May 4, 1937. M. KLAR ET AL 2,079,511

PROCESS FOR THE CONCENTRATION OF ACETIC ACID

Filed April 20, 1931 2 Sheets-Sheet 1

Fig. 1.

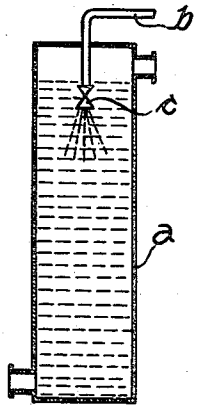


Fig. 2.

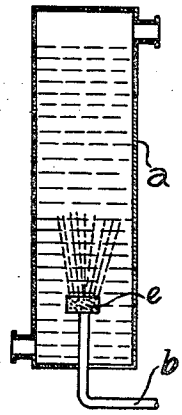


Fig. 3.

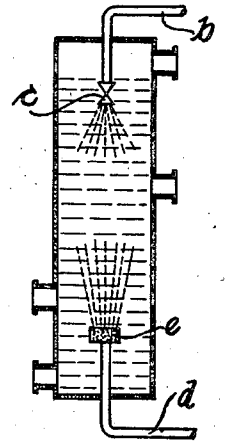
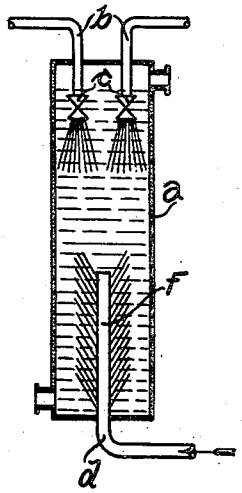


Fig. 4.



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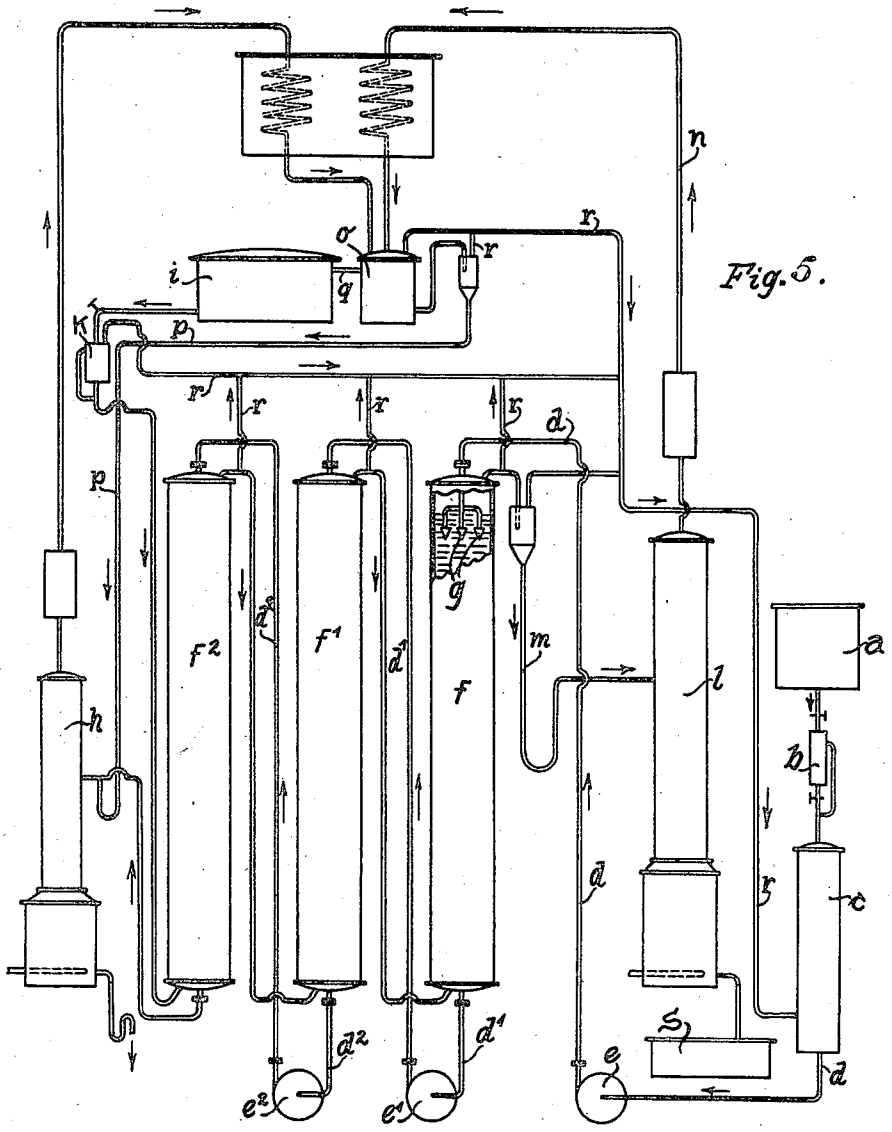


