March 18, 1958 D. FRAZIER 2,827,171

THERMAL DIFFUSION METHOD AND APPARATUS

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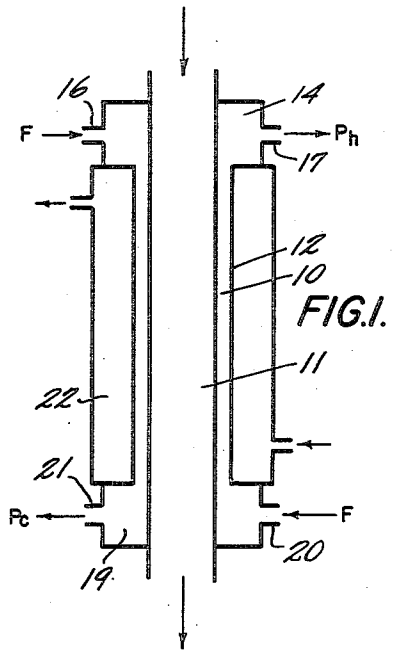


FIG.1.

FIG.2.

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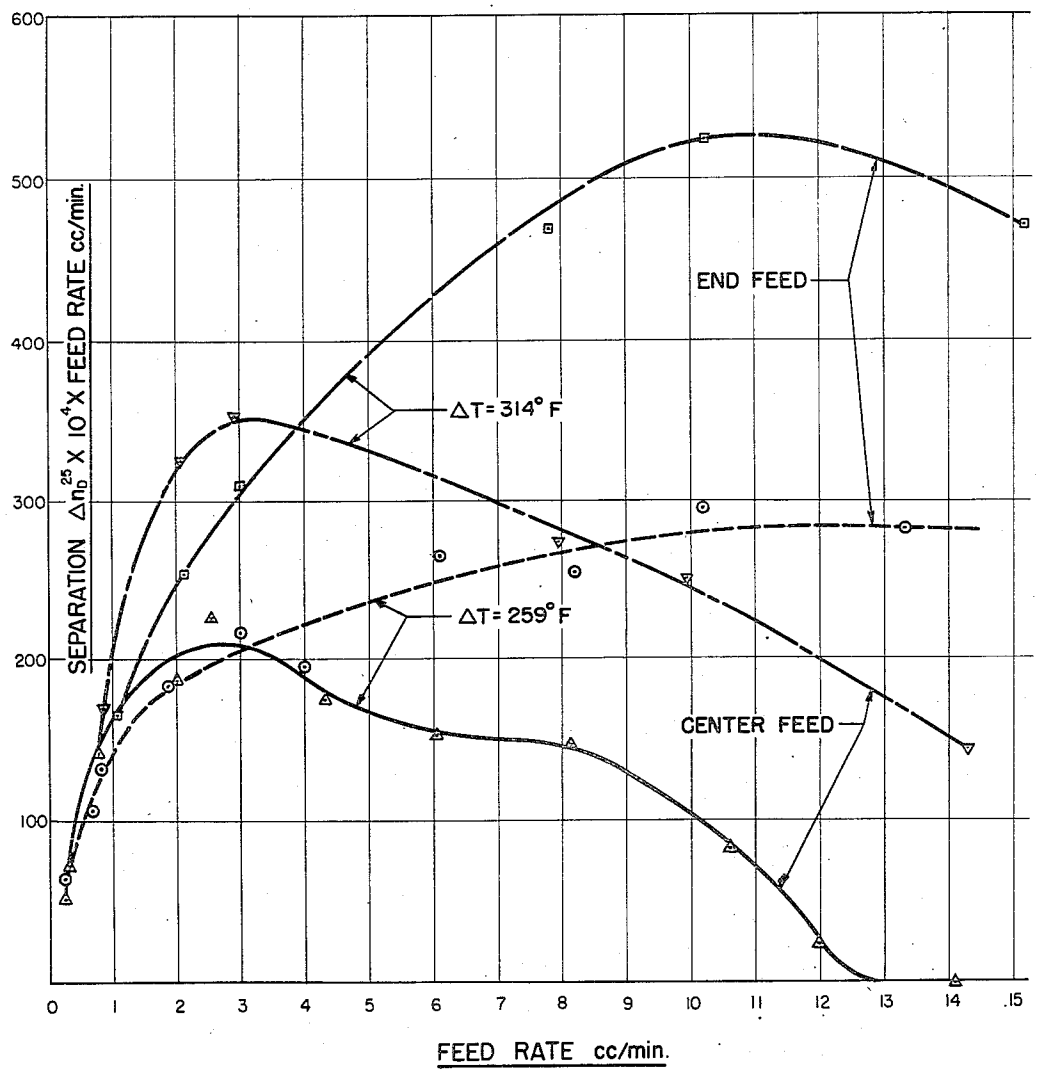
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THERMAL DIFFUSION METHOD AND APPARATUS

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FIG. 3.



