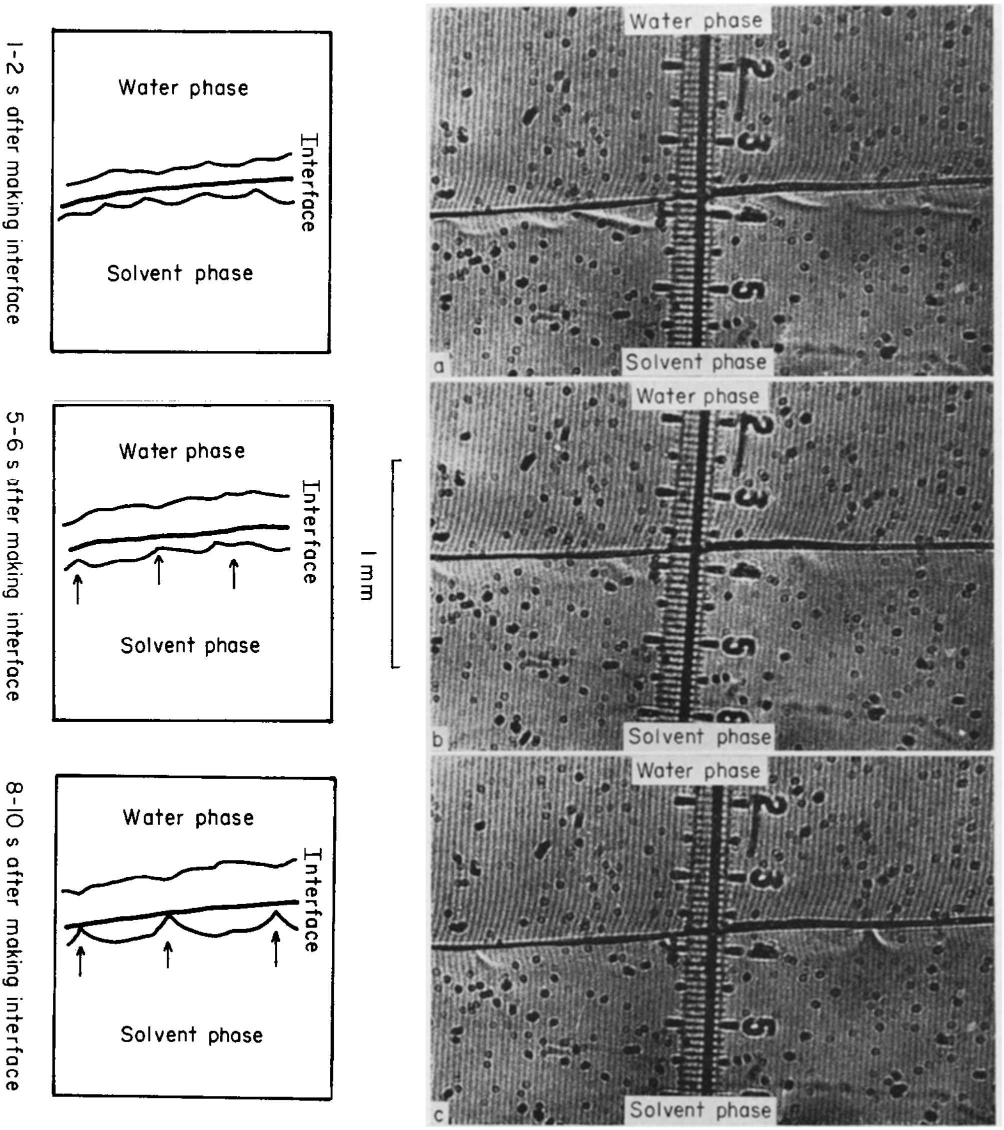


FIG. 8. Generation process of interfacial turbulence observed by the microinterferometric method.