Fig. 5.

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PROCESS FOR THE CONCENTRATION OF ACETIC ACID

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In Germany May 16, 1930

13 Claims. (Cl. 260—122)

This invention relates to a process for the concentration of acetic acid from aqueous solutions, also salt solutions, and the like, by extraction with suitable liquids, such as acetic acid ethyl ester, etc.

According to the invention a surprising and specially favourable effect is obtained in the known extraction of acetic acid from aqueous solutions or the like, with suitable liquids, such as, for instance, acetic acid ethyl ester, by extremely fine distribution or atomization of the one liquid within the other and preventing the atomized particles to coalesce into larger particles before they have an opportunity to react with the liquid in which they are introduced. The known high-power-, cloud-, dust-, spray- and atomizing nozzles are particularly suitable for this. Indeed those atomizers are to be preferred, which, for instance, at about 1–4 at. make an atomization of liquids in air and gases possible to drops of about 0.11–0.08 mm. in size.

The drawings illustrate diagrammatically both the separate adaptation of the distributing devices of the type described and the most varied combinations thereof, wherein not only the single extractors, but also an integrally co-operating system of the same is used.

In the drawings:—

Figure 1 shows an extraction liquid container *a* of any desired height with an inlet pipe *b*, which ends in an atomizer *c*, introduced at any place desired in the inner space, in the present case from above,

Figure 2 shows a similar extraction tower, i. e. a container *a* filled with extraction liquid; in the lower part are disposed an inlet pipe *b*, and an atomizing nozzle *e* as in Figure 1,

Figure 3 shows a combination of the forms of construction according to Figures 1 and 2,

Figure 4 shows by way of example a combined device, in which atomizing nozzles are provided in the upper portion of the container *a*, and, in the lower portion of the same, a perforated or porous pipe *f*, and

Figure 5 shows diagrammatically a complete apparatus for carrying out the process according to the invention.

Referring to Figure 4, the nozzles *c* and the pipe *f* may be of any desired size and of any desired number or position or arrangement.

These partly individual, partly combined forms of apparatus can be used for the concentration of dilute acetic acid in the following manner:

The procedure is such that, for example, aqueous acetic acid, is atomized through the pipe *b* and the nozzle *c* into the extraction chamber as shown in Figure 1 which is filled with acetic acid ethyl ester, to a point above the nozzle. The acetic acid solution, which sinks down through the ester column in an unusually fine state of division, is rapidly extracted at the relatively large distribution surface.

The process can naturally also be reversed, whereby the acetic ester is atomized in a column of aqueous acetic acid by means of the pipe *b* and the atomizing nozzle *e* disposed below in the extraction chamber according to Figure 2.

When using the apparatus according to Fig. 3, which is provided, at the top, with one or more pipes *b* and atomizing nozzles *c* and, at the bottom, with a pipe *d* and a nozzle *e* or a perforated or porous tube or other liquid-dispersing devices, a combined process can be carried out in which two layers of liquid are formed by suitable regulation of the inflow and outflow of the two liquids to be distributed; for example, an upper one of acetic acid ethyl ester and a lower one of aqueous acetic acid. If now dilute acetic acid is introduced into the column from above through the pipe *b*, and acetic acid ester through the lower distributing device *e*, then by means of the double sided distribution an extracting action in counterflow will result.

