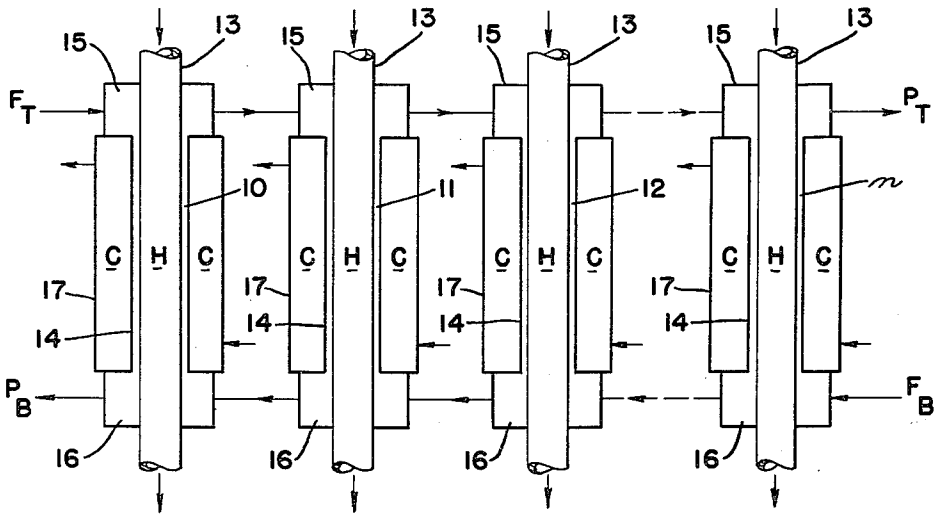
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THERMAL DIFFUSION METHOD

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THERMAL DIFFUSION METHOD

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This invention relates to an improved continuous method for separating materials by liquid thermal diffusion.

It has been known for some time that it is possible to separate by liquid thermal diffusion materials that are incapable of separation by any other known methods or that are separated by other methods with only great difficulty.

Succinctly stated, the process of liquid thermal diffusion consists of confining a liquid mixture, a term intended herein to include mixtures that are liquid under conditions of operation, in a narrow separation chamber and imposing a temperature gradient across the chamber. This is usually accomplished by confining the liquid mixture in a vessel or column having two closely spaced parallel or concentric walls, one of which is maintained at a substantially higher temperature than the other. The walls are vertical or have vertical components and thermal circulation of the contents of the chamber is therefore set up by virtue of the differences in density adjacent the relatively heated and cooled walls. One of the dissimilar fractions having a higher than initial concentration of one component accumulates adjacent the hot wall. Another fraction having a higher than initial concentration of a different component preferentially accumulates adjacent the cold wall. The fraction adjacent the hot wall ascends in the chamber as the top product. The fraction adjacent the cold wall descends in the chamber as the bottom product.

It has been found that in order to obtain satisfactory rates of separation by thermal diffusion, the opposed thermal diffusion chamber-forming surfaces of the hot and cold wall should be spaced apart no more than about ½ inch, spacings of the order of 0.15 inch or less, and preferably about 0.02 to 0.06 inch, being most effective. It will readily be seen that due to the small spacings involved, considerable difficulty is encountered in obtaining column efficiency which may be defined as the product of the degree of separation between the feed mixture and the desired product and the production rate of the desired product. Any improvement in either or both of these factors would be highly advantageous.

It has already been found that the degree of separation, particularly at relatively high feed rates, can be significantly improved over that which is obtainable with static thermal diffusion methods by employing a continuous "mixed-end feed system" for thermal diffusion. Such a thermal diffusion method and apparatus in which such method may be employed is disclosed in Patent No. 2,827,171 which is assigned to our assignee. The thermal diffusion unit disclosed therein offers excellent separation efficiency by providing upper and lower reservoirs communicating with the upper and lower ends of a vertical separation chamber. These reservoirs have sufficient volume to permit mixing of fresh feed with liquids entering the reservoirs from the chamber, as well as preventing a turn-about of the liquid within the separation chamber before it reaches the reservoir; i.e., the liquid ascending the hot wall, for example, does not start down the cold wall but rather is mixed with the feed in the reservoir and the mixtures start down the cold wall. The same is true at the bottom. Generally, these effects are obtained when

the reservoirs are at least about twice as wide and twice as high or deep as the spacing between the walls forming the separation chamber. In conducting this mixed-end feed system, the feed liquid mixture is continuously introduced into the upper and lower reservoirs and is thereby admixed in the reservoirs with the ascending and descending fractions; a portion of the liquid enters the separation chamber to replace the fractions entering the reservoirs; and the remainder, enriched with the components in the dissimilar fractions, is withdrawn from the upper and lower reservoirs, respectively.