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THERMAL DIFFUSION METHOD AND APPARATUS

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6 Claims. (Cl. 210—72)

This invention relates to a continuous method and apparatus for separating fluid mixtures into fractions enriched with dissimilar components.

It has been known for some time that it is possible to separate, by thermal diffusion, liquids and gases that are incapable of separation by any other known method, or that are separated by other methods with great difficulty. It was at first believed that thermal diffusion could be carried out only in a discontinuous, i. e., batchwise, manner. More recently, it has been discovered that it is possible to carry out thermal diffusion separation in a continuous manner.

The process of separation by thermal diffusion consists in essence of confining a fluid mixture, a term intended herein to include mixtures that are liquid or gaseous under the conditions of operation, in a narrow chamber and imposing across the chamber a temperature gradient. When the opposed walls defining such a chamber are non-horizontal, thermal circulation of the contents of the chamber is set up by virtue of the differences in density adjacent the relatively heated and cooled walls, referred to herein for convenience as the "hot" and "cold" walls. This thermal circulation is relied upon to separate the components of the mixture that preferentially accumulate adjacent the hot wall from those components that preferentially accumulate adjacent the cold wall.

It has been found that in order to obtain satisfactory rates of separation by thermal diffusion, the opposed thermal diffussion chamber-forming surfaces of the hot and cold walls should be spaced apart no more than about one-half inch, spacings of the order of 0.2 to 0.3 inch being preferred for separation of gaseous mixtures and smaller spacings of the order of 0.15 inch or less, preferably about 0.02 to 0.06 inch, being most effective for separation of liquid mixtures. In view of the small spacings involved, considerable difficulties have been encountered in taking advantage of the separation actually, obtained within a thermal diffusion separation chamber from the point of view of separately withdrawing the dissimilar fractions formed within the very narrow chamber and also in replacing these fractions with fresh fluid without undue turbulence and remixing of the separated fractions.

It has now been found that surprisingly good yields of dissimilar fractions are obtainable, particularly at relatively high feed rates, with thermal diffusion separation chambers formed by non-horizontal, equidistantly-spaced wall members, e. g., substantially vertical, concentric tubes. These excellent results are obtainable by providing upper and lower reservoirs communicating with the upper and lower ends of the chamber and having sufficient volume to permit mixing of fresh feed with fluids entering the reservoirs from the chamber and also to avoid a turnabout of the fluid within the separation chamber before it reaches the reservoir. 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.

By feeding the fresh fluid into the reservoirs, the separation chamber is filled with the fluid mixture which, by thermal diffusion, is separated into an ascending fraction containing a higher than initial concentration of one of the dissimilar components and a descending fraction containing a higher than initial concentration of another of the dissimilar components. The entering fluid mixture is admixed in the reservoirs with these ascending and descending fractions and continuously withdrawn therefrom.

The apparatus and method of the invention and the surprisingly good results obtainable therewith are described in greater detail by reference to the accompanying drawing wherein:

Figure 1 is a schematic illustration, in elevation, of a preferred embodiment of the apparatus of the invention; and

Figures 2 and 3 are graphs illustrative of the results obtainable with the method and apparatus of this invention as compared with those obtainable under comparable conditions with apparatus in which the entire feed of fluid mixture enters the chamber midway between the upper and lower ends.

The apparatus illustrated in Figure 1 comprises an annular thermal diffusion separation chamber **10** formed by the outer surface of an inner tube **11** and the inner surface of an outer tube **12**. The upper end of the annular chamber is provided with a reservoir **14** having an inlet connection **16** and an outlet connection **17**. The lower end of the chamber is similarly provided with a reservoir **19** having an inlet connection **20** and an outlet connection **21**. Suitable means not forming part of this invention are provided for relatively heating and cooling the chamber-forming walls. Thus, for example, the inner tube **11** may be relatively heated by passing steam through it, and the outer tube **12** may be relatively cooled by circulating water through a cooling jacket **22**.