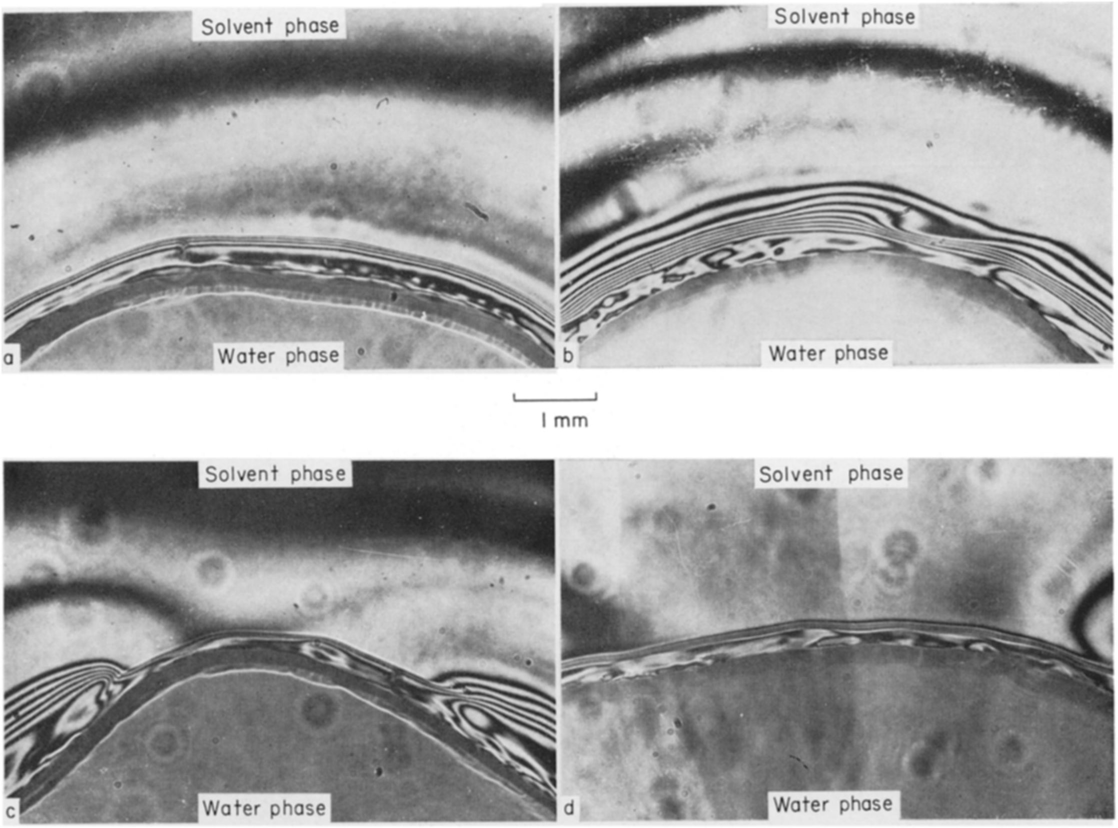
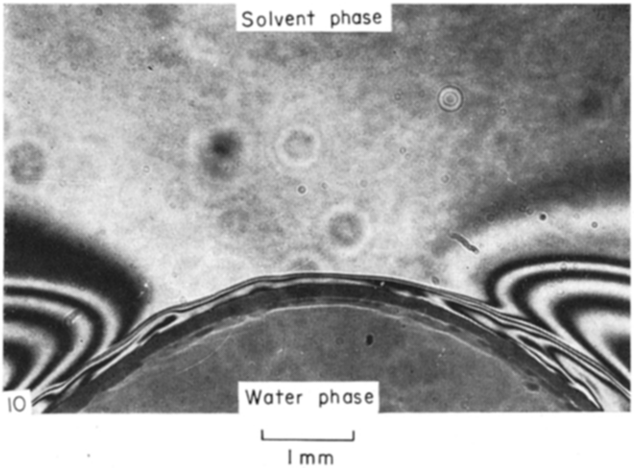
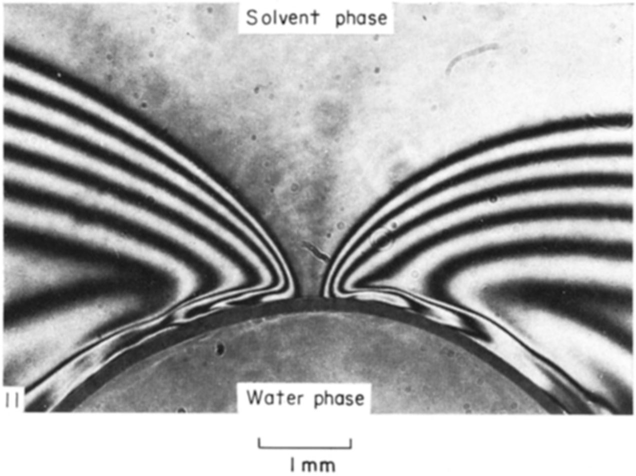


FIG. 9. Generation process of interfacial turbulence observed by the Mach–Zender interferometric method.





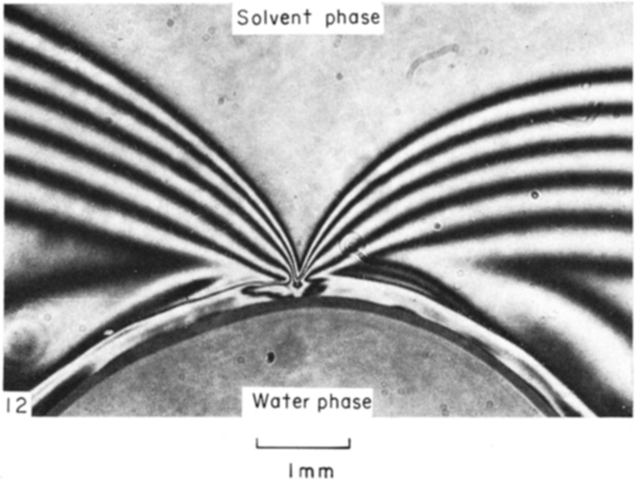
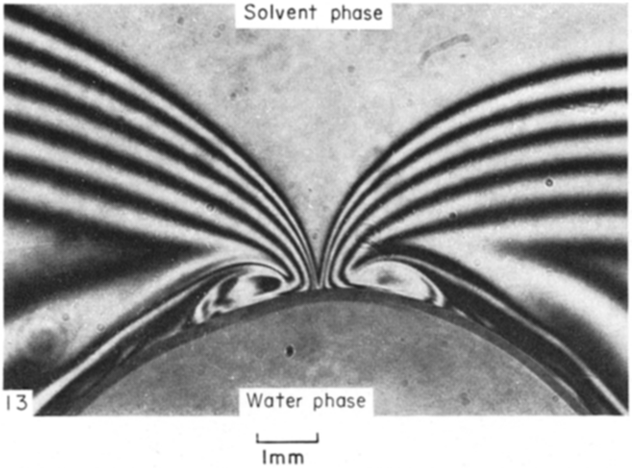
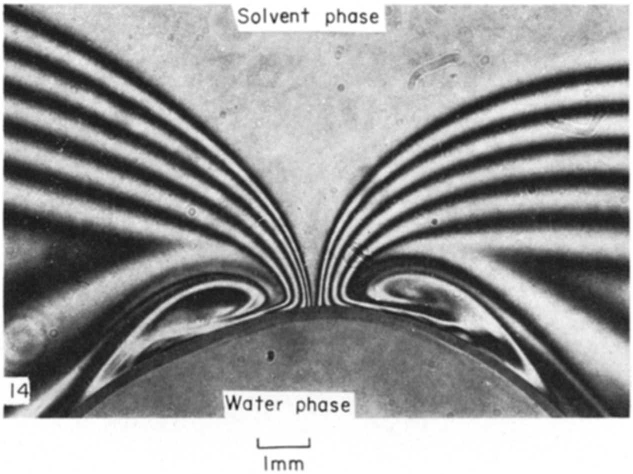


FIG. 10. Interfacial turbulence in the solvent phase 10 min after making the interface.