The present invention offers an improvement over such a mixed-end feed method of thermal diffusion by increasing the throughput of material or rate of flow of liquid across one end of the apparatus over the throughput or rate of flow of liquid across the other end. Although any ratio of throughput rates at the opposite ends of the apparatus greater than 1:1 is operable for the present invention to improve separation efficiency, it has been found that ratios from 2:1 to 20:1 are most effective and the preferred range is from 2:1 to 10:1. There is usually no economic advantage to be gained by increasing the ratio over 10:1 since the improvement gradually approaches but never reaches a limited value.

It is to be understood that the invention is contemplated for use with a mixed-end feed system to either a single thermal diffusion chamber such as is disclosed in Patent No. 2,827,171 or to a series of thermal diffusion chambers such as disclosed in Patent No. 2,827,172 wherein the flow is across the individual chambers and the flow at the top of the chambers may be either countercurrent or concurrent in relation to the flow at the bottom of the chambers. More detailed information relating to the apparatus and the method of these systems may be obtained from the foregoing cited references, and it is intended here to include such disclosure from these patents as may be necessary for a complete and clear understanding of the invention.

Several important advantages are to be gained over the conventional mixed-end feed system by the improvement of the present invention. It has been found, for example, that a higher quality product may be produced at the same rate. Furthermore, the rate at which a given quality product is produced may be increased. It has also been found that for any given level of througput to a chamber, a higher quality product may be produced by increasing the throughput at one end in relation to the other. Moreover, from an operational viewpoint, the method of the invention provides for better separation of viscous materials since this method inherently decreases the likelihood of thermal siphoning and bypassing.

A better understanding of the invention will be gained from the following discussion taken together with the accompanying patent drawing which is a schematic illustration of a preferred embodiment for practicing the present invention utilizing the apparatus shown in Patent No. 2,827,172. In the drawing, annular thermal diffusion chambers **10**, **11**, **12**, and *n*, are formed by an outer surface of inner tube **13** and an inner surface of outer tube **14**. Suitable means not forming a part of this invention are provided for relatively heating and cooling the chamber walls **13** and **14**, as indicated by the symbols "H" and "C" representing the hot and cold walls, respectively. Hence, tube **13** may be relatively heated by passing steam therethrough as indicated by the arrows, and outer tube **14** may be relatively cooled by circulating water through a cooling jacket **17** in the manner indicated by the arrows. The symbols F_T and F_B represent top feed liquid and bottom feed liquid, respectively, and the symbols P_T and P_B represent top product and bottom product,

respectively. The direction of flow across the series of separation chambers is indicated by the arrows, and flow across the top is countercurrent to flow across the bottom of the chambers.

In operation, top feed liquid F_T is continuously introduced into upper reservoir **15** of separation chamber **10** and bottom feed liquid F_B is continuously introduced into lower reservoir **16** of separation chamber *n*, while the inner and outer tubes forming the annular separation chambers **10**, **11**, **12**, and *n* are relatively heated and cooled. The fractions that accumulate adjacent the hot walls ascend to the upper reservoirs, become mixed with the feed therein, and are transferred to the next upper reservoir in the series until the feed enriched in that particular fraction P_T is withdrawn from the upper reservoir **15** of separation chamber *n* at a rate equivalent to F_T. The fractions that accumulate adjacent the cold walls descend into the lower reservoirs and likewise become mixed with the liquid therein. This liquid, enriched with the fraction descending into the lower reservoir, is passed into the next lower reservoir in the series until finally liquid that has been enriched with the cold wall fraction P_B is withdrawn from the lower reservoir **16** of separation chamber **10** at a rate equivalent to F_B. In accordance with the invention, the flow rates across the top and bottom are unequal and vary from 2 to 20, preferably from 2 to 10. Expressed mathematically:

$$\frac{F_T \doteq P_T}{F_B \doteq P_B} = \frac{1}{20} \text{ to } \frac{1}{2} \text{ and from 2 to 20}$$

The advantages offered by the method of the present invention will become further apparent from the discussion of results obtained from a series of tests conducted with a 4-column thermal diffusion unit operated on a countercurrent mixed-end feed system as schematically illustrated in the patent drawing. Each of the columns was six feet in height, having an equidistant annular separation chamber of 0.032 inch, a hot wall temperature of 450° F., and a cold wall temperature of 150° F. The feed stock for these tests was a lubricating oil stock obtained by solvent extraction of an oil refined from a Pennsylvania crude having a viscosity of 150 SSU at 100° F. and a viscosity index of 100. The feed was divided equally between the two end reservoirs in one run and then for comparison the feed was unequally divided in varying ratios. The runs were repeated for varying levels of throughput, and the results are reported in the following table:

Run No.	Feed Rate, ml./hr.	P_T Withdrawal Rate, ml./hr.	P_B Withdrawal Rate, ml./hr.	Bottom: Top Throughput	Viscosity Index of P_T
1	24	12	12	1:1	118
2	24	8	16	2:1	121
3	24	2.4	21.6	9:1	125
4	24	1.3	22.7	19:1	125.5
5	96	48	48	1:1	113
6	96	32	64	2:1	116
7	96	9.6	86.4	9:1	123
8	96	4.8	91.2	19:1	123
9	480	240	240	1:1	103
10	480	160	320	2:1	107
11	480	48	432	9:1	118
12	480	24	456	19:1	118.5

