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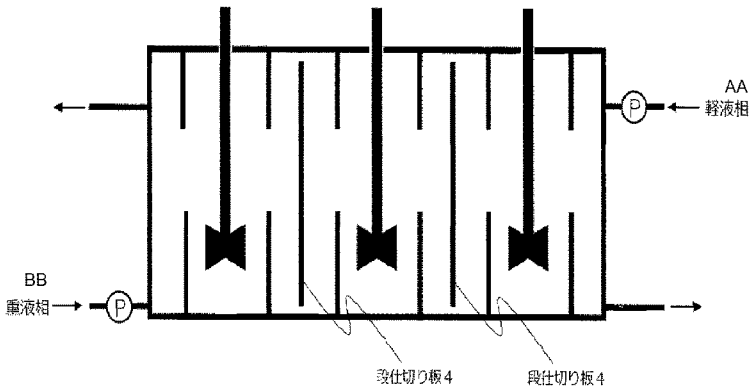
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JP 202194546

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Other: **JSTPlus/JSTChina/JST7580 (JDreamIII)**

(54) Title of the Invention: **Method for phase mixing of two liquid phases, and device therefor**
Abstract Title: **Method for phase mixing of two liquid phases, and device therefor**

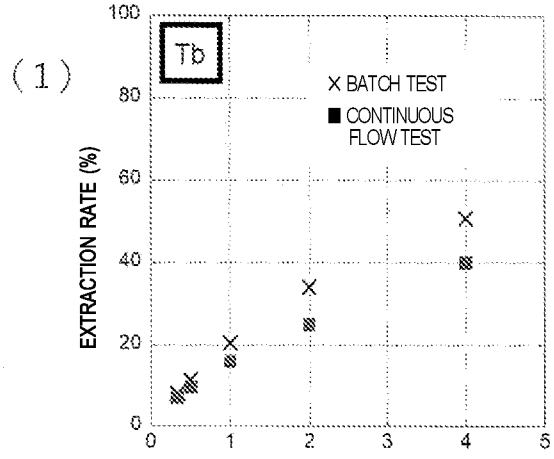
(57) [Problem] To make it possible to arbitrarily set a volume ratio between respective liquid phases involved in phase mixing, in a manner completely independent of the ratio between the liquid sending speeds, of the respective liquid phases, having an effective relationship expressed as a function regarding the extraction rate or the reverse extraction rate. [Solution] According to the present invention, during an operation to perform phase mixing while sending two liquid phases into a single reaction vessel, a region of phase mixing and a region of phase separation are made to coexist at a certain range within the reaction vessel.

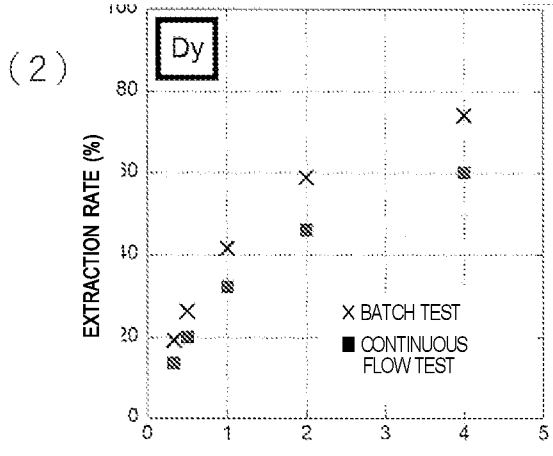


4 Stage partition plate
AA Light liquid phase
BB Heavy liquid phase

GB 2631184 A

FIG. 1(A)


(1)
Tb
X BATCH TEST
■ CONTINUOUS FLOW TEST
EXTRACTION RATE (%)
0
20
40
60
80
100
0
1
2
3
4
5
EFFECTIVE ORGANIC PHASE/AQUEOUS PHASE VOLUME RATIO (BATCH TEST)
EFFECTIVE ORGANIC PHASE/AQUEOUS PHASE FEED RATE RATIO (CONTINUOUS FLOW TEST)


(2)
Dy
X BATCH TEST
■ CONTINUOUS FLOW TEST
EXTRACTION RATE (%)
0
20
80
0
1
2
3
4
5
EFFECTIVE ORGANIC PHASE/AQUEOUS PHASE VOLUME RATIO (BATCH TEST) OR
EFFECTIVE ORGANIC PHASE/AQUEOUS PHASE FEED RATE RATIO (CONTINUOUS FLOW TEST)

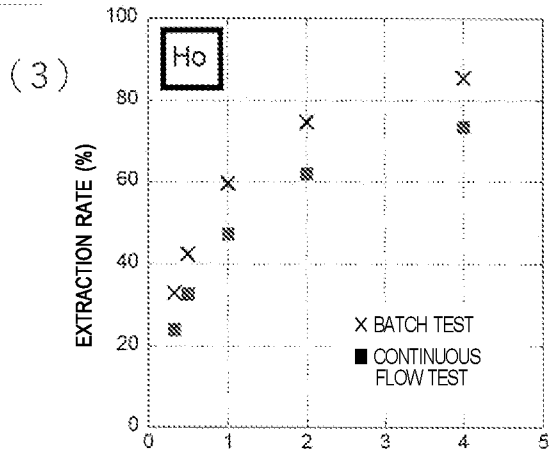

(3)
Ho
X BATCH TEST
■ CONTINUOUS FLOW TEST
EXTRACTION RATE (%)
0
20
40
60
80
100
0
1
2
3
4
5
EFFECTIVE ORGANIC PHASE/AQUEOUS PHASE VOLUME RATIO (BATCH TEST) OR
EFFECTIVE ORGANIC PHASE/AQUEOUS PHASE FEED RATE RATIO (CONTINUOUS FLOW TEST)

FIG. 1(B)

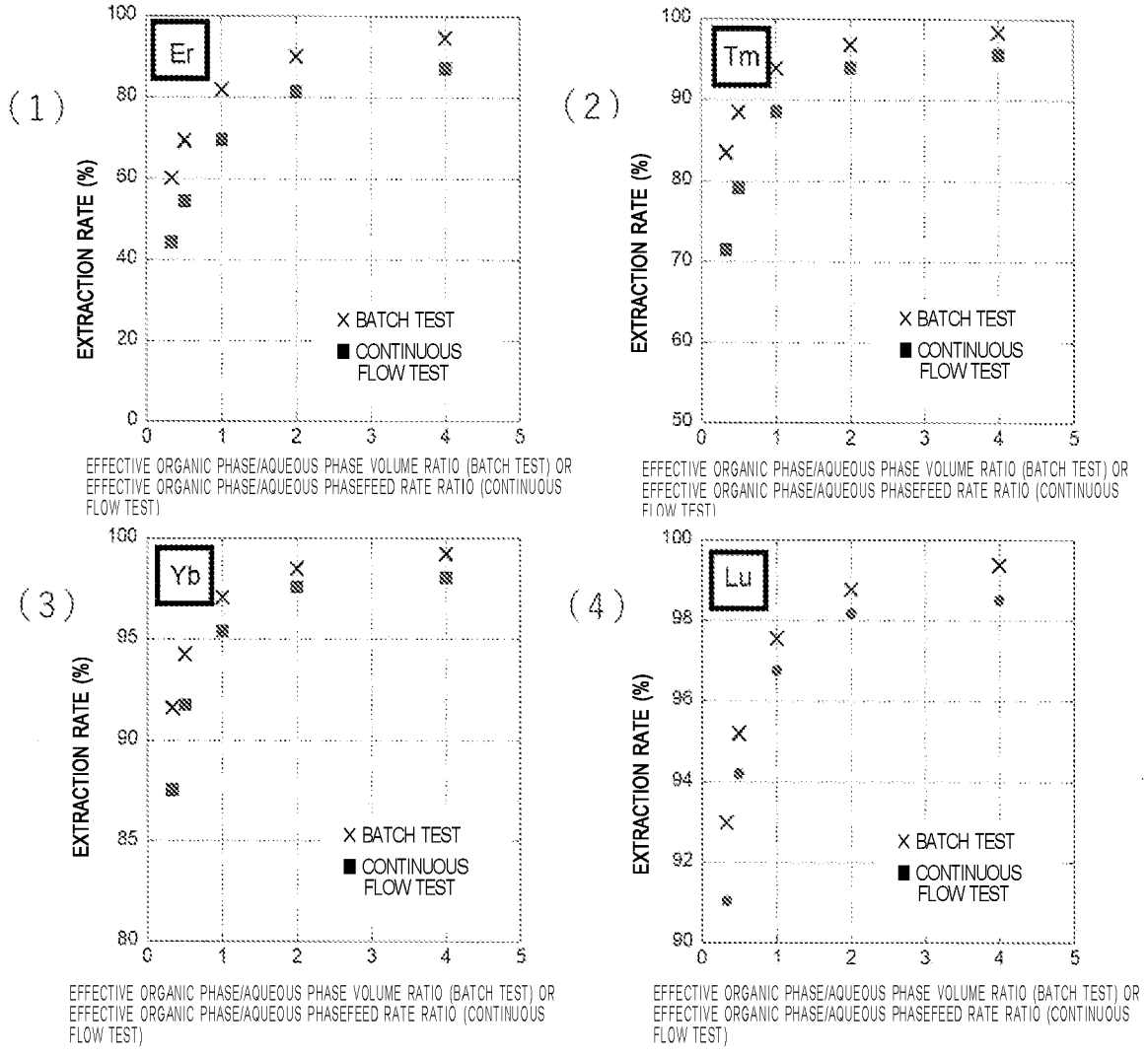

(1)
Er
EXTRACTION RATE (%)
X BATCH TEST
■ CONTINUOUS FLOW TEST
EFFECTIVE ORGANIC PHASE/AQUEOUS PHASE VOLUME RATIO (BATCH TEST) OR EFFECTIVE ORGANIC PHASE/AQUEOUS PHASEFEED RATE RATIO (CONTINUOUS FLOW TEST)
(2)
Tm
EXTRACTION RATE (%)
X BATCH TEST
■ CONTINUOUS FLOW TEST
EFFECTIVE ORGANIC PHASE/AQUEOUS PHASE VOLUME RATIO (BATCH TEST) OR EFFECTIVE ORGANIC PHASE/AQUEOUS PHASEFEED RATE RATIO (CONTINUOUS FLOW TEST)
(3)
Yb
EXTRACTION RATE (%)
X BATCH TEST
■ CONTINUOUS FLOW TEST
EFFECTIVE ORGANIC PHASE/AQUEOUS PHASE VOLUME RATIO (BATCH TEST) OR EFFECTIVE ORGANIC PHASE/AQUEOUS PHASEFEED RATE RATIO (CONTINUOUS FLOW TEST)
(4)
Lu
EXTRACTION RATE (%)
X BATCH TEST
■ CONTINUOUS FLOW TEST
EFFECTIVE ORGANIC PHASE/AQUEOUS PHASE VOLUME RATIO (BATCH TEST) OR EFFECTIVE ORGANIC PHASE/AQUEOUS PHASEFEED RATE RATIO (CONTINUOUS FLOW TEST)