In operation, fresh liquid is continously introduced into upper and lower reservoirs **14** and **19** by way of inlet connections **16** and **20** and a temperature gradient is imposed across the separation chamber **10** by relatively heating one of the inner and outer tubes and relatively cooling the other. The feed entering the reservoirs fills the thermal diffusion chamber **10** and while therein is resolved into a first fraction accumulating preferentially adjacent the hot wall ascending to the upper reservoir **14** and a second fraction accumulating preferentially adjacent the cold wall and descending to the lower reservoir **19**. Thus, for example, if the inner tube **11** is relatively heated and the outer tube **12** is relatively cooled, the ascending fraction containing a higher than initial concentration of one of the dissimilar components in the liquid mixture ascends along the outer surface of the inner tube **11** and the other fraction containing a higher than initial concentration of another of the dissimilar components descends along the inner surface of the outer tube **12** into the lower reservoir **19**. The fresh liquid mixture entering the reservoirs is admixed with these dissimilar fractions, a portion of the liquid enters the separation chamber **10** to replace the fractions entering the reservoirs and the remainder, enriched with the components in the dissimilar fractions, is separately withdrawn by way of withdrawal ports **17** and **21**.

A solvent extract neutral oil obtained by furfural extraction of an oil from a Mid-Continent crude and having a viscosity of 140 SUS at 100° F. and a refractive index of 1.4730 was subjected to separation by thermal diffusion at varying rates of feed in an annular separation chamber such as that illustrated in Figure 1 having a height of 6 feet, a mean diameter of one inch and a slit

width of 0.03 inch. One set of tests was conducted with the feed equally divided between the upper and lower reservoirs in accordance with the invention and was carried out at four different temperature gradients, the differences in temperature between the hot and cold walls being 259° F., 278° F., 291° F., and 314° F. The same tests were conducted with the same column but with all of the feed introduced midway between the upper and lower ends of the column. The feed rates, degree of separation in terms of change in index of refraction measured at 25° C., and efficiency in terms of the product of the feed rate and the degree of separation were as follows:

Center Feed Apparatus			Mixed End Feed Apparatus		
Feed Rate, cc./min.	Separation, $\Delta n_D^{25} \times 10^4$	Efficiency, Feed Rate × Separation	Feed Rate, cc./min.	Separation, $\Delta n_D^{25} \times 10^4$	Efficiency, Feed Rate × Separation
ΔT=259° F.					
0.256	202	52	0.367	179	66
0.273	199	54	0.376	186	70
0.384	196	75	0.740	143	106
0.876	162	142	0.912	144	131
2.04	92	188	1.99	93	185
2.59	87	226	3.05	69	218
4.44	40	177	4.06	48	195
6.11	25	153	6.12	43	263
8.20	18	148	8.26	31	256
10.62	8	85	10.24	29	297
12.0	2	24	13.40	21	281
14.10	0	0	--------	--------	--------
ΔT=278° F.					
1.04	163	170	1.16	135	157
2.99	106	317	2.00	110	220
4.26	76	324	3.34	80	267
6.1	40	244	3.80	72	274
9.0	20	180	6.52	60	391
11.44	13	149	9.30	40	372
14.95	7.5	112	12.34	31	382
--------	------	------	13.47	28	377
ΔT=291° F.					
1.04	174	181	1.16	146	169
2.0	139	278	1.8	125	225
4.0	89	356	3.9	88	343
6.0	52	312	5.75	64	368
8.0	31	248	8.0	51	408
10.0	16	160	9.9	42	416
12.4	6	74.4	12.4	32	397
ΔT=314° F.					
0.966	177	171	1.05	155	163
2.10	155	325	2.13	120	256
2.96	120	355	3.0	104	312
8.0	34	272	7.8	60	468
9.95	25	249	10.23	51	522
14.33	10	143	15.2	31	471

The results obtained with temperature differentials of 259 and 314° F. are graphically illustrated in Figures 2 and 3. Both graphs reveal that at feed rates above about 3 to 5 cc. per minute, the degree of separation obtainable in accordance with mixed end feed of the invention is outstandingly superior to that obtainable with the center feed method, all other factors, such as column height, mean diameter, slit width, feed stock and temperature gradient, being equal. Figure 3 shows the remarkably higher efficiency that is obtainable at such higher feed rates.

It is apparent that the apparatus and method of the invention have a number of important advantages, among the most important of which are the higher degree of separation and greater efficiency of separation obtainable at commercially practicable feed rates, and that the feed rates at which superior result are obtainable for any given set of conditions in accordance with the invention, as compared with those obtainable by the center feed method, are readily ascertainable in the manner illustrated herein by way of example.

