the up- and downstrokes. At $\theta=60^\circ$, a vortex appears partly only in the upper or lower portion of the vessel depending on the up- or downstroke while at $\theta=120^\circ$ a vortex develops more or less in the whole vessel. The systematic examination of 120 pictures reveals that the transition of flow pattern occurs more rapidly just after turning back of the impeller from up to downstroke and vice versa, and more gradually around the middle height of the impeller.

The velocity component projected to the vertical plane was measured from the path of a polystyrene tracer particle on the photograph. Typical results are shown in **Fig. 2** for six selected points in the vessel at three different impeller orientations. The direction of the local velocity varies over a wide angle during the whole cycle or 20 revolutions of the impeller, because of its up- and down motion. The velocity also fluctuates greatly in spite of the laminar flow. The velocity is large at 0° orientation since the impeller lies in this plane. Inward flow into the region enclosed by the six selected points is predominant at $\theta=60^\circ$, while outward flow is predominant at $\theta=120^\circ$.

The occurrence of various flow patterns and the frequent change in direction and magnitude of the local velocity corresponding to the impeller height are the most distinguishing features compared to conventional single-mode (rotational) agitation, in which only a single flow pattern and a single local velocity appear at a given orientation. When the flow is observed from stationary coordinates, the sequence of changes in flow patterns becomes too complicated to identify and follow by visual observation alone, because of the simultaneous changes in height and orientation of the impeller. Since any point in the vessel experiences various orientations, the direction of fluid velocity changes in almost all directions during the whole cycle or 20 revolutions. Such a frequent variation of flow

Fig. 2 Variation of local velocity during whole cycle at various orientations

pattern and local velocity increases the opportunity of replacement between fluid elements as well as the extent of deformation of fluid, which in turn results in improvement of mixing efficiency as stated previously²⁾.

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Nomenclature

θ = impeller orientation [deg]

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LETTER TO THE EDITOR

HETEROGENEITY OF THE REACTION SYSTEM OF ALKALINE HYDROLYSIS OF ETHYL ACETATE IN A BATCH STIRRED TANK

Dear Sir:

Alkaline hydrolysis of ethyl acetate has been frequently used to investigate the micromixing phenomenon in the continuous perfect macromixer. However, this reaction has been regarded as reaction controlling because the reaction rate is considerably slower than the rate of mixing for miscible liquids in

the usual batch operation^{2,3)}.

Here are presented some supplementary considerations as to heterogeneity due to the formation of droplets which has been observed for this particular reaction¹⁾.

Solid symbols in **Fig. 1** are the experimental data obtained in the system where pure ethyl acetate was poured into the sodium hydroxide solution without a viscosity-increasing reagent. The rate of reaction is depressed with increasing reaction time. Almost the same course of reaction was also observed in viscous

solutions of methyl cellulose. As can be seen from the figure, the reaction behavior is well explained by the modified parallel-type model^{2,4)} denoted by the segregation function $s(t)=p'=0.09$ where the observed fact that a fractional volume of added pure ethyl acetate was segregated is taken into consideration. Hiraoka¹⁾ interpreted the reaction in a viscous solution of glycerine as a heterogeneous reaction under the effect of droplets of ethyl acetate.

Here some attempts were made to photograph the droplets. These droplets in reacting mixture could not be made visible by a light-scattering method nor by a conventional microscope. A microscope equipped with phase-difference lens enables one to see the image of droplets by using difference of refractive index between transparent liquid mixtures. A small amount of reacting mixture was sampled with a pipette and then set on the slide glass after acidification by adding hydrochloric acid solution to stop the reaction.

Photographs of **Fig. 2** show the shapes of droplets. Figure 2(a) shows the suspended liquid droplets of excess pure ethyl acetate in water, (b) the droplets remaining after carrying out the reaction by adding sodium hydroxide to the state of (a), and (c) the droplets observed in a sampled mixture taken from the reacting glycerine solution of viscosity $1.91 \cdot 10^{-3} \text{ Pa} \cdot \text{s}$. The scale of observed droplets is less than 10 microns.

Nomenclature

C	= concentration of solution	$[\text{mol} \cdot \text{dm}^{-3}]$
k_r	= 2nd-order reaction rate constant	$[\text{dm}^3 \cdot \text{mol}^{-1} \cdot \text{s}^{-1}]$
n	= impeller speed	$[\text{s}^{-1}]$
p'	= micromixing parameter for the parallel-type model ⁴⁾	$[-]$
t	= time	$[\text{s}]$
v	= volume	$[\text{dm}^3]$

<Subscripts>

a	= reactive liquid A or species A
b	= reactive liquid B or species B
m	= maximum mixedness
s	= complete segregation
0	= time zero or initial

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Fig. 1 Analysis of experimental data under droplet effect by modified parallel-type model

Fig. 2 Photographs of liquid droplets of ethyl acetate taken by microscope equipped with phase-difference lens

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