FIG. 2

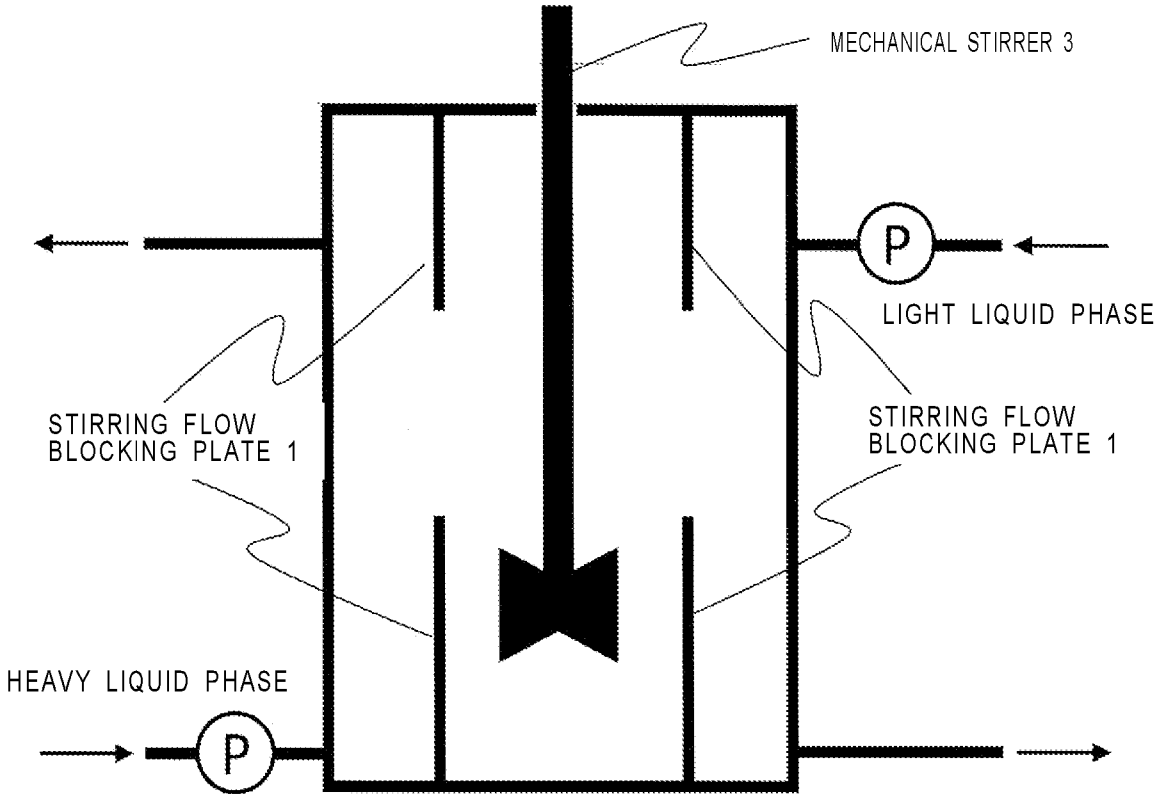

MECHANICAL STIRRER 3
P
LIGHT LIQUID PHASE
STIRRING FLOW BLOCKING PLATE 1
STIRRING FLOW BLOCKING PLATE 1
HEAVY LIQUID PHASE
P

FIG. 3

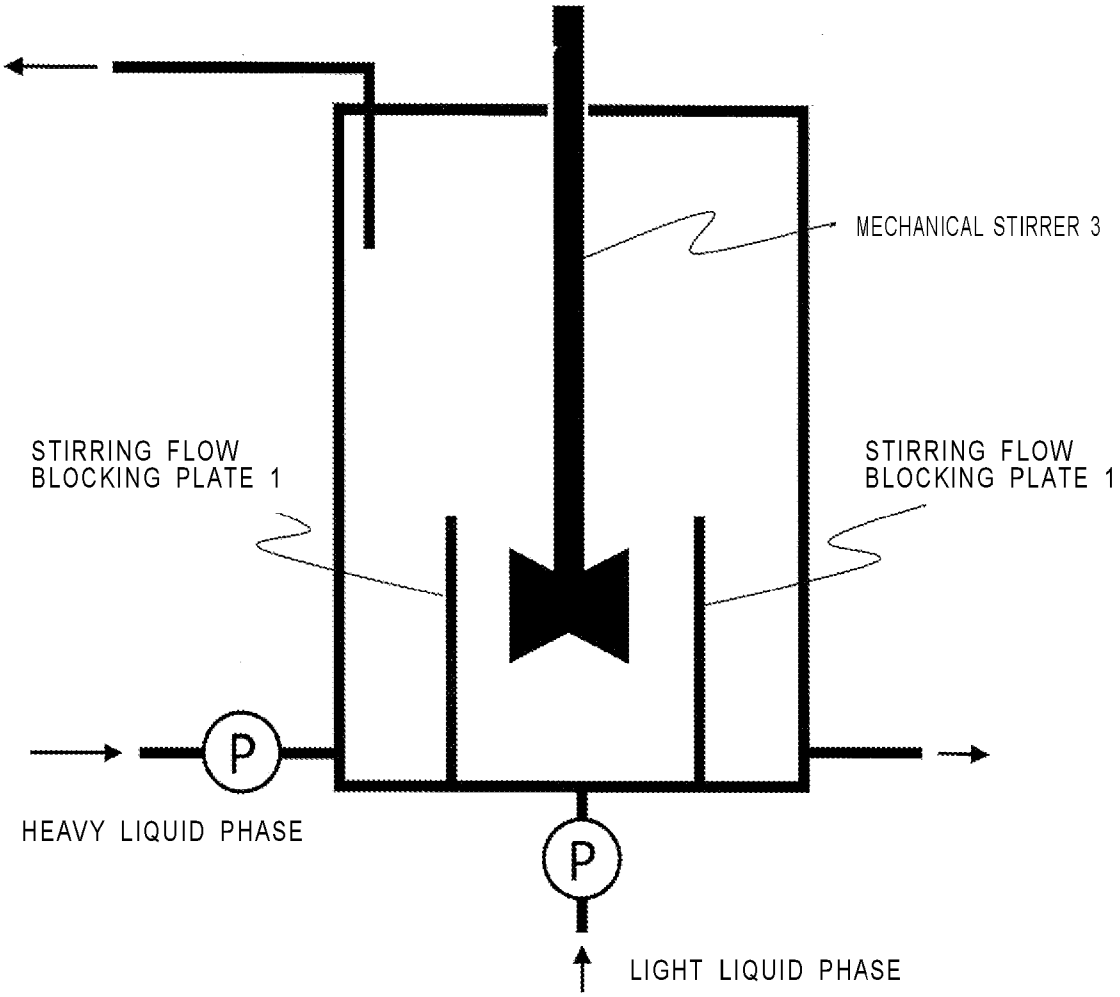

MECHANICAL STIRRER 3
STIRRING FLOW BLOCKING PLATE 1
STIRRING FLOW BLOCKING PLATE 1
P
P
HEAVY LIQUID PHASE
LIGHT LIQUID PHASE

FIG. 4

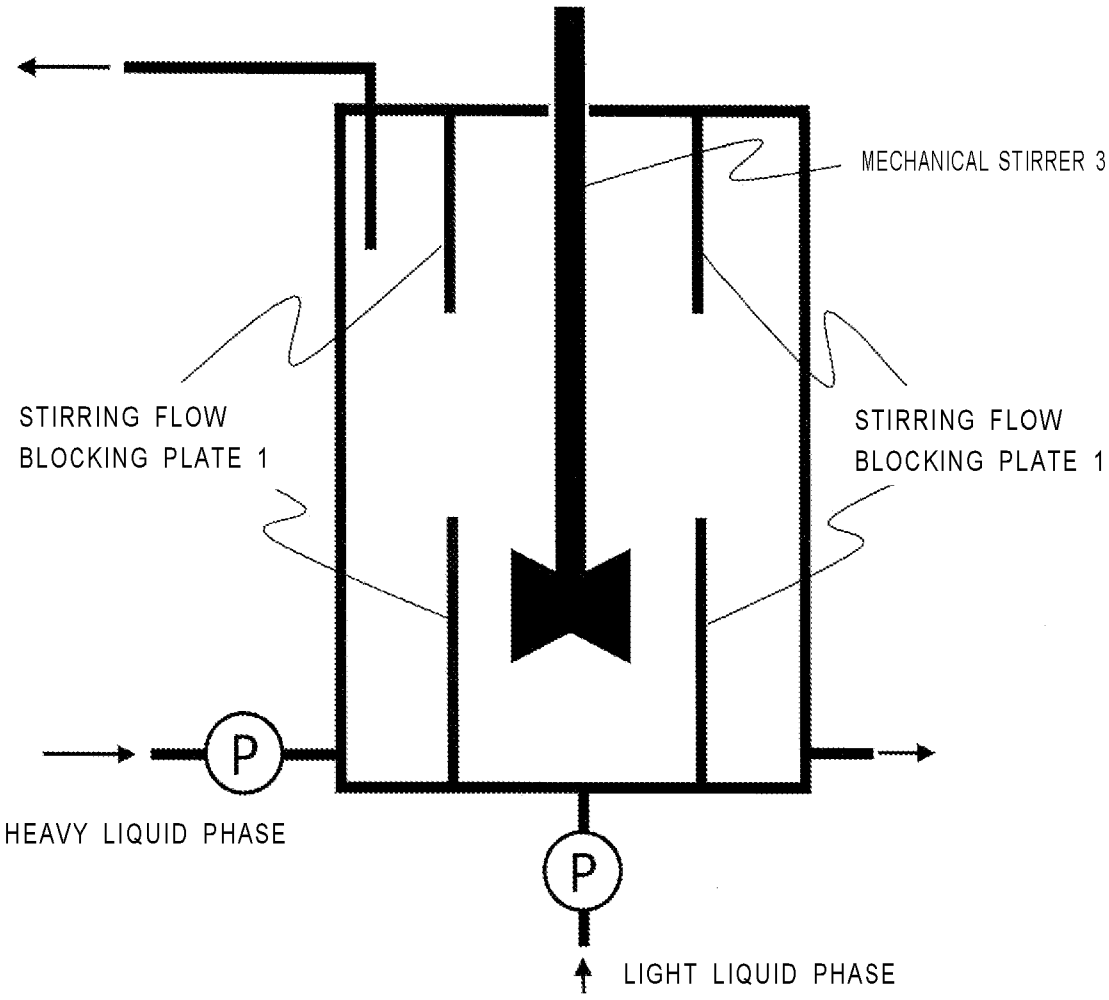

MECHANICAL STIRRER 3
STIRRING FLOW
BLOCKING PLATE 1
STIRRING FLOW
BLOCKING PLATE 1
P
HEAVY LIQUID PHASE
P
LIGHT LIQUID PHASE