FIG. 11. Interfacial turbulence in the solvent phase 15 min after making the interface.

FIG. 12. Figures 12 and 13 show interfacial turbulence (oscillatory turbulence) in the solvent phase 17 min after making the interface. In Fig. 12 the interface turbulence is stopped.





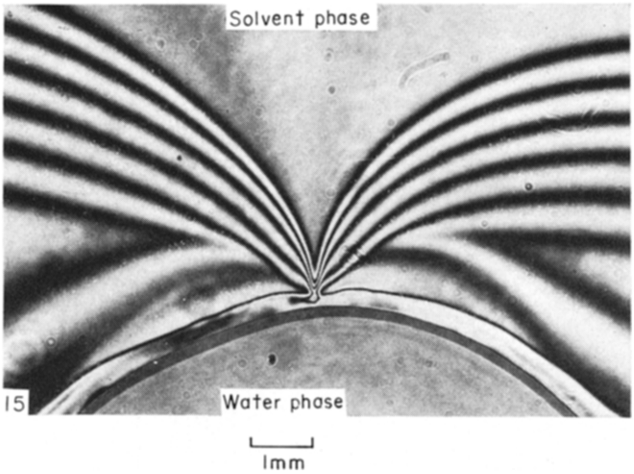
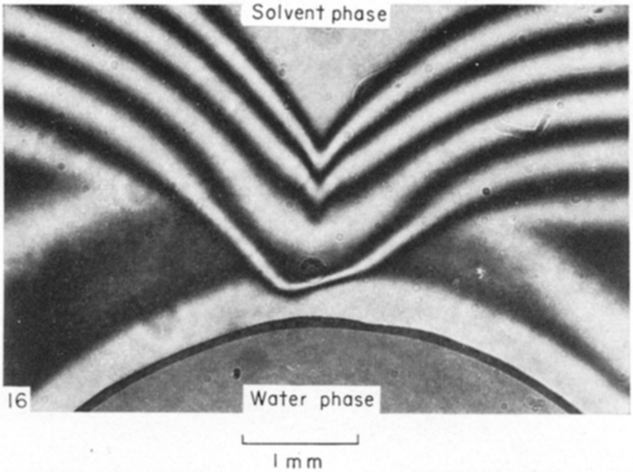
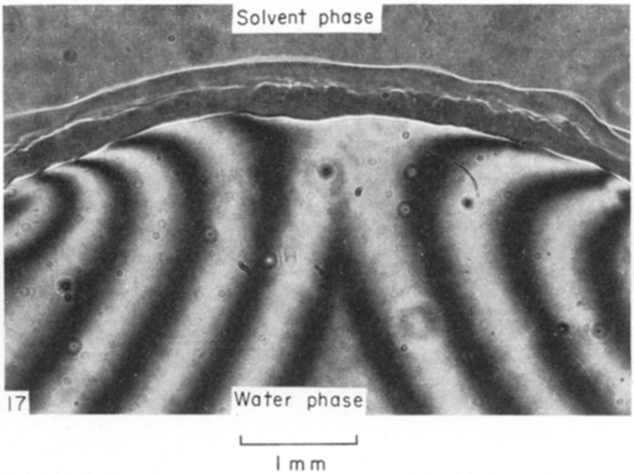


FIG. 13. The interfacial turbulence is generated.

FIG. 14. Interfacial turbulence in the solvent phase 22 min after making the interface.

FIG. 15. Interfacial turbulence in the solvent phase 25 min after making the interface.





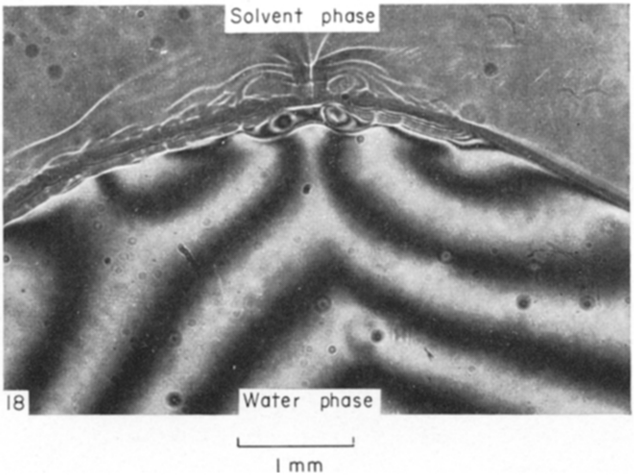
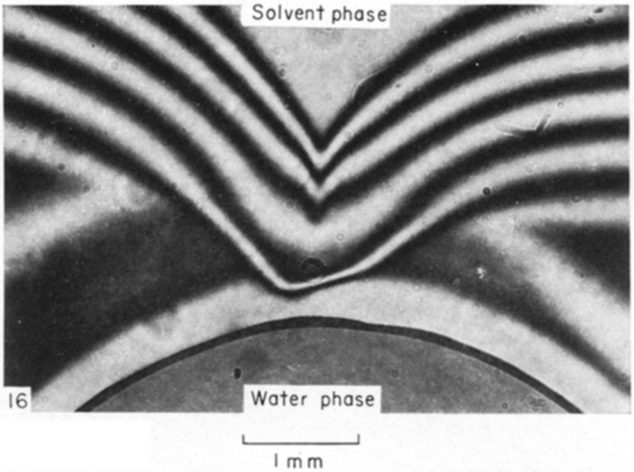
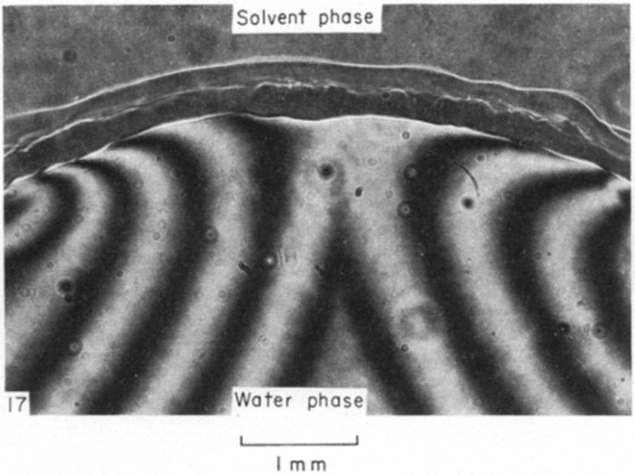