It may be noted from the above table that as the throughput increases, the effectiveness of the separation decreases, as is understood in the thermal diffusion art, but that at any given level of throughout to the thermal diffusion unit the top product quality in terms of viscosity index numbers improves (i.e., the viscosity index number increases) as the ratio of bottom throughput to top throughput is increased. There is a marked improvement in changing the ratio from 1:1 to 2:1. Obviously from the results shown, the major improvement is gained from increasing this ratio to about 9:1, and the subsequent improvement gained by increasing this ratio from 9:1 to 19:1 is nominal and generally not justifiable economically.

Furthermore, it will be observed from the foregoing table that if the top product quality is held constant while the bottom to top throughput ratio is increased, the rate of top product production will be increased. Compare, for example, Run No. 1 where 12 ml./hr. of 118 VI top product is produced at a bottom to top throughput ratio of 1:1 with Run No. 11 where 48 ml./hr. of 118 VI top product is produced when this ratio is increased to 9:1. Thus the amount of 118 VI product is increased fourfold without any increase in the consumption of heat.

Moreover, if the top product rate is held constant while the bottom to top throughput ratio is increased, the quality of the top product will be improved. Compare, for example, Run No. 5 where a 113 VI top product is obtained at a rate of 48 ml./hr. at a ratio of 1:1 with Run No. 11 of the table where a 118 VI top product is obtained at the same rate of 48 ml./hr. when the ratio is increased to 9:1. In this case the VI can be raised 5 numbers in accordance with the invention with no increase in heat consumption.

It is to be expected that various modifications of the method of the present invention will suggest themselves to those skilled in the art upon a reading of the foregoing description. All such modifications are intended to be included as are defined in the appended claims.

We claim:

1. In a thermal diffusion method which comprises confining a liquid mixture in a vertical thermal diffusion separation chamber comprising opposed walls and terminating in reservoirs at its upper and lower ends, said reservoirs having a width and height greater than the distance between said opposed walls, imposing a temperature gradient across the liquid in the chamber, whereby dissimilar fractions are formed, one of which contains a higher than initial concentration of one component and ascends to enter the upper reservoir and another of which contains a higher than initial concentration of another component and descends to enter the lower reservoir, continuously feeding fresh liquid into the upper and lower reservoirs for admixture with the dissimilar fractions from said thermal diffusion separation chamber, directing portions of the admixed liquid in the respective reservoirs into the upper and lower ends of the thermal diffusion separation chamber, and continuously and separately withdrawing dissimilar fractions from the upper and lower reservoirs, the improvement providing a higher rate of separation and an improved quality enriched fraction from one of said reservoirs in which the rate of the fresh feed to the upper reservoir F_T is approximately equal to the rate of withdrawal from the upper reservoir P_T and in which the rate of fresh feed to the lower reservoir F_B is approximately equal to the rate of withdrawal from the lower reservoir P_B and in which

$$\frac{F_T}{F_B} = X$$

where X has a value within the range from $\frac{1}{20}$ to $\frac{1}{2}$ and from 2 to 20, and the greater rate of feed is introduced through the reservoir at the end of the chamber opposite that from which an improved quality enriched fraction is being withdrawn.

2. In a thermal diffusion method for continuously separating a liquid mixture into fractions enriched with dissimilar components which comprises imposing a temperature gradient across a plurality of separation zones defined by opposed, substantially vertical surfaces substantially equidistantly spaced apart and communicating at their upper and lower ends with reservoirs having dimensions greater than the distance between said vertical surfaces and constituting a series of mixing zones; continuously introducing liquid mixture containing said dissimilar components into a first member of said series of mixing zones in said upper and lower reservoirs, whereby the

separation zones are filled with liquid mixture for separation into ascending fractions containing higher than initial concentrations of one of the dissimilar components and descending fractions containing higher than initial concentrations of another of said dissimilar components; admixing introduced liquid in said upper and lower reservoirs with the ascending and descending fractions entering said reservoirs from the separation zones; and continuously and separately withdrawing fractions enriched with dissimilar components by admixture with said ascending and descending fractions from a member of said series of mixing zones remote from the place of introduction of said liquid mixture in said upper and lower reservoirs, respectively, the improvement in which the rate of the fresh feed to the upper reservoirs F_T is approximately equal to the rate of withdrawal from the upper reservoirs P_T and in which the rate of fresh feed to the lower reservoirs F_B is approximately equal to the rate of withdrawal from the lower reservoirs P_B and in which

$$\frac{F_T}{F_B}=X$$

where X has a value within the range from $\frac{1}{20}$ to $\frac{1}{2}$ and from 2 to 20, and the greater rate of feed is introduced through the reservoirs in the ends of the separation zones opposite those from which an improved quality enriched fraction is being withdrawn.

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