FIG. 5

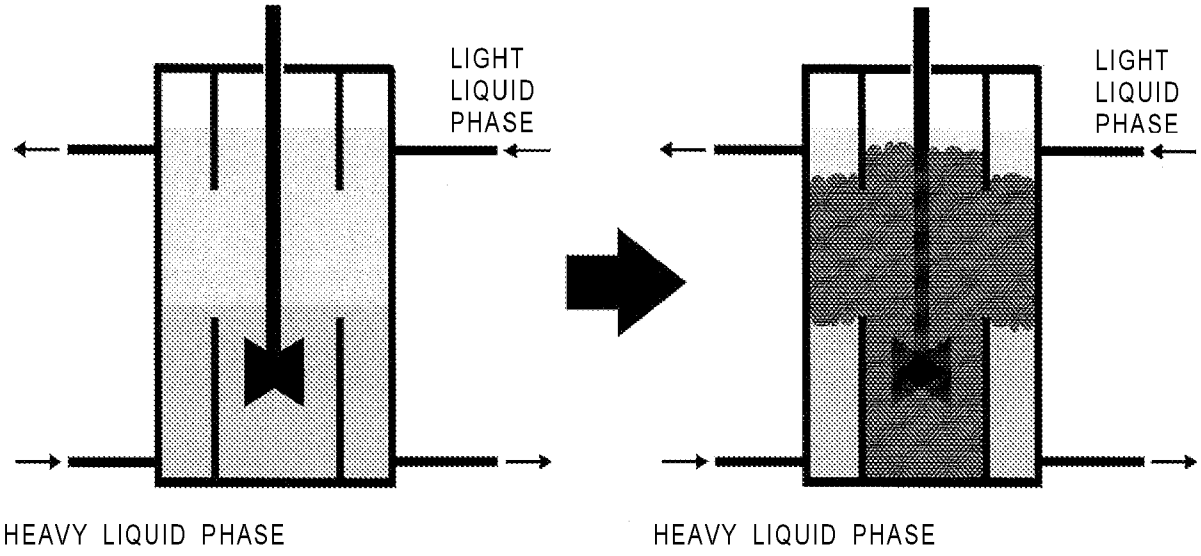

LIGHT
LIQUID
PHASE
HEAVY LIQUID PHASE
LIGHT
LIQUID
PHASE
HEAVY LIQUID PHASE

FIG. 6

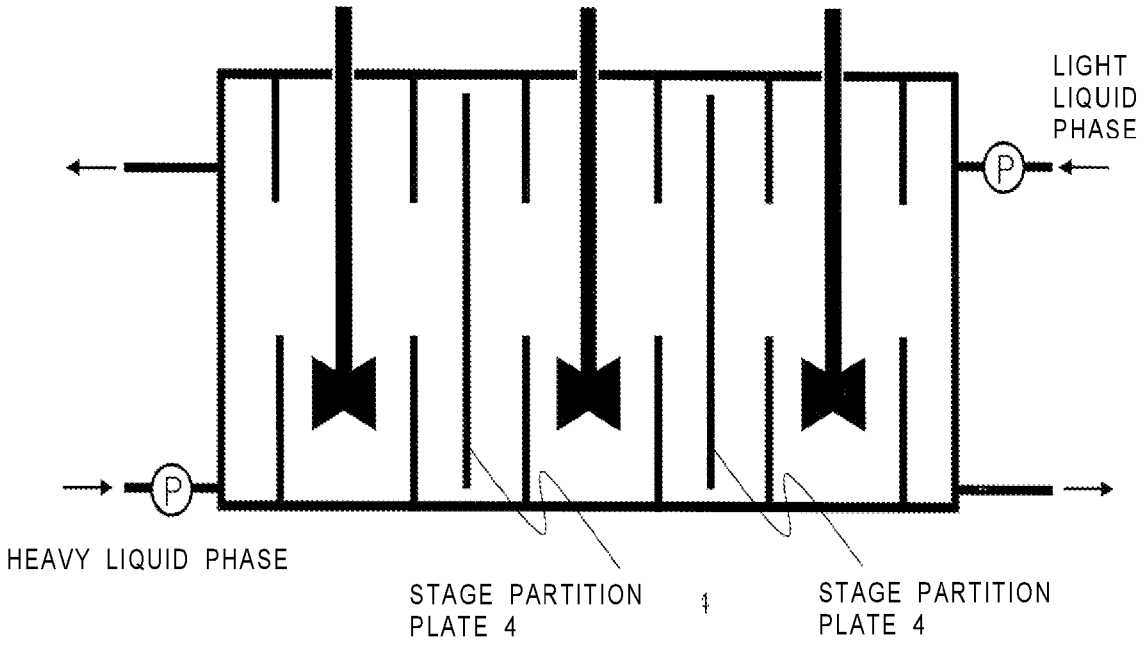

LIGHT
LIQUID
PHASE
P
P
HEAVY LIQUID PHASE
STAGE PARTITION
PLATE 4
STAGE PARTITION
PLATE 4

FIG. 7

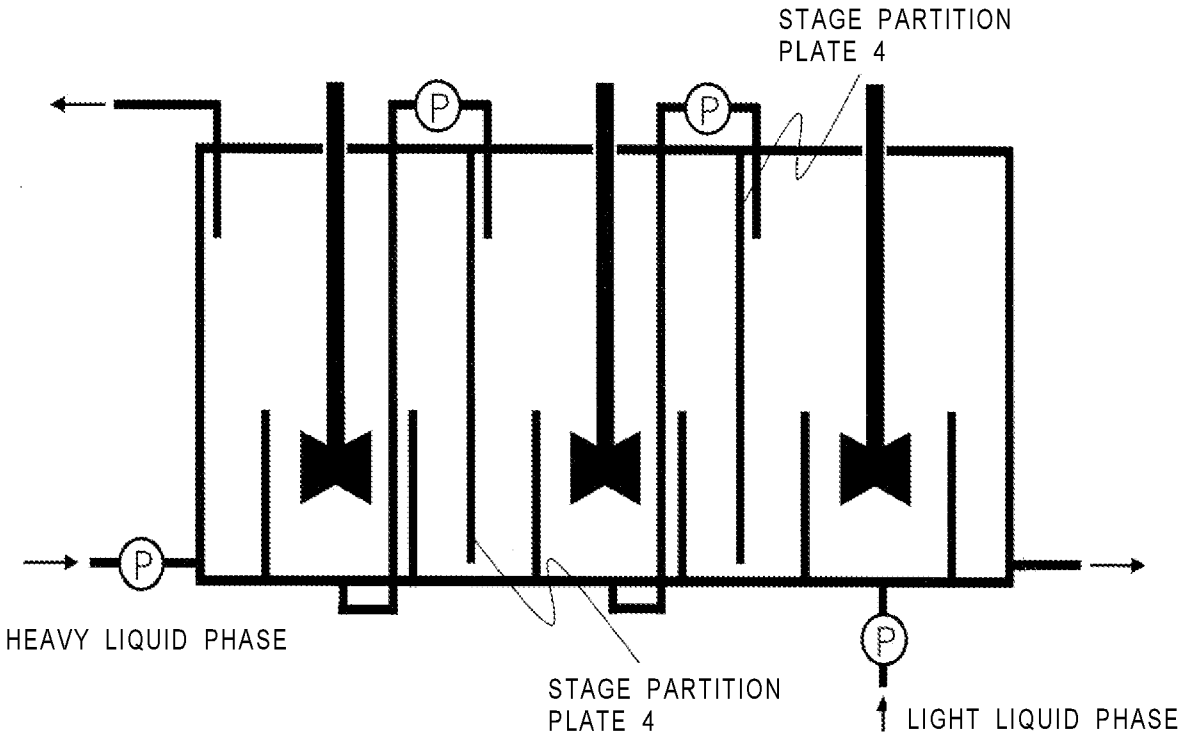

STAGE PARTITION
PLATE 4
P
P
P
P
HEAVY LIQUID PHASE
STAGE PARTITION
PLATE 4
LIGHT LIQUID PHASE

FIG. 8

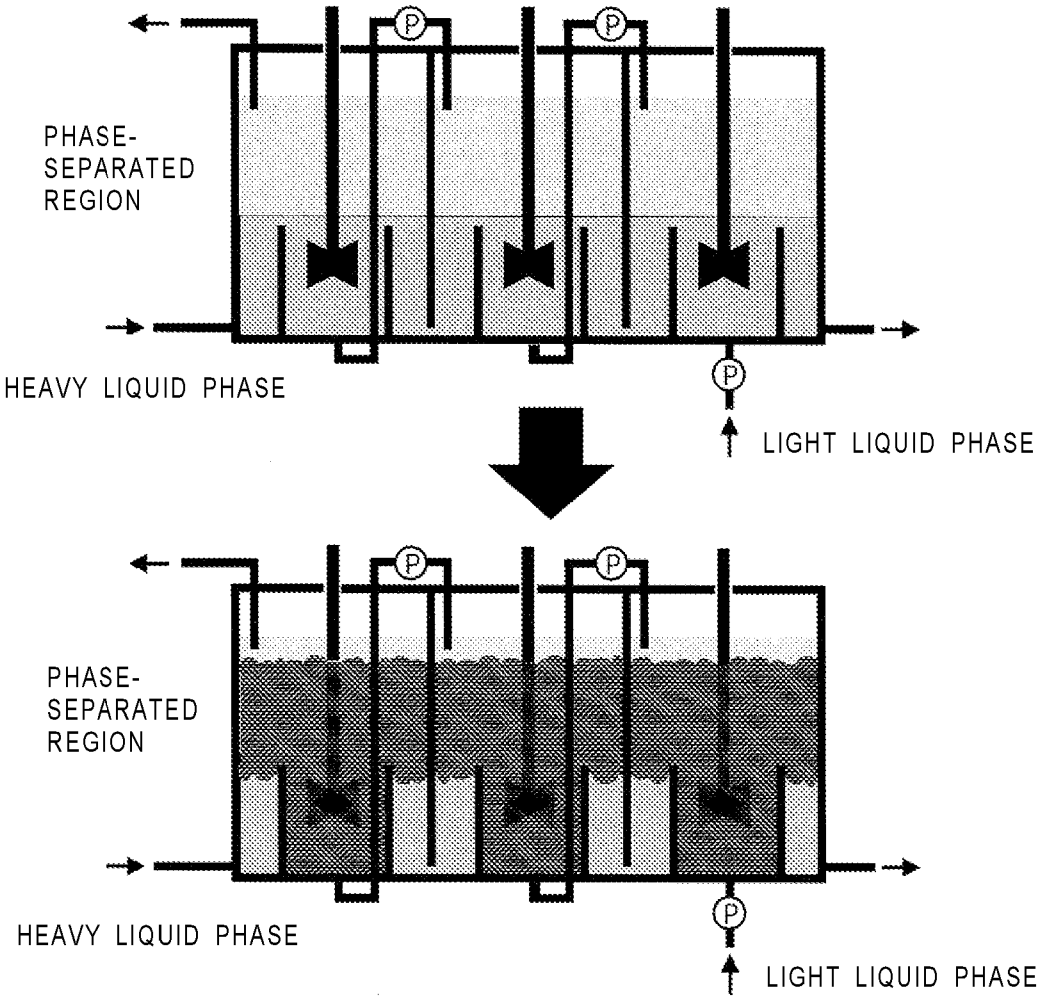

PHASE-
SEPARATED
REGION
HEAVY LIQUID PHASE
LIGHT LIQUID PHASE
PHASE-
SEPARATED
REGION
HEAVY LIQUID PHASE
LIGHT LIQUID PHASE