FIG. 16. Interfacial turbulence in the solvent phase 27 min after making the interface.

FIG. 17. Interfacial turbulence in the water phase 5 min after making the interface.

FIG. 18. Interfacial turbulence in the water phase 10 min after making the interface.





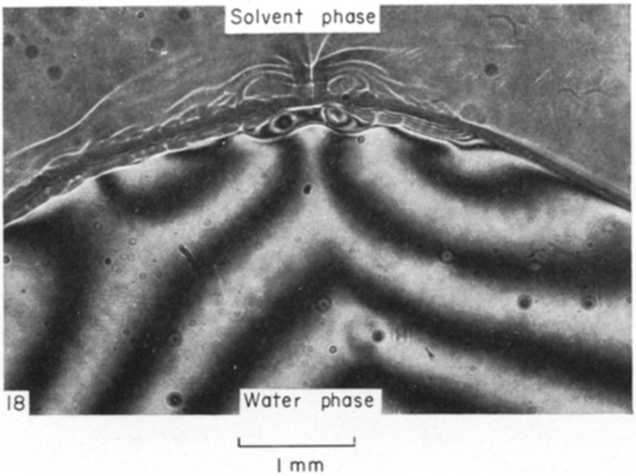


FIG. 19. Interfacial turbulence in the water phase 13 min after making the interface.

FIG. 20. The interfacial condition in the water phase similar to liquid boiling.

FIG. 21. The motion of fine particles around the turbulent point (corresponds to Fig. 14).

(2) *Mach–Zender interferometric method*

Figures 4a and 4b show schematic diagrams of the Mach–Zender interferometric method and the main part of that equipment, respectively. The photographic equipment consists of a lens with long focal length ($f = 130$ mm) and a microscope movie camera. The object and the eye lenses of the microscope are of four and ten magnifications, respectively. A movie camera, type H16RX made by Bolex Co. Ltd., and a 16 mm Sakura High Speed Negative film No. 6162 are used in this experiment. The test cell consists of two pieces of window glass with sample injecting and gas exhausting holes as shown in Fig. 5, and these glasses are carefully set in the frame. The sensitivity of the Mach–Zender interferometer is such that even a slight strain on the glass plate produces a deviation of the interference fringes. Therefore two pieces of the glass are used in this experiment after checking strain on the glass plate, flatness and parallelism of both surfaces. The two pieces of glass plate are adjusted to be parallel to each other by an auto-collimator in the construction of the cell. The distance between the plates is measured with the microscope. The cell for the compensation of the optical path length has the same construction as the test cell.

The experimental procedure is as follows: First, solvent or water are put into both the test and compensation cells, and the optical path lengths for the test and the compensation cell are adjusted to make one fringe on the film, and then, in order to produce the interface in the test cell, a part of the liquid is taken out from the gas exhausting hole and at the same time another liquid is put into the cell from the sample injecting hole. When the surface is inactive, the mass-transfer rate is determined by the molecular diffusion only, the interference fringes become parallel to the interface of the two liquids, as shown in Fig. 6, and these fringes depart from the interface as time elapses. The experiment is performed at a constant room temperature of 20 ± 1°C.

In this experiment di-n-butylphthalate–water system is used as the solvent and propionic acid dissolved in the di-n-butylphthalate transfers to the water phase. The reason for using the di-n-butylphthalate–water system is that the solubility of both liquids is very small in each other and as the difference of viscosity between them is very large, interfacial turbulence can easily be generated. In order to observe the flow pattern of interfacial turbulence near the interface, powders of tetra-phenyltin and cellulose are dispersed in the di-n-butylphthalate and water phases, respectively.

EXPERIMENTAL RESULTS

(1) *General type of interfacial turbulence*

Interfacial turbulence is a very complicated phenomenon and several different types of interfacial turbulence are found by the micro-interferometric method. Classification of these phenomena is quite hard. However, the types of interfacial turbulence can be roughly classified by the style of the bulk equal concentration curve as shown in Figs. 7a–e. Both solid and dotted lines in Fig. 7 show bulk equal concentration curves. If the solid and dotted lines are in the same figure, this means that the curve can be changeable between those two curves periodically.

Interfacial turbulence has the following different types.

Type A. The tip of the bulk equal concentration curve keeps in touch with the interface in the solvent phase as shown in Fig. 7a and looks as if it does not move. The lifetime of this type of turbulence is comparatively long.

Type B. Interfacial turbulence of type B is almost the same style as type A except that the tip of the curve moves slowly to the left or right along the interface, as shown in Fig. 7b.

Type C. The tip of the bulk equal concentration curve in the solvent phase periodically touches and detaches from the interface, as shown in Fig. 7c.