The method described hereinabove can be carried out just as well in the apparatus shown in Figure 4, in which the perforated tube connected to the pipe *d* is arranged in the lower part of the container *a*.

In passing in dilute acetic acid, for example from above, and the extraction material, e. g. the acetic acid ethyl ester from below into an extraction chamber, it is advantageous to regulate the upper and lower inflow of the liquids being mixed together, so that two separate layers thereof are formed in the extraction chamber. In the upper portion of the container for the extraction liquid, dilute acetic acid is atomized into a liquid column of acetic acid ethyl ester and, in the lower portion of the container for the extraction liquid acetic acid ethyl ester is atomized into a liquid column of dilute acetic acid. By this counter-current action, and the fine state of division of the one liquid in the other, an intensive extraction results.

It is important that when carrying out the process the components of the mixture in the liquid columns formed should always be in counterstream, and that the mixing process should take place in a hollow space of the container, which has no partition walls, filling bodies or

other obstructions, so as to offer as little hindrance as possible to the free flow of the droplets.

The single extractors may either be located one above the other, or next to one another in rows, and so that they can be connected one behind the other by means of pipe connections, as illustrated in Fig. 5, which illustrates preferred apparatus for practicing the method of the present invention.

Here, the dilute acetic acid passes from a supply tank *a*, arranged at a suitable height, through a measuring and regulating device *b* into a scrubber *c*, which is filled with suitable fillers, for instance porcelain rings, so called Raschig-rings or the like.

From the lower part of this scrubber *c* the diluted acid is sucked through a pump *e* by means of the pipe *d* and led into the upper portion of an extractor *f*, which is partly broken away in the figure, and is atomized therein by means of one or more rotary power nozzles *g* under an excess pressure of 1–3 atmospheres. This atomization takes place in the chamber *f* in the extraction medium, with which the extractor *f* is filled to a point above the nozzles *g*.

As the small droplets travel downward, from the upper portion to the lower portion of the chamber *f*, the dilute acetic acid becomes considerably weakened, and the weakened acetic acid collects in a layer at the bottom of the chamber *f*. The weakened liquid may be passed out of an opening at the bottom of the chamber *f* and the acetic acid ethyl ester, containing acetic acid, out of an opening at the top of the chamber *f*. The separation of the acetic acid may be further brought about by decantation and distillation.

The weakened, dilute, acetic-acid layer collected at the bottom of the chamber *f* is now forced, by a pump *e'*, through a pipe *d'* and further nozzles (not shown), into a second extraction chamber *f'*, which may be of the same type, and which is also filled with an extraction liquid, in the same manner as before described. The atomized acetic acid thus forced into the extraction liquid in the chamber *f'* likewise travels downward therein to form a layer of still further weakened dilute acetic acid at the bottom of this chamber *f'*.

A pump e^2 forces this still further weakened acetic acid, through a pipe d^2 and additional nozzles (not shown), into a third extraction chamber f^2, in the same manner as already described.

A three step extraction with extractors of about 10 m. in height is sufficient for all dilute acids ordinarily met with, and make it possible to obtain about 95–98% of the absolute acid value introduced. Any height and number of extractors, if structural circumstances will allow it, can be chosen, which make the effect more favourable.

The exhausted waste water with a content of about 0.4–0.5% acetic acid and about 8–9% acetic ester passes out from the lower portion of the last extractor f^2 into a spirit extraction column *h* in which the acetic ester is driven off continuously by direct steam.

The acetic acid ethyl ester, running in countercurrent to the dilute acid, passes continuously out of the supply container *i*, through a regulating device *k*, and into the lower portion of the extractor f^2. After rising up through the chamber f^2, it passes again from the top downwards, and into the bottom of the extraction chamber *f'*. After leaving the top of the extraction chamber *f'*, it pursues a similar course through the extraction chamber *f*, finally leaving the extraction apparatus at the upper part of the extractor *f* laden with acetic acid, through the pipe *m* to the concentration column *l*, in which the acetic acid is freed completely from ethyl ester and water by indirect steam.