FIG. 9

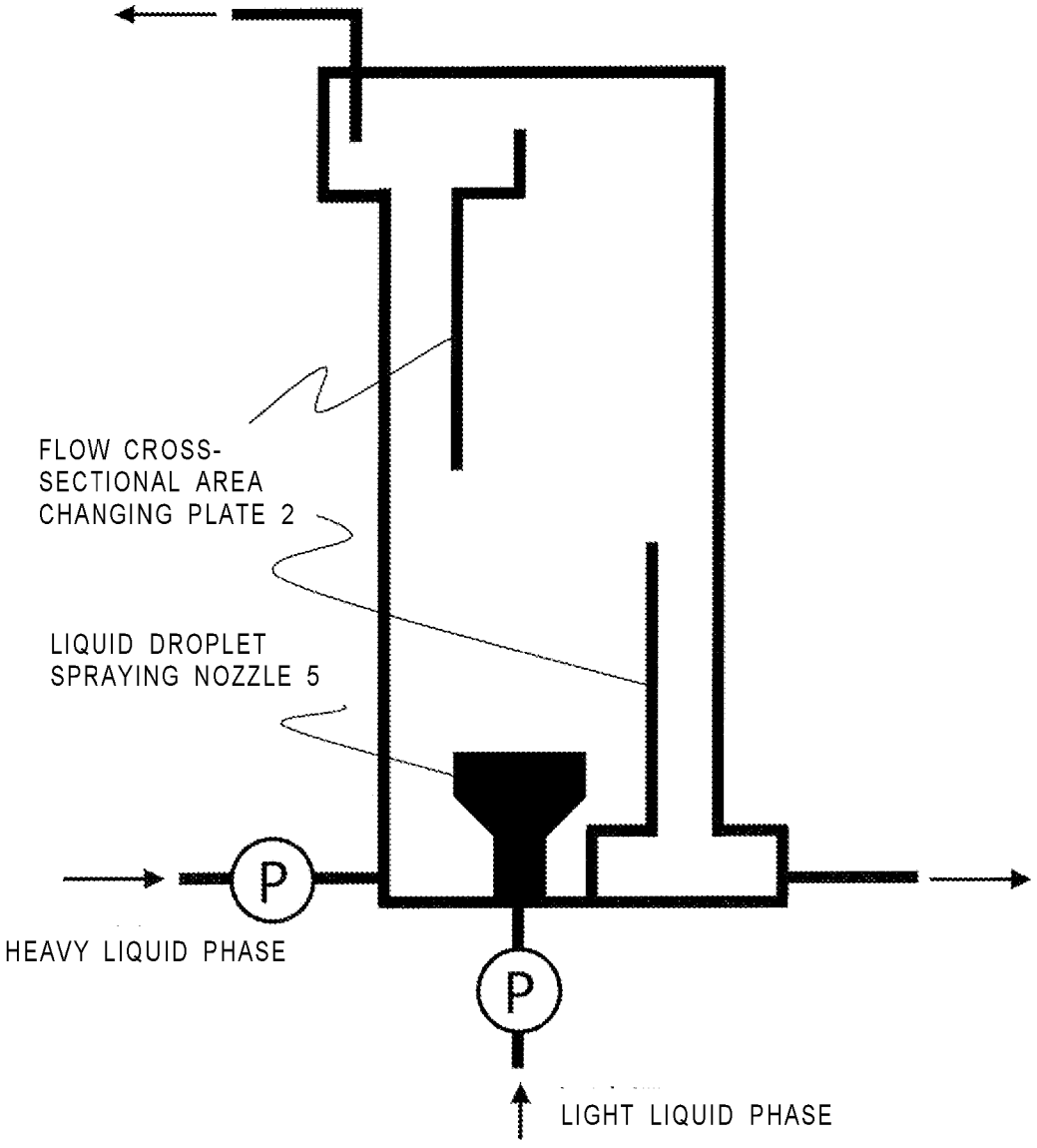

FLOW CROSS-SECTIONAL AREA CHANGING PLATE 2
LIQUID DROPLET SPRAYING NOZZLE 5
P
P
HEAVY LIQUID PHASE
LIGHT LIQUID PHASE

FIG. 10

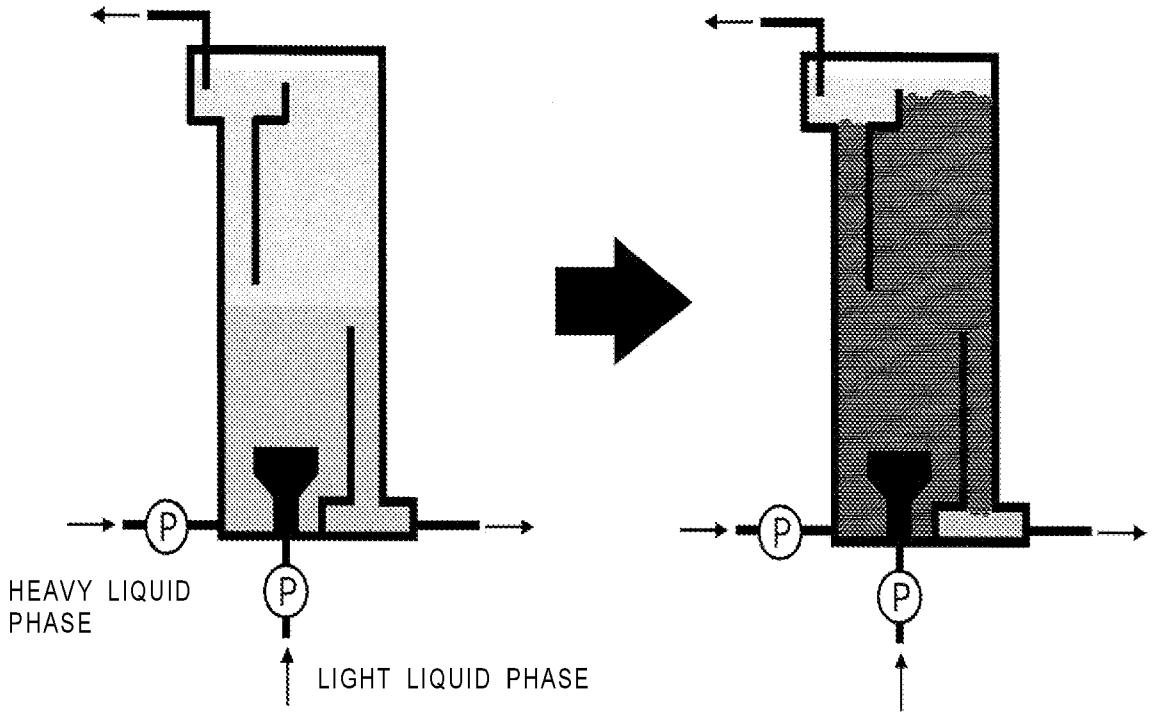


FIG. 11

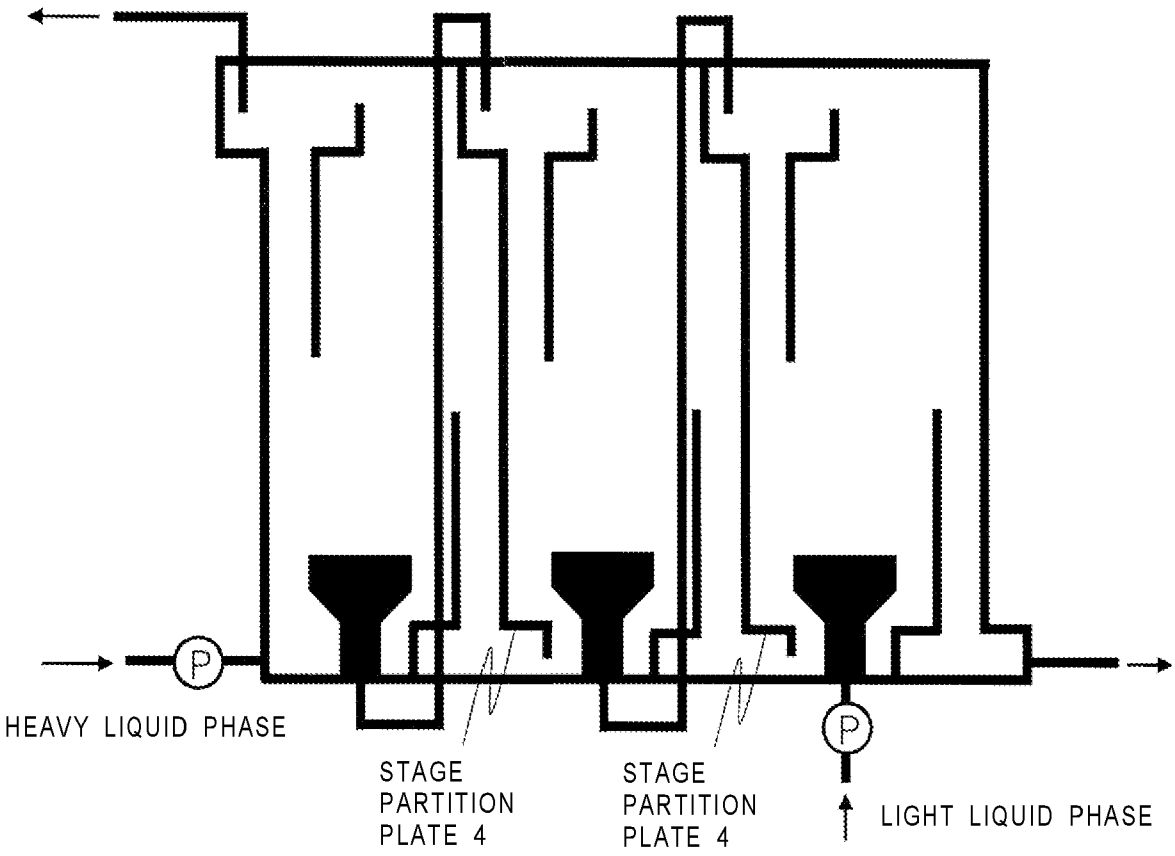
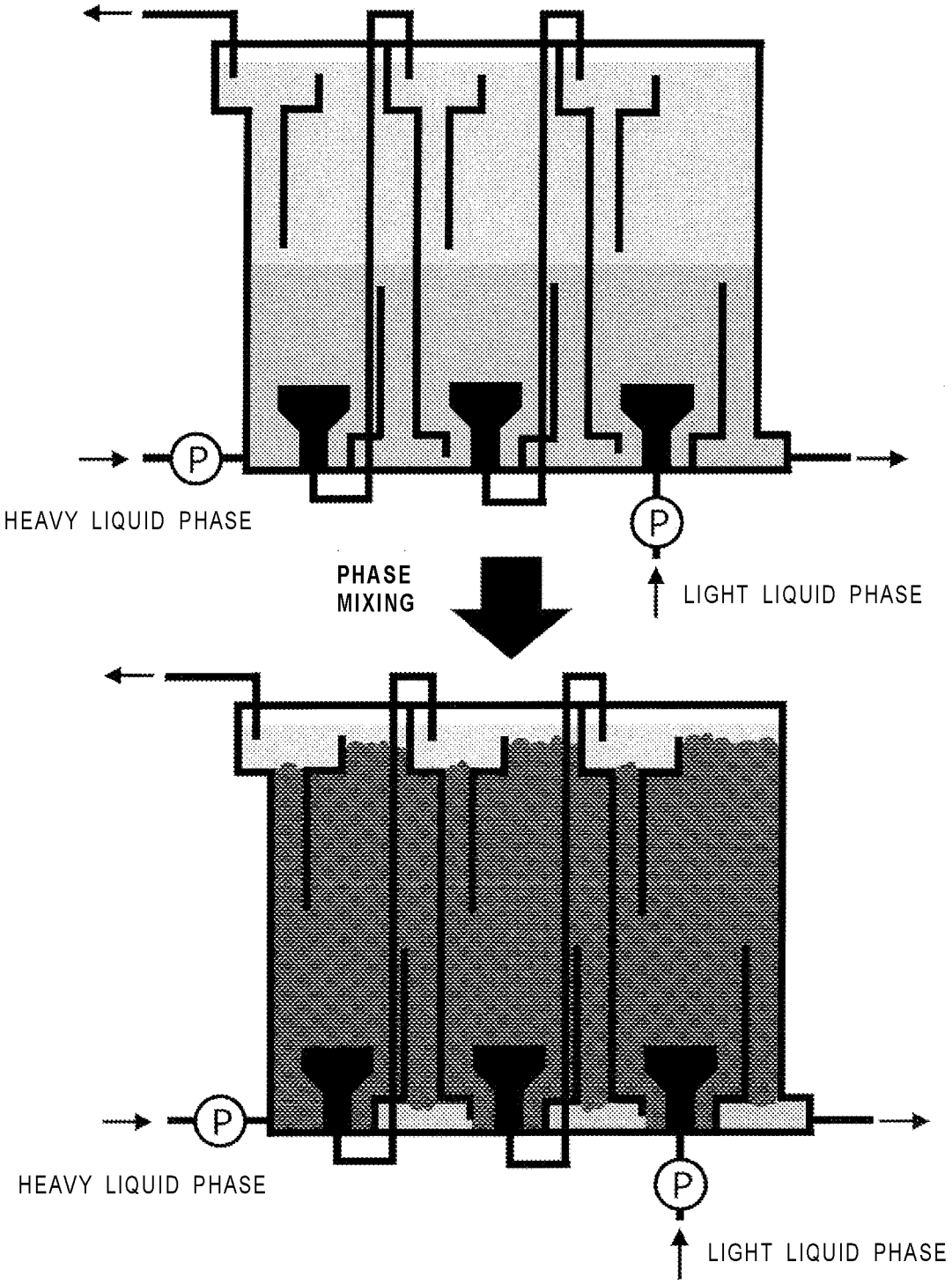

P
HEAVY LIQUID PHASE
STAGE
PARTITION
PLATE 4
STAGE
PARTITION
PLATE 4
P
LIGHT LIQUID PHASE

FIG. 12


P
HEAVY LIQUID PHASE
P
LIGHT LIQUID PHASE
PHASE
MIXING
P
HEAVY LIQUID PHASE
P
LIGHT LIQUID PHASE

SPECIFICATION

METHOD FOR PHASE MIXING OF TWO LIQUID PHASES, AND DEVICE THEREFOR

[Technical Field]

[0001]

The present invention relates to a method and an apparatus for phase mixing, characterized in that the volume ratio of liquid phases involved in phase mixing in one reaction vessel can be set arbitrarily, completely independent of the ratio of the feed rate of each liquid phase, which is an effective functional relationship to the extraction rate and the back-extraction rate.

[Background Art]

[0002]

Two-phase extraction, which consists of two immiscible liquid phases, is widely used in chemical techniques such as solvent extraction. This method, for example, is one of the most important separation and purification techniques for metallic elements and organic compounds, which support key industries such as the metal and chemical industries, as well as rare metals that are essential in high-tech industries.

[0003]

On the other hand, solvent extraction is considered to be a method with a large environmental impact because the entrainment of organic components into the wastewater is likely to occur. In addition, depending on the volume ratio of the aqueous and organic phases to be mixed, the region involved in phase mixing (called the mixed phase) may be unstable, the mixed phase may not flow easily, or phase separation after the extraction reaction may be difficult, which may cause solvent extraction to proceed inefficiently.

[0004]

Phase mixing refers to mixing two immiscible liquid phases that form an interface by stirring, shaking, etc. Thereby, if each liquid phase is refined and the interface area is increased, an emulsion state is reached. In an emulsion state, the transfer of substances between phases is promoted, making it easier to reach chemical equilibrium in the extraction reaction. Stirring and shaking are common methods of phase mixing, but recently, phase mixing to an emulsion state by droplet spraying is also known (e.g., Patent Document 1 and Patent Document 2). Phase separation refers to the process of separating two mixed liquid phases into their respective liquid phases again. Common methods are natural separation using gravity and buoyancy, and mechanical separation using centrifugal force.

[0005]

When a batch test is performed in which a aqueous phase containing the component to be extracted and an organic phase mainly made of a solvent that is not miscible with water are placed in a test tube or other container at a fixed volume ratio and shaken sufficiently until the extraction reaction reaches chemical equilibrium, the extraction rate of the component to be extracted into the organic phase depends on the volume ratio of the two phases at that time (Here, the volume ratio of the organic phase to the aqueous phase at this time is referred to as the effective organic phase/aqueous phase volume ratio.). In other words, at the time of chemical equilibrium of