Type D. Bulk equal concentration curve has the same style as type C except that the tip of the curve departs considerably from the inter-

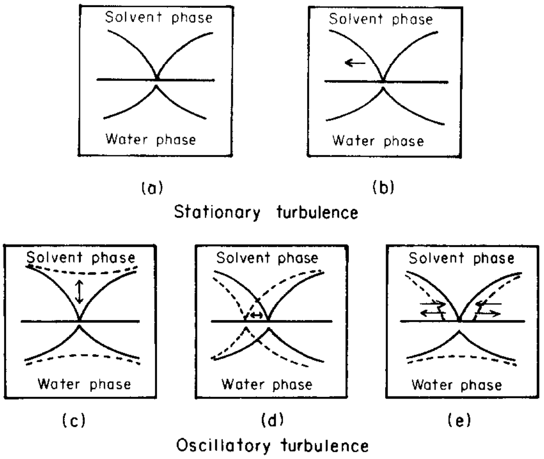


FIG. 7. The type of interfacial turbulence.

face and moves along the interface as shown in Fig. 7d.

Type E. The bulk equal concentration curve in solvent phase is divided into two curves that get together periodically with explosion accompanied by waving, as shown in Fig. 7e. Such a turbulence happens only with very high solute concentration in the solvent phase.

Though there are many types of interfacial turbulence as shown above, they can be roughly classified into two types; Stationary turbulence as shown Figs. 7a and b and oscillatory turbulence as shown Figs. 7c–e.*

(2) *Generation mechanism of interfacial turbulence*

After producing the interface the generation of interfacial turbulence is observed by the transformation of bulk equal concentration curve by taking photographs at the rate of two per second. The results are shown in Figs. 8a–c. Arrow marks in Fig. 8b show the position of a big concentration disturbance. Arrow marks in Fig. 8c show the position of a well-developed interfacial turbulence. Figures 9a–d are photographs of a concentration profile in the solvent side using the Mach–Zender interferometric method.

According to these observations, there are many small concentration disturbances at several points near the interface as shown in Fig. 9a just after the interface is produced, but there are interference fringes parallel to the interface a little way from it, as shown in Fig. 9a. This means that the size of the concentration disturbances just after making the interface is small and their effect is confined to near the interface. But these small concentration disturbances interfere with each other and grow to large concentration disturbances as in Fig. 8b. Figures 9b and c are photographs taken by Mach–Zender interferometric method in that situation. A part of the interface contacts with bulk concentration at a very small distance as shown in Fig. 9c and eventually that distance becomes 4–5 mm and small concentration disturbances develop perfectly to the interfacial turbulence as shown in Figs. 8c and 9d.

(3) *Relation between the concentration of solute and generation of interfacial turbulence*

The generation of interfacial turbulence was observed in the field of the microscope by a microinterferometric method under the various concentrations of solute in the solvent phase. Table 1 shows these results. In low concentration of solute, (for example, in less than 7·66 wt %,) frequency of turbulence generation is so small that we can only find turbulence by moving the test cell. But when the solute concentration is increased, the frequency of turbulence generation increases and the intensity of turbulence becomes powerful. In some extreme cases the turbulence is so strong that an emulsion is generated in the water phase near the interface and the interface itself is broken and pushed into the other phase.

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*The names stationary and oscillatory turbulence have already been used in the theoretical study of Sternling and Scriven. But it is not clear in their study which phenomenon corresponds to stationary or oscillatory turbulence. The names of stationary and oscillatory turbulence defined by Sternling and Scriven are used only to explain our experimental results. So we are not sure whether our concept of those names agrees with their concept or not.

These phenomena happen in the small test cell with less than 0·1 mm thickness and as the effect of wall and curvature of interface on the turbulence can not be ignored, these results can not be applied to the general case quantitatively. These results supply a useful suggestion on the fundamental study of interfacial turbulence.

Table 1. Relation between the interfacial turbulence and solute concentration

Solute concentration (wt.%)	Observation results
1·35	Interfacial turbulence is not observed
3·23	The same as the above
4·88	Many small concentration disturbances are observed near the interface in the solvent phase, but these disturbances do not usually develop into interfacial turbulence
7·66	Two or three concentration disturbances are generated near the interface in the field of camera vision just after making the interface. After a few seconds, those disturbances sometimes develop into interfacial turbulence
9·15	Number of concentration disturbances is the same as that of 7·66 wt.% solute concentration. These disturbances develop to the interfacial turbulence and this turbulence sometimes keeps up for more than 10 min
19·2	Three or five concentration disturbances are generated in the field of camera vision just after making the interface, but after a few seconds, one or two of these disturbances develop to interfacial turbulence and after 10 min only one interfacial turbulence remains in vision
24·9	The number of concentration disturbances is the same as that of 19·2 wt.% solute concentration. About 20 min after making the interface, an emulsion is generated in the water phase. A phenomenon like waving is observed near the turbulent point* in the solvent phase
36·0	The phenomenon like waving is observed in both solvent and water phases. Emulsion is generated around turbulent point in the water phase. Interface is sometimes pushed away to the water phase by the fluid flow to the turbulent point from the bulk in the solvent phase
68·5	Frequency of turbulence generation is increased more and intensity of turbulence hardens. Interface near the turbulent point is more often pushed away to the water phase. Emulsion in the water phase increases and some of the emulsion is moved along the interface

* Turbulent point is the point where the tip of the bulk equal concentration curve is in contact with the interface.

(4) *Change of interfacial turbulence with time*

After establishing interfacial turbulence, change of interfacial turbulence is observed by the Mach–Zender interferometric method at 10 wt% solute concentration. The difference of refraction ratio between the solvent phase and water phase is so large that it is impossible to find fringes for both solvent and water phases at the same time. Accordingly the change of interfacial turbulence is observed by taking movie photographs of solvent and water phases, respectively.