It is to be understood of course that numerous variations and applications will readily occur to those skilled in the art upon reading this description. All such variations and applications are intended to be included within the scope of the invention as defined in the appended claims.

I claim:

1. Thermal diffusion apparatus for separating a fluid mixture into fractions enriched with dissimilar components which comprises substantially vertical concentric tubes defining between their opposed vertical walls an annular thermal diffusion separation chamber having an upper end and a lower end, means for relatively heating and cooling the chamber-forming concentric tubes to maintain a temperature gradient across the separation chamber, an upper reservoir communicating with the upper end of the separation chamber for receiving from the separation chamber an ascending fraction having a higher than initial concentration of one of the dissimilar components, said upper reservoir having a width and height greater than the distance between said vertical walls so that the volume of the upper reservoir permits mixing of fresh feed with fluid entering and leaving the upper end of the separation chamber, and a lower reservoir communicating with the lower end of the separation chamber for receiving from the separation chamber a descending fraction having a higher than initial concentration of another of the dissimilar components, said lower reservoir having a width and height greater than the distance between said vertical walls so that the volume of the lower reservoir permits mixing of fresh feed with fluid entering and leaving the lower end of the separation chamber, means for continuously feeding fluid to the upper and lower reservoirs and admixing it with the ascending and descending fractions, respectively, and means for continuously withdrawing from said upper and lower reservoirs fractions enriched with the dissimilar components in said ascending and descending fractions.

2. Thermal diffusion apparatus for separating a liquid mixture into fractions enriched with dissimilar components which comprises substantially vertical concentric tubes defining between their opposed vertical walls an annular thermal diffusion separation chamber having an upper end and a lower end, means for relatively heating and cooling the chamber-forming concentric tubes to maintain a temperature gradient across the separation chamber, an upper reservoir communicating with the upper end of the separation chamber for receiving from the separation chamber an ascending fraction having a higher than initial concentration of one of the dissimilar components, said upper reservoir having a width and height at least about twice the distance between said vertical walls so that the volume of the upper reservoir permits mixing of fresh feed with fluid entering and leaving the upper end of the separation chamber, and a lower reservoir communicating with the lower end of the separation chamber for receiving from the separation chamber a descending fraction having a higher than initial concentration of another of the dissimilar components, said lower reservoir having a width and height at least about twice the distance between said vertical walls so that the volume of the lower reservoir permits mixing of fresh feed with fluid entering and leaving the lower end of the separation chamber, means for continuously feeding liquid to the upper and lower reservoirs and admixing it with the ascending and descending fractions, respectively, and means for continuously withdrawing from said upper and lower reservoirs fractions enriched with the dissimilar components in said ascending and descending fractions.

3. Thermal diffusion method which comprises confining a fluid 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 fluid 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 fluid into the upper and lower reservoirs for admixture with the dissimilar fractions from said thermal diffusion separation chamber, directing portions of the admixed fluid 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.

4. 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 tempreature 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.

5. Method for continuously separating a fluid mixture into fractions enriched with dissimilar components which comprises imposing a temperature gradient across an annular separation chamber defined by the opposed walls of substantially vertical concentric tubes and communicating with upper and lower reservoirs at its upper and lower ends; said reservoirs having a width and height at least about twice the distance between the opposed walls, continuously feeding fresh fluid into said reservoirs, whereby the separation chamber is filled with fluid mixture for separation into an ascending fraction containing a higher than initial concentration of one of the dissimilar components and a descending fraction containing a higher than initial concentration of another of the dissimilar components and the entering fluid mixture is admixed in said reservoirs with the ascending and descending fractions; and continuously and separately withdrawing, from the upper and lower reservoirs, fractions enriched with dissimilar components by admixture with the ascending and descending fractions.

6. Method for continuously separating a liquid mixture into fractions enriched with dissimilar components which comprises imposing a temperature gradient across an annular separation chamber defined by the opposed walls of substantially vertical concentric tubes and communicating with upper and lower reservoirs at its upper and lower ends; said reservoirs having a width and height at least about twice the distance between the opposed walls, continuously feeding fresh liquid into said reservoirs, whereby the separation chamber is filled with liquid mixture for separation into an ascending fraction containing a higher than initial concentration of one of the dissimilar components and a descending fraction containing a higher than initial concentration of another of the dissimilar components and the entering liquid mixture is admixed in said reservoirs with the ascending and descending fractions; and continuously and separately withdrawing, from the upper and lower reservoirs, fractions enriched with disimilar components by admixture with the ascending and descending fractions.

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