the extraction reaction, the relationship between the extraction rate (= E%) and the effective organic phase/aqueous phase volume ratio (= RV) is a function expressed as $E\% = 100 \times RV \times D/(RV \times D + 1)$ via the distribution ratio (the ratio D of the concentration of the solute in the organic phase to the concentration of the solute in the aqueous phase). For example, when the extraction rate of the extraction target component is to be increased, the effective organic phase/aqueous phase volume ratio should be increased.

[0006]

When industrially performing continuous solvent extraction while pumping the aqueous and organic phases, the ratio (referred to as the organic phase/aqueous phase feed rate ratio) of the organic phase feed rate to the aqueous phase corresponds to the organic phase/aqueous phase volume ratio (referred to as the organic phase/aqueous phase volume ratio in the mixed phase) involved in the phase mixing. This coincides with the effective organic phase/aqueous phase volume ratio mentioned in the batch test above. That is, in conventional continuous solvent extraction methods and apparatuses, the relationship is such that the organic phase/aqueous phase feed rate ratio = the organic phase/aqueous phase volume ratio in the mixed phase = the effective organic phase/aqueous phase volume ratio.

[0007]

In this case, the volume ratio of the two phases when they are mixed, i.e., the volume ratio of the two phases in the mixed phase, is determined by the ratio of the feed rate of the aqueous phase and the organic phase. On the other hand, the physical properties of the mixed phase (stability, phase separation, fluidity, ease of entrainment, etc.) depend on the volume ratio of the aqueous phase and the organic phase that form the mixed phase. In other words, in conventional methods and apparatuses, the physical properties of the mixed phase are dependent on the ratio of the feed rate of the two liquid phases.

[Prior Art Documents]

[Patent Documents]

[0008]

[Patent Document 1] JP 5305382 B

[Patent Document 2] JP 5565719 B

[Summary of the Invention]

[Problems to be Solved by the Invention]

[0009]

When two liquid phases are phase-mixed at different volumes in one reaction vessel, the liquid phase of the smaller volume tends to mix with the other phase as fine droplets. In other words, the smaller liquid phase tends to cause so-called entrainment (droplet entrainment).

[0010]

In many cases, the physical properties of a mixed phase consisting of two liquid phases differ greatly depending on the mixing ratio. For example, for a mixed phase consisting of an aqueous phase and an organic phase, the stability of the mixed phase, ease of flow of the mixed phase, ease of entrainment, and time required for phase separation may differ significantly depending on whether the mixed phase is aqueous phase rich or organic phase rich.

[0011]

Especially in apparatus with a multi-stage structure in which multiple stages are set up and unit operations are repeated, the above differences in the ease of entrainment and the physical properties of the mixed phase have a significant impact on whether the apparatus/plant can be operated or not. In other words, problems arise such as the inability to separate the phases, not being able to proceed to the next stage due to the lack of flow, or not being able to remove impurities according to the set number of stages due to entrainment.