(a) *Concentration profile of interfacial turbulence in the solvent phase* 10 *min after making interface.* As already mentioned in the previous section, well-developed interfacial turbulence, with an interface of 4–5 mm in length, contacting with bulk concentration at a very small distance (as shown in Fig. 10), begins to shrink gradually.

15 *min after making interface.* The length of interface contacting with bulk concentration at a very small distance narrows as shown in Fig. 11 and the bulk equal concentration curve becomes like a nozzle with only the tip of this curve keeping in contact with the interface. (This situation is called stationary turbulence in the previous section.) In this situation, there is convective flow inside two bulk equal concentration curves to the interface from the surroundings and after reaching the interface, this fluid is separated to left and right at the turbulent point and flows along the interface. (Fluid flow near the active interface will be mentioned in more detail in next section.)

17 *min after making interface.* The tip of the nozzle style bulk equal concentration curve begins to move slightly and this movement becomes bigger and bigger in proportion to the time elapsed until at last the tip of the nozzle style curve departs from the interface as shown

in Fig. 12. (In this condition, fluid flow near the interface stops.) After a while, the tip of the nozzle style curve begins to approach the interface. When the tip of this curve attains the interface, the bulk equal concentration curve stays as shown in Fig. 13. Afterwards these departures from and approaches to the interface are repeated periodically. These departing or approaching rates slow as time elapses and the periodic time of this repeating becomes longer and longer. (This situation corresponds to the oscillatory turbulence already mentioned.)

22 *min after making interface.* Periodic time between the generating and stopping of interfacial turbulence becomes much more longer. When interfacial turbulence is generated, the bulk liquid itself contacts directly with the interface as shown in Fig. 14.

25 *min after making interface.* The periodic repeating of the turbulent generation slows and the tip of the nozzle style bulk equal concentration curve oscillates somewhere near the interface as shown in Fig. 15. The intensity of turbulence is too weak to produce a large periodic movement and the interfacial turbulence gradually becomes inactive.

27 *min after making interface.* The interface reaches an inactive state and the bulk equal concentration curve gradually departs from the interface as shown in Fig. 16.

(b) *Concentration profile of interfacial turbulence in the water phase* 5 *min after making interface.* Usually, generation of the interfacial turbulence can be recognized by the tip of the bulk equal concentration curve approaching the interface. The tip of this curve still does not attain the interface as shown in Fig. 17.

10 *min after making interface.* As the intensity of turbulence is too weak to maintain the interfacial turbulence continuously, the periodic repeating of turbulent generation and interruption is observed as shown in Fig. 18.

13 *min after making interface.* The intensity of turbulence becomes much weaker and only a slight disturbance of liquid near the interface is recognized (Fig. 19).

These experimental results on interfacial turbulence are one example of many experiments. Generally speaking, concentration profile near active interface is affected by the method of interface formation and curvature of interface. Therefore, quantitative correspondence between the turbulence behaviour and the time elapsed after making interface is still not clear.

Intensity of interfacial turbulence becomes stronger as the concentration of solute is increased. Such a violent phenomenon as the liquid boiling is observed when the interface is in contact with the bulk equal concentration curve at a very small distance (Fig. 20). When the solute concentration is increased, lifetimes of stationary and oscillatory turbulences are increased, but the process from generation to disappearance is independent of the solute concentration.

(5) *Liquid flow near the active interface*

The existence of the liquid flow is supposed from the concentration profile around the turbulent point and from the phenomenon like a rippling at the active interface. In order to show this liquid flow experimentally, fine particles of tetra-phenyltin and cellulose are dispersed in the solvent and water phases, respectively, and the movement of those fine particles is observed. Figure 21 shows one of those experimental results (corresponding to Fig. 14). Even if the movement of fine particles in the bulk of solvent phase is not clear, fine particles near the interface move from side to side along the interface.

In the case of turbulence accompanied by a long interface contacted with the bulk concentration at a very small distance, convective flow of particles near the interface is not clear, but the fine particles near that area have complicated movement based upon the fluid movement like a boiling of liquid at the interface. On the other hand, in the case of stationary turbulence with tip of the nozzle style bulk equal concentration curve contacting with interface, fine particles move to the turbulent point from the bulk and after reaching the turbulent point, these particles are separated from side to side and most of them move along the interface while some circulate near the turbulent point. The velocity of those fine particles is the highest at the turbulent point.

In the case of oscillatory turbulence with periodic repeating of generation and interruption, when the turbulence is generated, fine particles near the active interface move like particles in the stationary turbulence, but even if the turbulence is interrupted, fine particles sometimes have a slight movement along the interface.

From these experimental results, typical fluid flow near the active interface in the solvent phase exists only in the case of the tip of the bulk equal concentration curve directly in contact with the interface (Fig. 14) and there is no typical fluid flow around the interface in the other types of turbulence.

Convective fluid flow in the water phase is observed by movement of the fine particles dispersed and by displacement of the bulk equal concentration curve. But the intensity of convective fluid flow in the water phase is weaker than the flow in the solvent phase. Therefore symmetrical fluid flow to the interface introduced in the Sternling and Scriven model is not recognized in this experiment.

DISCUSSION

Discussion about the experimental method

Experimental method of this study has three typical characteristics as follows:

1. Quantitative measurement of solute concentration is possible near the interface.
2. The solute transfers horizontally from the solvent phase to the water phase in a very small space (less than 0·1mm in micro-interferometric method and 0·8mm in Mach–Zender interferometric method) of the test cell set horizontally.
3. Observation of concentration profile near the active interface is possible just after producing an interface.