The acetic acid ethyl ester and the water form a constant boiling mixture and leave the column continuously as a distillate of about 93% ester and 7% water, while the water free acetic acid is continuously drawn off into a container *s* as concentrate.

The distillates pass through the pipe *n* into a common separator *o*, which separates the excess water and returns it through a gooseneck and the pipe *p* back again into the spirit extraction column *h*, while the acetic ethyl ester runs back, by means of the connecting pipe *q* into the supply tank *i*, and from there starts its circuit again.

All the air removed from the several parts *f*, f^1, f^2, *k*, *o*, etc. of the apparatus is returned to the scrubber *c* through the collecting pipe system *r*.

Instead of acetic acid ethyl ester, other extraction liquids as, i. e., acetic-acid absorbing liquids may be employed in connection with the devices shown in Figs. 1, 3, 4 and 5, if the specific gravity of said liquids is less than the specific gravity of water. Extraction liquids having a greater specific gravity than water may, however, be used, if they are introduced into the upper part of the extracting column and if the dilute acetic acid is introduced into the lower part of said column by means of a nozzle *e* producing a very fine atomization, as shown in Fig. 2.

An important feature of the invention resides in the fact that at least one of the liquids, either the dilute acetic acid or the extracting liquid, is atomized by a nozzle or nozzles adapted to produce droplets in the region of 0.1 millimeter or less in air. Both said liquids may be atomized by such a nozzle or nozzles. Preferably, however, the dilute acetic acid is atomized by the above-referred to nozzle or nozzles, and the extracting liquid, such as acetic acid ethyl ester, may be atomized by other means, as perforated tubes, porous-candle devices or the like, producing a coarser atomization.

The path of the liquid in the above-referred-to rotary, high-power atomizing nozzles is spiral. By the time that the liquid particles reach the end of the spiral path, the centrifugal force becomes so great that the liquid is atomized.

Modifications will occur to persons skilled in the art and all such are considered to fall within the spirit and scope of the invention, as defined in the appended claims.

What is claimed is:—

1. Process for the concentration of acetic acid contained in liquid solutions by extraction with a suitable extraction liquid, comprising the steps in combination of introducing the acetic acid solution and the extraction liquid into the upper and lower portions respectively of two superimposed layers consisting of the extraction liquid and the acetic acid solution, the position of the two layers corresponding to the specific gravities of the acetic acid solution and the extraction liquid and being such that the point of introduction of the acetic acid solution is in the extraction liquid layer and the point of introduction of the extraction liquid is in the acetic acid layer, at least one of the liquids being introduced by atomization which in air would yield droplets of a size

in the region of 0.11 to 0.08 mm., and maintaining the layers of the extraction liquid and the acetic acid solution free of flow-affecting bodies in the region substantially between the said two points of introduction of the extraction liquid and the acetic acid solution so as to offer as little hindrance as possible to the free flow of the droplets.

2. Process for the concentration of acetic acid contained in liquid solutions by extraction with a suitable extraction liquid comprising the steps in combination of introducing the acetic acid solution and the extraction liquid into the upper and lower portions respectively of two superimposed layers consisting of the extraction liquid and the acetic acid solution, the position of the two layers corresponding to the specific gravities of the acetic acid solution and the extraction liquid and being such that the point of introduction of the acetic acid solution is in the extraction liquid layer and the point of introduction of the extraction liquid is in the acetic acid layer, at least one of the liquids being introduced by atomization which in air would yield droplets of a size of about 0.1 mm., and maintaining the layers of the extraction liquid and the acetic acid solution free of flow-affecting bodies in the region substantially between the said two points of introduction of the extraction liquid and the acetic acid solution so as to offer as little hindrance as possible to the free flow of the droplets.