[0012]

In the usual method of phase mixing where an area of phase mixing (mixed phase) is created throughout the entirety of a reaction vessel, the ratio of the volume of each liquid phase comprising the mixed phase in the vessel is equal to the ratio of the liquid feed rate of each liquid phase. In other words, the liquid phase with a slower feed rate is also smaller in volume of the mixed phase, and thus entrainment is more likely to occur. In addition, depending on the ratio of the feed rate of each liquid phase, the mixed phase may be unstable or difficult to flow, which may cause operational problems. Furthermore, the tendency to entrainment and the long time required for phase separation may also

cause problems with drainage and other processes.

[0013]

Because the actual solvent extraction process often requires increasing the feed rate of the organic phase to increase the extraction rate into the organic phase or increasing the feed rate of the aqueous phase to increase the back extraction rate into the aqueous phase, each time such adjustments are made, we are faced with the problems described above. In addition, since these problems cannot always be solved, the solvent extraction process itself may become unfeasible.

[0014]

As a result of extensive research aimed at solving these problems, we have found that by utilizing a mechanism in which phase mixing and phase separation occur simultaneously in one reaction vessel, even when the volume ratio of each liquid phase involved in the phase mixing is set arbitrarily and completely independent of the ratio of the liquid flow rates of each liquid phase, the ratio of the feed rate of each liquid phase has an effective functional relationship with the extraction rate or back-extraction rate.

[0015]

In other words, while improving the extraction rate or back-extraction rate by adjusting the feed rate ratio, the volume ratio of the aqueous and organic phases involved in phase mixing can be arbitrarily set to a ratio that is less likely to cause entrainment due to high stability, flowability, and phase separation of the mixed phase.

[0016]

Therefore, the object of the present invention is to provide a method and an apparatus for mixing two liquid phases, which allows the volume ratio of each liquid phase involved in the phase mixing to be set arbitrarily completely independent of the ratio of the liquid delivery speeds of each liquid phase, which has an effective functional relationship with the extraction rate and back extraction rate. As for the apparatus, we are strongly aware of multi-stage apparatus, where the

susceptibility to entrainment and the influence of differences in the physical properties of the mixed phases become more pronounced.

[Means for Solving the Problems]

[0017]

By utilizing a mechanism in which phase mixing and phase separation occur simultaneously in a single reaction vessel, the volume ratio of each liquid phase subjected to phase mixing can be set arbitrarily and completely independently of the ratio of the liquid feed rate of each liquid phase, and the ratio of the feed rate of each liquid phase can be made to have an effective functional relationship to the extraction rate or the back-extraction rate.

[0018]

The basic feature of this invention is that in the operation of phase mixing while pumping two liquid phases into a single reaction vessel, the organic phase/aqueous phase volume ratio in the phase-mixed region (mixed phase) where the aqueous phase and organic phase undergo phase mixing can be set arbitrarily and completely independent of the organic phase/aqueous phase feed rate ratio by allowing the phase-mixed region and phase-separated region to coexist within a certain range in said reaction vessel. It is a key point of the invention that the above-mentioned organic phase/aqueous phase feed rate ratio is ideally identical to the effective organic phase/aqueous phase volume ratio, which is a parameter in the chemical equilibrium function for the extraction rate and the back-extraction rate.

[Effect of the Invention]

[0019]

Conventionally, in terms of the stability, fluidity, phase separation, or entrainment of the mixed phase, the volume ratio of the aqueous phase and the organic phase involved in the phase mixing had to be set under unfavorable conditions. However, according to the

method and apparatus of the present invention, it becomes possible to arbitrarily set the volume ratio under favorable conditions in which these problems do not occur.

[0020]

In conventional methods, the aqueous phase and the organic phase are mixed throughout the reaction vessel, and then the mixed phase flows into a settling vessel located next to the reaction vessel, where the two phases are separated by gravity. In contrast, in the method of the present invention, the mixed phase and the separated aqueous phase and organic phase coexist within a certain range in the reaction vessel, so no settling vessel is required, and there is no need to wait for the mixed phase to separate by gravity.

[0021]

Next, the gist of the present invention is explained below. In the conventional method, when the feed rate of the aqueous or organic phase or both is changed, the volume ratio of the aqueous and organic phases in the mixed phase also changes, and accordingly, the physical properties and entrainment state of the mixed phase also change. Therefore, it is essential to adjust the position of the interface in the settling vessel, for example, to accommodate this change. In contrast, in the method of the present invention, where the volume ratio of the aqueous and organic phases in the mixed phase is not affected by the feed rate of the two phases, the physical properties and entrainment state of the mixed phase are always constant. Therefore, the method does not require a number of adjustments (e.g., adjusting the position of the interface in a settling vessel) that are indispensable in the conventional method.

[0022]

Furthermore, in the conventional method, the aqueous and organic phases are phase-mixed throughout the reaction vessel, which inevitably results in the mixed phase (the area disturbed by phase mixing) being exposed to air. In contrast, in the method of the present

invention, the mixed phase is always located below the phase-separated organic phase (undisturbed region), so that the disturbed mixed phase is never exposed to air. Thus, for example, if a target component that is easily oxidized (easily affected by oxygen in the air) is the subject of solvent extraction, the extraction operation can be performed while suppressing the effect.

[Brief Description of Drawings]

[0023]

Fig. 1(A) shows the relationship of the extraction rates of Tb, Dy, and Ho to effective volume ratio of organic phase/aqueous phase in a batch test or feed rate ratio of organic phase/aqueous phase in a continuous flow test.

Fig. 1(B) shows the relationship of the extraction rates of Er, Tm, Yb, and Lu to effective volume ratio of organic phase/aqueous phase in a batch test or Feed rate ratio of organic phase/aqueous phase in a continuous flow test.

FIG. 2 is a diagram showing a mechanically stirred phase mixing unit having upper and lower stirring flow blocking plates, and shows a mechanism in which a light liquid phase is introduced from above while being discharged from above, and a heavy liquid phase is introduced from below while being discharged from below.

FIG. 3 is a diagram showing a mechanically stirred phase mixing unit having a stirring flow blocking plate only on the lower side, and shows a mechanism in which a light liquid phase is introduced from below while being discharged from above, and a heavy liquid phase is introduced from below while being discharged from below.

FIG. 4 is a diagram showing a mechanically stirred phase mixing unit having upper and lower stirring flow blocking plates, and shows a mechanism in which a light liquid phase is introduced from below while being discharged from above, and a heavy liquid phase is introduced from below while being discharged from below.

FIG. 5 is a diagram showing the distribution of phase-mixed regions and phase-separated regions in the mechanically stirred phase-mixing unit shown in FIG. 2.

FIG. 6 is a diagram showing a three-stage mechanically stirred phase mixing apparatus having upper and lower stirring flow blocking plates, showing a mechanism in which a light liquid phase is introduced from above while being discharged from above, and a heavy liquid phase is introduced from below while being discharged from below.

FIG. 7 shows a three-stage mechanically stirred phase mixing apparatus having a stirring flow blocking plate only on the lower side, and shows a mechanism in which the light liquid phase is introduced from the bottom while being discharged from the top, and the heavy liquid phase is introduced from the bottom while being discharged from the bottom.

FIG. 8 is a diagram showing the distribution of phase-mixed regions and phase separation regions in the three-stage mechanically stirred phase mixing apparatus shown in FIG. 7.

FIG. 9 is a diagram showing a droplet spraying type phase mixing unit having flow path cross-sectional area changing plates on the top and bottom, showing a mechanism in which a light liquid phase is introduced from below while being discharged from above, and a heavy liquid phase is introduced from below while being discharged from below.

FIG. 10 is a diagram showing the distribution of phase-mixed regions and phase-separated regions in a phase mixing unit of a droplet spraying type shown in FIG. 9.

FIG. 11 is a diagram showing a three-stage droplet spraying type phase mixing apparatus having flow path cross-sectional area changing plates on the top and bottom, showing a mechanism in which a light liquid phase is introduced from below while being discharged from above, and a heavy liquid phase is introduced from below while being discharged from below.

FIG. 12 is a diagram showing the distribution of phase-mixed

regions and phase separation regions in the three-stage droplet spraying type phase mixing apparatus shown in FIG. 11.

[Mode for Carrying Out the Invention]

[0024]

Usually, the volume ratio of the aqueous and organic phases in the phase-mixed region in solvent extraction corresponds to the ratio of the feed rate of the aqueous and organic phases that are fed into that phase-mixed region. For example, in the mixer section of a mixer-settler, which is a typical apparatus for industrial solvent extraction, the entire region is the phase-mixed region (mixed phase), and the mixing volume ratio (volume ratio of both phases in the mixed phase) and the feed rate ratio (feed ratio of both phases) are identical.

[0025]

In this case, feed rate ratio of organic phase/aqueous phase is equal to the volume ratio of organic phase/aqueous phase in the phase-mixed region (mixed phase) and can be equated with the effective volume ratio of organic phase/aqueous phase (volume ratio of organic phase/aqueous phase in the above-mentioned batch test), which is a function of the extraction rate and back extraction rate. In fact, when the liquid in the phase-mixed region (mixed phase) is collected with a pipette or other dispensing apparatus and then phase-separated, the volume ratio of organic phase/aqueous phase is then identical to feed rate ratio of organic phase/aqueous phase.

[0026]

On the other hand, if the phase-mixed region in the reaction vessel where the aqueous and organic phases are subjected to phase mixing is partial and remains within a certain range, the relationship that the above-mentioned mixing volume ratio and the above-mentioned feed rate ratio are equal does not hold. In other words, in a system where phase mixing and phase separation occur simultaneously in one reaction vessel and each region coexists within a certain range, the ratio of the

feed rate of the aqueous phase to the organic phase can be freely changed while keeping the volume ratio of the aqueous phase to the organic phase in the phase-mixed region constant.

[0027]

In this case, when the liquid in the phase-mixed region (mixed phase) was actually collected with a pipette or other dispensing apparatus and then phase separated, the volume ratio of organic phase/aqueous phase at that time was always constant and independent of feed rate ratio of organic phase/aqueous phase. This is why feed rate ratio of organic phase/aqueous phase has not been discussed in relation to the effective volume ratio of organic phase/aqueous phase (the volume ratio of organic phase/aqueous phase in the above-mentioned batch study). In other words, the reality is that the volume ratio of organic phase/aqueous phase in the mixed phase is independent of feed rate ratio of organic phase/aqueous phase and is maintained in the state of setting, such as the volume ratio of both phases in the vessel. Therefore, it is difficult to believe that a difference in the effective volume ratio of organic phase/aqueous phase, which should be the same as the volume ratio of organic phase/aqueous phase in the mixed phase, would be caused by a difference in the mixing way, i.e., a difference in feed rate, while the volume ratio of organic phase/aqueous phase in the mixed phase remains the same in reality.