Compared with other methods, our experimental methods have the following concrete characteristics. Schlieren method is one of the suitable methods for the qualitative observation of solute concentration near the interface, but it is quite difficult to obtain the quantitative concentration profile. The quantitative observation of concentration profile near the interface is possible by either methods of this study and sensitivity of this measurement is 10–100 times larger than that of the schlieren method. As concentration changes are expressed by the displacement of Fizeau fringes in the micro-interferometric method, it takes time to obtain the local concentration profile by this method, but this method can observe local concentration change of both solvent and water phases at the same time. On the other hand, as interference fringes of the Mach–Zender interferometric method express equal concentration curves, the observation of concentration profiles near the interface is quite easy, but when the difference of refraction ratio between the two phases is large, interference fringes of either water phase or solvent phase appear in the picture. Both phases cannot be observed at the same time.

In this experiment, as the interface is made vertical to the window glass in the very small space of the test cell set horizontally, capillary action, curvature of the interface and wall effects influence the interfacial turbulence, concentration disturbances near the interface interfere with each other and only strong interfacial turbulence can be generated. Of course, since natural convection based on the density change accompanied by mass transfer can be taken away, complicated turbulent phenomena are simplified extremely. Therefore fluid flow and solute concentration profiles near the turbulent point can be clearly observed and the build up of interfacial turbulence can be investigated using these experimental methods together. This experiment is performed under special conditions but is one of the most interesting ways to pursue the principle of interfacial turbulence.

Change of interfacial turbulence with time

Equal concentration curves are parallel to the interface just after the interface is produced. After a few seconds, many small local concentration disturbances occur near the interface and most of those disturbances disappear as time elapses, but some develop to interfacial turbulence. From these facts, solute in the solvent phase is transferred by molecular diffusion just after making the interface and some small local concentration disturbances, coming from the instability of the interface, produce variation of local interfacial tension and develop to strong interfacial turbulence in cooperation with supplying the solute.

The well-developed turbulence produces an interface of 4–5 mm in length contacting with bulk concentration at a very small distance (Fig. 10). Figure 22 shows the interfacial tension model near the interface estimated by the momentum transfer theory. As the concentration of solute at the turbulent point "A" in the solvent side is higher than at point "B" in Fig. 22, the force acts from point "A" to point "B" and the reverse direction force acts on the water side. When the force acting on the solvent side is stronger than that of the water side, the interface contacting with bulk concentration at a very small distance is enlarged by this force balance and reaches a maximum in the equilibrium state. After a while, the region of interface contacting with bulk concentration at a very small distance is decreased. This decrease is due to the relatively increased force in the direction from "B" to "A" on the water side by the increase of solute concentration at that point (Fig. 22). The reason why the tip of the nozzle style bulk equal concentration curve contacts with or departs from the interface periodically is explained as a periodic change of the force balance accompanied by a periodic change of solute concentration at the points "A" and "B" (Fig. 22). Finally concentration of solute at points "A" and "B" is decreased by the mass transfer across the interface and the acting force on the points "A" and "B" weakens until the interfacial turbulence finally disappears. A fundamental factor for the generation of the interfacial turbulence is attributed to the local concentration disturbance in the liquid–liquid system.

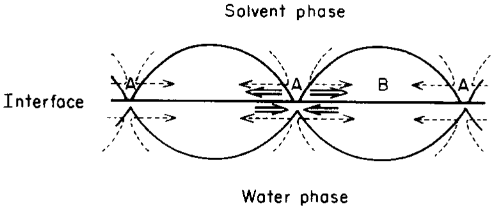

FIG. 22. Interfacial fluid model based on the momentum transfer theory.

Fluid flow near the interface

It is very interesting that fluid flow observed near the interface is, in some parts, quite analogous to the fluid model in interfacial turbulence introduced by Sternling and Scriven. But the fluid flow from the interface to bulk as mentioned in the Sternling and Scriven model is not observed in this study.

Circulation flow near the turbulent point is also observed and this flow sometimes occurs in the symmetrical position to the turbulent point in the solvent phase and sometimes only on one side in the solvent phase near the turbulent point. In the former case, the turbulent point remains in the same position, but in the latter case, the turbulent point moves along the interface in the direction where there is no circulation flow. This movement is due to the decrease of the interfacial tension accompanied by a local increase of solute concentration promoted by the circulation of fluid at that point. Though there is the circulation flow in the stationary turbulence, no fluid suck exist. Therefore, fluid elements along the interface may gradually return to the bulk after departing from the turbulent point. If this assumption is correct, contact time of the fluid element in the solvent phase with the water phase is not equal to the interfacial effective length divided by average fluid velocity, but becomes the distribution function of the residence time of the individual fluid element in contact with the interface. To establish the fluid flow around the turbulent point becomes important for the analysis of mass-transfer mechanisms accompanied by interfacial turbulence.

The reason why the tip of the bulk equal concentration curve in the water phase departs from the interface, even if the tip of that curve in the solvent phase is in contact with the interface, is that fluid flow in the solvent phase is stronger than that in the water phase. These differences of fluid flow between two phases are considered to depend on the difference of physical properties between the two phases.

Thus, fluid flow around the turbulent point is not as symmetrical as the "roll cell model" of Sternling and Scriven and some modifications may be necessary. In order to establish the fluid flow near the interface more exactly, further experiments are necessary.