3. Process for the concentration of acetic acid contained in liquid solutions by extraction with a suitable extraction liquid, comprising the steps in combination of introducing the acetic acid solution and the extraction liquid into the upper and lower portions respectively of two superimposed layers consisting of the extraction liquid and the acetic acid solution, the position of the two layers corresponding to the specific gravities of the acetic acid solution and the extraction liquid and being such that the point of introduction of the acetic acid solution is in the extraction liquid layer and the point of introduction of the extraction liquid is in the acetic acid layer, at least one of the liquids being introduced by atomization which when employing a pressure between 1 and 4 atmospheres would yield in air droplets of a size between 0.11 and 0.08 mm., and maintaining the layers of the extraction liquid and the acetic acid solution free of flow-affecting bodies in the region substantially between the said two points of introduction of the extraction liquid and the acetic acid solution so as to offer as little hindrance as possible to the free flow of the droplets.

4. Process for the concentration of acetic acid contained in liquid solutions by extraction with acetic acid ethyl ester, comprising the steps in combination of introducing the acetic acid solution and the acetic acid ethyl ester into the upper and lower portions respectively of two superimposed layers of acetic acid ethyl ester as the top layer and acetic acid solution as the lower layer, the acetic acid solution being introduced by atomization which in air would yield droplets of a size in the region of 0.11 to 0.08 mm., and maintaining the layers of the acetic acid ethyl ester and the acetic acid solution free of flow-affecting bodies in the region substantially between the points of introduction of the acetic acid ethyl ester in the layer of the acetic acid solution and the point of introduction of the acetic acid solution in the layer of acetic acid ethyl ester so as to offer as little hindrance as possible to the free flow of the droplets.

5. Process for the concentration of acetic acid contained in liquid solutions by extraction with acetic acid ethyl ester, comprising the steps in combination of introducing the acetic acid solution and the acetic acid ethyl ester into the upper and lower portions respectively of two superimposed layers of acetic acid ethyl ester as the top layer and acetic acid solution as the lower layer, the acetic acid solution being introduced by atomization which in air would yield droplets of a size of about 0.1 mm., and maintaining the layers of the acetic acid ethyl ester and the acetic acid solution free of flow-affecting bodies in the region substantially between the points of introduction of the acetic acid ethyl ester in the layer of the acetic acid solution and the point of introduction of the acetic acid solution in the layer of acetic acid ethyl ester so as to offer as little hindrance as possible to the free flow of the droplets.

6. Process for the concentration of acetic acid contained in liquid solutions by extraction with acetic acid ethyl ester, comprising the steps in combination of introducing the acetic acid solution and the acetic acid ethyl ester into the upper and lower portions respectively of two superimposed layers of acetic acid ethyl ester as the top layer and acetic acid solution as the lower layer, the acetic acid solution being introduced by atomization which when employing a pressure between 1 and 4 atmospheres would yield in air droplets of a size between 0.11 and 0.08 mm., and maintaining the layers of the acetic acid ethyl ester and the acetic acid solution free of flow-affecting bodies in the region substantially between the points of introduction of the acetic acid ethyl ester in the layer of the acetic acid solution and the point of introduction of the acetic acid solution in the layer of acetic acid ethyl ester so as to offer as little hindrance as possible to the free flow of the droplets.

7. A process for concentrating acetic acid, comprising introducing, into the lower portion of a column of liquid consisting of an upper layer of an acetic acid absorbing liquid and a lower layer of dilute acetic acid solution, a flow of absorbing liquid, introducing into the upper portion of the liquid column within the upper layer a flow of dilute acetic acid, atomizing at least one of the liquid flows under conditions which in air would yield droplets of a size of 0.11 to 0.08 mm., and maintaining the column of liquid free of flow-affecting bodies in the region substantially between the point of introduction of the absorbing liquid in the lower portion of the column and the point of introduction of the acetic acid in the upper portion of the column so as to offer as little hindrance as possible to the free flow of the droplets.