[0028]

For example, if the volume ratio of the organic phase/aqueous phase (the volume ratio of organic phase/aqueous phase in the mixed phase) in the phase-mixed region is set to 1/1, and the feed rate of the organic phase is increased while the feed rate of the aqueous phase remains the same, the extraction rate actually increases. This phenomenon was interpreted as a result of enhanced phase mixing and closer approach to chemical equilibrium. In other words, in the above, the effective volume ratio of organic phase/aqueous phase was considered to remain at 1/1. Therefore, in a system where phase mixing

and phase separation coexist within a certain range in the same reaction vessel, the relationship between the effective volume ratio of organic phase/aqueous phase and the feed rate ratio of organic phase/aqueous phase has never been considered so far.

[0029]

On the other hand, even if volume ratio of organic phase/aqueous phase is always constant in the mixed phase, if, for example, the feed rate of the organic phase to the aqueous phase increases, more of the organic phase will have the opportunity to come into contact with the aqueous phase at a frequency corresponding to that feed rate ratio. Therefore, while the volume ratio of organic phase/aqueous phase in the mixed phase is maintained constant, feed rate ratio of organic phase/aqueous phase can be considered to correspond to the effective volume ratio of organic phase/aqueous phase described above.

[0030]

The primary significance of this invention is that even in cases where the volume ratio of the organic phase/aqueous phase in the phase-mixed region (volume ratio of organic phase/aqueous phase in the mixed phase) is always maintained constant regardless of the feed rate of both phases, feed rate ratio of organic phase/aqueous phase corresponds to the effective volume ratio of organic phase/aqueous phase (the volume ratio of organic phase/aqueous phase in the above batch test). In fact, in a system where phase mixing and phase separation occur simultaneously in one reaction vessel, feed rate ratio of organic phase/aqueous phase corresponds to the effective volume ratio of organic phase/aqueous phase, even though the volume ratio of organic phase/aqueous phase is constant, and the extraction rate of rare earths changes according to this feed rate ratio. The results are shown in Example 1.

[Embodiment 1]

[0031]

Rare-earth extraction test in a single unit of droplet spraying type

[0032]

As the aqueous phase, an aqueous nitric acid solution at pH 2 containing Tb, Dy, Ho, Er, Tm, Yb, and Lu at a concentration of 100 ppb each, and as the organic phase, an organic solution of 35 mM (2-ethylhexyl) phosphonic acid 2-ethylhexyl (PC88-A) dissolved in a mixed solution of 90 vol% D70 (alkane solvent) and 10 vol% 2-ethyl hexanol were prepared and used for the following rare earth extraction tests. Rare earths extracted into the organic phase were back-extracted with 0.5 M nitric acid solution, and then their concentrations were measured by inductively coupled plasma optical emission spectrometry (ICP-OES).

[0033]

The extraction rate (%) of the above rare earth relative to feed rate ratio of organic phase/aqueous phase was determined by continuous flow testing using a droplet spraying unit and compared to the effective volume ratio of organic phase/aqueous phase extraction rate (%) of the rare earth obtained by batch testing. That is, we verified whether the change in feed rate ratio of organic phase/aqueous phase in the continuous flow test would correspond to a change in effective volume ratio of organic phase/aqueous phase in the batch test, and whether changing feed rate ratio of organic phase/aqueous phase would result in the same extraction rate (%) of rare earths as when the effective volume ratio of organic phase/aqueous phase was changed.

[0034]

First, in the continuous flow test, the feed rate of the organic phase was fixed at 30 mL/min, and the feed rate of the aqueous phase was varied to 7.5, 15, 30, 60, and 90 mL/min. In this case, feed rate ratio of organic phase/aqueous phase (= the feed rate of organic phase/the feed rate of aqueous phase) is 4, 2, 1, 1/2, and 1/3, respectively.

[0035]

In the continuous flow test, the volume ratio of organic phase/aqueous phase in the phase mixing area (mixed phase) was set to 1/1. In other words, the same volume of aqueous phase and organic phase were installed in the unit of a droplet spraying type. The volume ratio of organic phase/aqueous phase in the mixed phase can be set arbitrarily according to the volume ratio of the aqueous and organic phases to be installed.

[0036]

In batch tests, on the other hand, the effective volume ratio of organic phase/aqueous phase (the volume ratio of the organic phase/aqueous phase actually fed into the test tube) was set to 4, 2, 1, 1/2, and 1/3, as in the continuous flow test, and they were shaken sufficiently until the extraction reaction reached chemical equilibrium.

[0037]

In addition, in the continuous flow test, whenever feed rate ratio of organic phase/aqueous phase was changed, the mixed phase was sampled with a pipette to confirm that volume ratio of organic phase/aqueous phase had not changed from the set value of 1/1.

[0038]

FIG. 1(A) and FIG. 1(B) show a comparison of the results of the continuous flow test and the batch test. FIG. 1(A) shows the results for Tb, Dy, and Ho, while FIG. 1(B) shows the results for Er, Tm, Yb, and Lu.

[0039]

The extraction rate (%) of each rare earth in the continuous flow test is slightly smaller than that in the batch test because the extraction reaction does not reach chemical equilibrium. However, the relationship to the rare earth extraction rate (%) between feed rate ratio of organic phase/aqueous phase in the continuous flow test and the effective volume ratio of organic phase/aqueous phase in the batch test is in good agreement. That is, it was shown that, ideally, feed rate ratio of organic phase/aqueous phase can be equated with effective volume ratio of organic phase/aqueous phase.

[0040]

In normal continuous phase mixing (e.g., phase mixing in a mixer-settler), feed rate ratio of organic phase/aqueous phase, which can be regarded as the effective volume ratio of organic phase/aqueous phase, is uniquely equal to the volume ratio of organic phase/aqueous phase in the mixed phase. However, the results shown in Example 1 indicate that, according to the present method, the effective volume ratio of organic phase/aqueous phase can be freely changed by the feed rate ratio of organic phase/aqueous phase, while the volume ratio of organic phase/aqueous phase in the mixed phase can be set arbitrarily (e.g., 1/1).

[0041]

As described above, the present invention is based on a phenomenon that occurs in a mechanism in which phase mixing and phase separation occur simultaneously in a single reaction vessel. Therefore, examples of an apparatus are shown below, in which phase mixing and phase separation occur simultaneously in a single reaction vessel, but the present invention is not limited to this.

[0042]

In the Figures below, the distinction between heavy and light liquid phases is used rather than the distinction between aqueous and organic phases. This is because in the mechanism of the apparatus, it is important to know which liquid phase has greater specific gravity and is located lower down through the interface. In many cases, the organic phase is lighter than the aqueous phase, but conversely, there is also an organic phase that is heavier than the aqueous phase. In other words, the organic phase is not necessarily the lighter liquid phase and the aqueous phase is not necessarily the heavier liquid phase.

[0043]

There is a difference in the mechanism of phase separation between the mechanism of phase mixing by mechanical stirring (referred to as the mechanical stirring type) and the mechanism of phase mixing

by droplet spraying (referred to as the droplet spraying type).

[0044]

When phase mixing is done by mechanical stirring, phase separation can be encouraged by changing the flow produced by the rotation of the stirring blade. Examples of such apparatus are shown in FIGS. 2, 3, and 4, but are not limited to these.

[0045]

FIG. 2 shows a mechanism in which the impeller of the mechanical stirrer is positioned slightly below the vessel of the apparatus, the light liquid phase is pumped from above while being discharged from above, and at the same time, the heavy liquid phase is pumped from below while being discharged from below. FIG. 2 shows a apparatus for mechanical stirring type phase mixing, with two stirring flow blocking plates at the top and bottom. This apparatus is a unit that can be connected in series and can be assembled as a multi-stage apparatus with any number of stages (see FIG. 6).

[0046]

FIG. 3 shows a mechanism in which the arrangement of the stirring blades of the mechanical stirrer is the same as that in FIG. 2, and the light liquid phase is pumped from below while being discharged from above, and at the same time, the heavy liquid phase is pumped from below while being discharged from below. FIG. 3 shows a mechanically stirred phase mixing apparatus with two stirring flow blocking plates only at the bottom. As in FIG. 2, this apparatus is a unit that can be connected in series and can be assembled as a multi-stage apparatus with any number of stages (see FIG. 7).

[0047]

FIG. 4 shows a mechanism in which the arrangement of the stirring blades of the mechanical stirrer is the same as in FIGS. 2 and 3, and the light liquid phase is pumped from below while being discharged from above, and at the same time, the heavy liquid phase is pumped from below while being discharged from below. FIG. 4 shows a phase

mixing apparatus of mechanical stirring type with two stirring flow blocking plates, one at the top and one at the bottom. As in FIGS. 2 and 3, this apparatus is a unit that can be connected in series and can be assembled as a multi-stage apparatus with any number of stages (not shown).

[0048]

Hereinafter, as Example 2, an example of the distribution of phase-mixed regions and phase-separated regions coexisting in a mechanical stirring type unit will be shown.

[Embodiment 2]

[0049]

Distribution of phase-mixed and phase-separated regions in a unit of mechanical stirring.

[0050]

The apparatus in FIG. 2 was selected as a representative example, and the heavy liquid phase and the light liquid phase were placed at the same volume inside the apparatus. The distribution of the phase-mixed region and phase separation region when the apparatus was operated as described above is shown in FIG. 5. Water containing inorganic salts was used as the heavy liquid phase, and D70 (alkane-based solvent) was used as the light liquid phase. The stirring blades of the mechanical stirrer were also placed slightly below the liquid-liquid interface. At this time, it was found by actually sampling the mixed phase with a pipette that the light liquid phase/heavy liquid phase volume ratio in the mixed phase generated by mechanical stirring was always 1/1, regardless of the ratio of light liquid phase/heavy liquid phase feed rate. Note that if the position of the stirring blades of the mechanical stirrer is raised or lowered too much, the light liquid phase/heavy liquid phase volume ratio in the mixed phase will no longer be 1/1, so the stirring blades must be installed in an appropriate position.

[0051]

As shown in FIG. 5, in the apparatus shown in FIG. 2, the area of phase mixing occurred in a cross shape in the center of the apparatus vessel. Phase separation were developed in two areas of the bottom of the apparatus vessel (right side and left side) and three areas of the top of the apparatus vessel (right side, left side, and top center) due to the blockage of the stirred liquid flow generated by the rotation of the stirrer blades by the action of the stirring flow blocking plates, which were installed two at the top and two at the bottom. The ratio of the phase-mixed region to the phase-separated region varied slightly by changing the rotation of the stirrer blades, but the ratio of the two regions was not affected by the light liquid phase/heavy liquid phase feed rate ratio.