CONCLUSION

The summary of this study is as follows:

1. Interfacial turbulence is classified into stationary turbulence and oscillatory turbulence.
2. Generation of interfacial turbulence initially comes from local small concentration disturbances. Most of those disturbances disappear by interference with each other, but some develop to the interfacial turbulence in cooperation with the supply of solute from the bulk of the solvent phase.
3. Each interfacial turbulence has the following processes; generation of turbulence, stationary turbulence, oscillatory turbulence and and decease of turbulence.
4. Increase of solute concentration not only increases the frequency of turbulence generation, but also increases the life time of turbulence.
5. There is typical convective flow near the interface in stationary turbulence and the velocity of this flow reaches a maximum at the turbulent point. But no flow suck as mentioned in the Sternling and Scriven model is observed in this experiment.
6. In oscillatory turbulence, the generation and interruption of turbulence are repeated periodically. When turbulence occurs the same type of flow as in stationary turbulence is observed near the interface, but when the turbulence decreases, only small concentration discrepancies are observed near the interface.
7. The change of interfacial turbulence with time can be explained by an interfacial fluid model based on the momentum transfer across the interface.
8. In stationary turbulence with convective fluid flow around the interface, contact time of fluid element in the solvent phase with the other phase may be determined by the distribution function of the resident time of the individual fluid element.

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UNE ÉTUDE OPTIQUE DE LA TURBULENCE INTERFACIALE DANS LE SYSTÈME LIQUIDE-LIQUIDE

Résumé—La turbulence interfaciale genérée près de l'interface dans un système liquide–liquide est un des problèmes les plus intéressants. Mais ce phénomène est si compliqué que l'on doit dégager beaucoup de facteurs inconnus.

Afin de dégager la relation entre le flux de transfert massique et le mécanisme de turbulence interfaciale, l'étude fondamentale de la turbulence interfaciale est conduite qualitativement. Dans cette expérience, on observe le transfert massique d'acide propionique dans le système di-n-butyl-phthalate/eau dans une cellule d'essai bien conçue et le profil de concentration d'acide propionique près de l'interface est observé par une méthode micro-interférométrique et par la méthode interférométrique Mach–Zender. Quand ce profil de concentration a une configuration typique, on définit ce phénomène comme turbulence interfaciale et on l'observe qualitativement.

Les résultats suivants sont obtenus à partir de ces expériences:

(1) L'existence d'une turbulence stable et oscillatoire dans la turbulence interfaciale.

(2) Le processus suivant lequel quelques petits écarts de concentration apparaissent après que l'interface ait atteint la turbulence interfaciale.

(3) Le processus par lequel la turbulence interfaciale change avec le temps. Les mouvements des fluides autour de la turbulence interfaciale sont étudiés en suivant le mouvement de petites particules dispersées dans le système et la relation entre la turbulence interfaciale et l'écoulement du fluide est discutée.

EINE OPTISCHE UNTERSUCHUNG DER TRENNSCHICHT-TURBULENZ IM SYSTEM FLÜSSIG-FLÜSSIG.

Zusammenfassung—Die Trennschicht-Turbulenz, die sich in der Nähe der Trennfläche eines Systems flüssig-flüssig bildet, ist so kompliziert, dass viele Faktoren noch unbekannt sind.

In der Absicht, die Beziehung zwischen der Stoffübertragung und dem Mechanismus der Turbulenz der Trennfläche zu klären, wurde qualitativ eine grundlegende Studie durchgeführt. In diesem Experiment wurde die Stoffübertragung einer propionischen Säure in einem di-n-butyl-phthalat-Wasser-System in einer geeigneten Versuchskammer untersucht. Das Konzentrationsprofil der propionischen Säure in der Nähe der Trennfläche wurde mit der Mikro- und mit der Mach-Zender-Interferometermethode beobachtet. Wenn dieses Konzentrationsprofil ein typisches Muster aufwies, wurde dies als Trennflächenturbulenz bezeichnet und qualitative Beobachtungen durchgeführt.

Folgende Ergebnisse wurden ermittelt.

(1) Es existiert eine stabile und oszillierende Turbulenz in der Trennschicht.

(2) Der Einfluss der Konzentrationsstörungen zur Erzielung von Trennschichtturbulenzen wurde untersucht.

(3) Der zeitliche Einfluss auf die Änderung der Trennschichtturbulenz wurde fes-gestellt.

Die Flüssigkeitsströmung in der Trennschicht wurde durch Schwebeteilchen sichtbar gemacht und die Verbindungen zwischen der Trennschichtturbulenz und der Strömung wurden diskutiert.

ОПТИЧЕСКОЕ ИССЛЕДОВАНИЕ ТУРБУЛЕНТНОСТИ НА ПОВЕРХНОСТИ РАЗДЕЛА В СИСТЕМЕ ЖИДКОСТЬ-ЖИДКОСТЬ

Аннотация—Межфазная турбулентность, возникающая вблизи поверхности раздела системы жидкость-жидкость, являегся одной из интересных проблем. Однако это явление очень сложное, т.к. требует учёта многих неизвестных факторов.

Для того, чтобы выяснить связь между скоростью массообмена и механизмом турбулентности на поверхности раздела, проведено фундаментальное качественное исследование турбулентности на поверхности раздела. В этом исследовании проводилисъ наблюдения массопереноса пропановой кислоты в дибутилфталат—вода на экспериментальной ячейке специальной конструкции. На интерферометре Цендера-Маха методом микроинтерферометрии проводились наблюдения за распределением концентрации пропановой кислоты вблизи поверхности раздела. Когда эти профили концентрации имеют обычный вид, эти явления определялись как межфазная турбулентность и наблюдались качественно.

Из этих экспериментов получены следующие результаты:

1. Существование устойчивой и осциллирующей турбулентности в межфазной туроулентности.

2. Процесс перехода небольших разностей концентрации после образования поверхности раздела в межфазную турбулентность.

3. Процесс изменения во времени межфазной турбулентности.

Исследуется течение жидкости при межфазной турбулентности, а такжепрослеживается движение мелкодисперсных частиц в системе. Обсуждается соотношение между межфазной турбулентностью и потоком жидкости.