8. A process for concentrating acetic acid, comprising introducing, into the lower portion of a column of liquid consisting of an upper layer of an acetic acid absorbing liquid and a lower layer of dilute acetic acid solution, a flow of absorbing liquid, introducing into the upper portion of the liquid column within the upper layer a flow of dilute acetic acid, atomizing at least one of the liquid flows under conditions which in air would yield droplets of a size of about 0.1 mm., and maintaining the column of liquid free of flow-affecting bodies in the region substantially between the point of introduction of the absorbing liquid in the lower portion of the column and the point

of introduction of the acetic acid in the upper portion of the column so as to offer as little hindrance as possible to the free flow of the droplets.

9. A process for concentrating acetic acid, comprising introducing, into the lower portion of a column of liquid consisting of an upper layer of an acetic acid absorbing liquid and a lower layer of dilute acetic acid solution, a flow of absorbing liquid, introducing into the upper portion of the liquid column within the upper layer a flow of dilute acetic acid, atomizing at least one of the liquid flows under conditions which when employing a pressure between 1 and 4 atmospheres would yield in air droplets of a size of 0.11 to 0.08 mm., and maintaining the column of liquid free of flow-affecting bodies in the region substantially between the point of introduction of the absorbing liquid in the lower portion of the column and the point of introduction of the acetic acid in the upper portion of the column so as to offer as little hindrance as possible to the free flow of the droplets.

10. A process for concentrating acetic acid comprising introducing, into the lower portion of a column of liquid consisting of an upper layer of acetic acid ethyl ester and a lower layer of dilute acetic acid solution, a flow of acetic acid ethyl ester, introducing into the upper portion of the liquid column within the upper layer a flow of dilute acetic acid, atomizing at least one of the liquid flows under conditions which in air would yield droplets of a size of 0.11 to 0.08 mm., and maintaining the column of liquid free of flow-affecting bodies in the region substantially between the point of introduction of the acetic acid ethyl ester in the lower portion of the column and the point of introduction of the acetic acid in the upper portion of the column so as to offer as little hindrance as possible to the free flow of the droplets.

11. A process for concentrating acetic acid, comprising introducing, into the lower portion of a column of liquid consisting of an upper layer of acetic acid ethyl ester and a lower layer of dilute acetic acid solution, a flow of acetic acid ethyl ester, introducing into the upper portion of the liquid column within the upper layer a flow of dilute acetic acid, atomizing at least one of the liquid flows under conditions which in air would yield droplets of a size of about 0.1 mm., and maintaining the column of liquid free of flow-affecting bodies in the region substantially between the point of introduction of the acetic acid ethyl ester in the lower portion of the column and the point of introduction of the acetic acid in the upper portion of the column so as to offer as little hindrance as possible to the free flow of the droplets.

12. A process for concentrating acetic acid, comprising introducing, into the lower portion of a column of liquid consisting of an upper layer of acetic acid ethyl ester and a lower layer of dilute acetic acid solution, a flow of acetic acid ethyl ester, introducing into the upper portion of the liquid column within the upper layer a flow of dilute acetic acid, atomizing at least one of the liquid flows under conditions which when employing a pressure between 1 and 4 atmospheres would yield in air droplets of a size of 0.11 to 0.08 mm., and maintaining the column of liquid free of flow-affecting bodies in the region substantially between the point of introduction of the acetic acid ethyl ester in the lower portion of the column and the point of introduction of the acetic acid in the upper portion of the column so as to offer as little hindrance as possible to the free flow of the droplets.

13. Process for the concentration of acetic acid contained in liquid solutions by extraction with a suitable extraction liquid, comprising the steps in combination, of introducing the acetic acid solution into a layer of extraction liquid and introducing the extraction liquid into a layer of acetic acid solution, said layers being superposed in positions corresponding to the specific gravities of the acetic acid solution and the extraction liquid, the acetic acid solution being introduced by atomization which in air would yield droplets substantially of a size in the region of 0.11 to 0.08 mm., the extraction liquid being introduced in a coarser atomization than the acetic acid solution, and maintaining the layer of extraction liquid free of flow-affecting bodies in the region substantially between the point of introduction of the acetic acid in the layer of extraction liquid and the surface of the acetic acid layer so as to offer as little hindrance as possible to the free flow of the droplets of acetic acid within the layer of extraction liquid.

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