[0052]

FIGS. 2, 3, and 4 show mechanically stirred phase mixing apparatus, which are designed as a unit with a structure that can be easily made into multi-stage units. As examples of multi-stage apparatus, FIG. 6 shows a multi-stage (3-stage) apparatus using the unit of FIG. 2, and FIG. 7 shows a multi-stage (3-stage) apparatus using the unit of FIG. 3, but is not limited to the above.

[0053]

FIG. 6 shows a three-stage apparatus using the unit shown in FIG. 2 as a single unit. In this apparatus, each stage is separated by a stage partition plate. The top and bottom of the stage partition plates are communicated, so that the light liquid phase moves to the next stage from the upper communication port, and the heavy liquid phase moves to the next stage from the lower communication port.

[0054]

FIG. 7 shows a three-stage apparatus using the unit shown in FIG. 3. In this apparatus, as in FIG. 6, each stage is separated by a stage partition plate. In FIG. 6, the upper and lower stages of the stage partition plates are connected, but in FIG. 7 the upper stages are not connected, and the light liquid phase moves to the next stage by pumping rather than through a communication port. On the other hand,

the heavy liquid phase moves to the next stage from the lower communication port, as in FIG. 6.

[0055]

Next, as Example 3, an example of the distribution of phase-mixed regions and phase-separated regions coexisting in a mechanical stirring type multi-stage apparatus will be shown.

[Embodiment 3]

[0056]

Distribution of phase-mixed and phase-separated regions in a mechanical stirring type multi-stage apparatus

[0057]

As one example of a multi-stage apparatus with mechanical stirring, FIG. 8 shows the distribution of the phase mixing and phase separation regions when the heavy liquid phase (water containing inorganic salts) and light liquid phase (D70) are installed in the same volume in the three-stage apparatus shown in FIG. 7. The stirring blades of the mechanical stirrer were installed slightly below the liquid-liquid interface in all stages. In the multi-stage (3-stage) apparatus, as in the unit apparatus, the light liquid phase to heavy liquid phase volume ratio in the mixed phase was always 1/1 in all stages, regardless of the light liquid phase to heavy liquid phase feed rate ratio (Actual pipetting of the mixed phase).

[0058]

As shown in FIG. 8, in the apparatus of FIG. 7, phase-mixed regions occurred in the form of T-shaped configurations connected in series. The two stirring flow blocking plates installed under each stage blocked the stirred liquid flow caused by the rotation of the stirring blades, resulting in two phase-separated regions (on the right and left) at the bottom of each stage. At the connection points of each stage, the left and right phase separation regions (both are heavy liquid phase separation regions) are connected via communication ports provided

below the stage partition plates. On the other hand, although no stirring flow blocking plate was installed above the apparatus, the light liquid phase was easily separated when pumped from below.

[0059]

Next, an example of an apparatus in which phase mixing is achieved by droplet spraying is shown. In the droplet spraying type, phase separation can be promoted by increasing the cross-sectional area of the part through which the flow generated by droplet spraying passes. An example of such an apparatus is shown in FIG. 9, but is not limited to this.

[0060]

FIG. 9 shows a mechanism in which the light liquid phase is pumped in from below through a droplet spraying nozzle while being discharged from above, and at the same time the heavy liquid phase is pumped in from below while being discharged from below. FIG. 9 shows a droplet spraying type phase mixing apparatus with one flow cross-sectional area changing plate arranged above and one below. Note that this apparatus is a unit that can be connected in series, and can be assembled into a multi-stage apparatus with any number of stages (see FIG. 11).

[0061]

Hereinafter, as Example 4, an example of the distribution of phase-mixed regions and phase-separated regions coexisting in a droplet spraying type unit will be described.

[Embodiment 4]

[0062]

Distribution of phase-mixed and phase-separated regions in a droplet spraying type unit

[0063]

FIG. 10 shows the results of investigating the distribution of phase mixing and phase separation regions when equal volumes of

heavy liquid phase (water containing inorganic salts) and light liquid phase (D70) were placed in the droplet spraying type unit shown in FIG. 9 and operated as described above. In this case, it was found by actually sampling the mixed phase with a pipette that the light liquid phase/heavy liquid phase volume ratio in the mixed phase generated by droplet spraying was always 1/1, regardless of the light liquid phase/heavy liquid phase feed rate ratio.

[0064]

As shown in Fig. 10, the apparatus in Fig. 9 can develop regions of phase mixing over a wide area. The mixed phase generated by droplet spraying can be phase separated by increasing the cross-sectional area of the region through which the flow passes. The regions of phase separation can appear on either the left or right side of the top and bottom of the vessel. The ratio of the phase-mixed region to the phase-separated region hardly changed when the feed rate of the heavy liquid phase, light liquid phase, or both was changed, as long as the feed rate was within the appropriate range (i.e., the feed rate was not too fast for the size of the apparatus). Similarly, the ratio of the two regions was hardly affected by the ratio of the light liquid phase/heavy feed rate as long as the feed rate was within the appropriate range.

[0065]

FIG. 9 shows an apparatus for phase mixing by droplet spraying, but it is designed as a unit with a structure that can be easily multi-staged. As an example of the multi-stage system, a multi-stage (3-stage) apparatus in which the apparatus of FIG. 9 is used as a unit is shown in FIG. 11, but is not limited to this.

[0066]

FIG. 11 shows a three-stage apparatus using the apparatus of FIG. 9 as a unit, where each stage is partitioned by a stage partition plate, but the top of the partition plate is not communicated, and only the bottom partition is communicated. In other words, the light liquid phase does not pass through the communication port and is transferred to the

next stage by pumping, while the heavy liquid phase is transferred to the next stage from the lower communication port of the partition plate.

[0067]

Hereinafter, as Example 5, an example of the distribution of phase-mixed regions and phase-separated regions coexisting in a droplet spraying type multi-stage apparatus will be described.

[Embodiment 5]

[0068]

Distribution of phase-mixed and phase-separated regions in a droplet spraying type multi-stage apparatus

[0069]

The droplet spraying type multi-stage apparatus shown in FIG. 11 will be used as an example. In the multi-stage (3-stage) apparatus shown in FIG. 11, the distribution of phase mixing and phase separation regions are shown in FIG. 12 when the heavy liquid phase (water containing inorganic salts) and light liquid phase (D70) are installed in the same volume in the apparatus. In the multi-stage (3-stage) apparatus, as in the single-unit apparatus, the volume ratio of the light liquid phase/heavy liquid phase in the mixed phase was always 1/1 in all stages, regardless of the ratio of the light liquid phase/heavy liquid phase flow rate (The mixed phase was actually collected with a pipette).

[0070]

As shown in FIG. 12, it was found that even in a multi-stage apparatus, a phase-mixed region can be generated over a wide range, as in a single unit. The mixed phase generated by droplet spraying in each stage can be phase-separated by increasing the cross-sectional area of the part through which the flow passes, and the phase-separated area appears on either the left or right side of the upper or lower part of each stage. The separated heavy liquid phase moves to the next stage through a communication port provided at the bottom of the stage partition plate that separates each stage, while the light liquid phase

moves to the next stage by pumping, not through a communication port.

[Industrial Applicability]

[0071]

This invention relates to a new method for mixing two liquid phases and an apparatus (particularly a multi-stage apparatus) that utilizes the method, and is expected to be used in a wide range of industrial fields, such as solvent extraction and oil-water separation. For example, it is expected to be used in key industries such as the metal industry and chemical industry. In particular, it is expected to be used in the manufacture of high-tech parts that require rare metals, such as lithium-ion batteries and neodymium magnets, and in a separation and purification technology in rare metal recycling.

[Illustration of sign]

[0072]

1: Stirring flow blocking plate

2: Flow cross-sectional area changing plate

3: Mechanical stirrer

4: Stage partition plate

5: Liquid droplet spraying nozzle

What Is Clamed Is:

1. A phase mixing method, wherein

by allowing a phase mixing region and a phase separation region to coexist within a certain range within the reaction vessel, in an operation in which two liquid phases are fed into a single reaction vessel and phase mixing is performed within the reaction vessel, the light liquid phase/heavy liquid phase volume ratio in the mixed phase in which both phases participate in the phase mixing in the phase mixing region can be set freely and completely independent of the light liquid phase/heavy liquid phase feed rate ratio which is ideally equated with the effective light liquid phase/heavy liquid phase volume ratio, which is a parameter in a function of chemical equilibrium for extraction rate and back extraction rate.

2. The phase mixing method according to claim 1, characterized in that the phase mixing is performed by mechanical stirring or droplet spraying.

3. A phase mixing unit connectable in series, which has a mechanism for allowing a phase mixing region and a phase separation region to coexist within a certain range within the reaction vessel in an operation of performing phase mixing while feeding two liquid phases into a single reaction vessel, wherein

said unit comprises one reaction vessel for performing phase mixing of two liquid phases, and a means for feeding the two liquid phases into the reaction vessel and performing phase mixing using mechanical stirring or droplet spraying.

4. A multi-stage phase mixing apparatus characterized in that the unit recited in claim 3 are connected in series to form a multi-stage configuration in which similar operation is repeatedly performed.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP2023/001154

A. CLASSIFICATION OF SUBJECT MATTER

B01D 11/04(2006.01)i
FI: B01D11/04 A

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

B01D11/04

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Published examined utility model applications of Japan 1922-1996
Published unexamined utility model applications of Japan 1971-2023
Registered utility model specifications of Japan 1996-2023
Published registered utility model applications of Japan 1994-2023

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

JSTPlus/JSTChina/JST7580 (JDreamIII)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP 2021-94546 A (JAPAN ATOMIC ENERGY AGCY.) 24 June 2021 (2021-06-24) claims, paragraphs [0022]-[0025], [0027]-[0032], [0104], [0105], [0129], [0130], fig. 1(p)-(t), 2(p)-(t)	1-2
Y		2-4
Y	JP 2016-123907 A (JAPAN ATOMIC ENERGY AGCY.) 11 July 2016 (2016-07-11) paragraph [0005], fig. 1	2-4
X	JP 2013-144270 A (SHIN-ETSU CHEMICAL CO., LTD.) 25 July 2013 (2013-07-25) claims, paragraphs [0020]-[0026], [0031], fig. 1	3-4
Y		3-4

☐ Further documents are listed in the continuation of Box C. ☑ See patent family annex.

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
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"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
13 February 2023	**28 February 2023**
Name and mailing address of the ISA/JP **Japan Patent Office (ISA/JP)** **3-4-3 Kasumigaseki, Chiyoda-ku, Tokyo 100-8915** **Japan**	Authorized officer Telephone No.

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.
PCT/JP2023/001154

Patent document cited in search report	Publication date (day/month/year)	Patent family member(s)	Publication date (day/month/year)
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