

March 11, 1941.

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2,234,166

VACUUM DISTILLATION

Filed Nov. 3, 1939

2 Sheets-Sheet 1

Fig. 1.

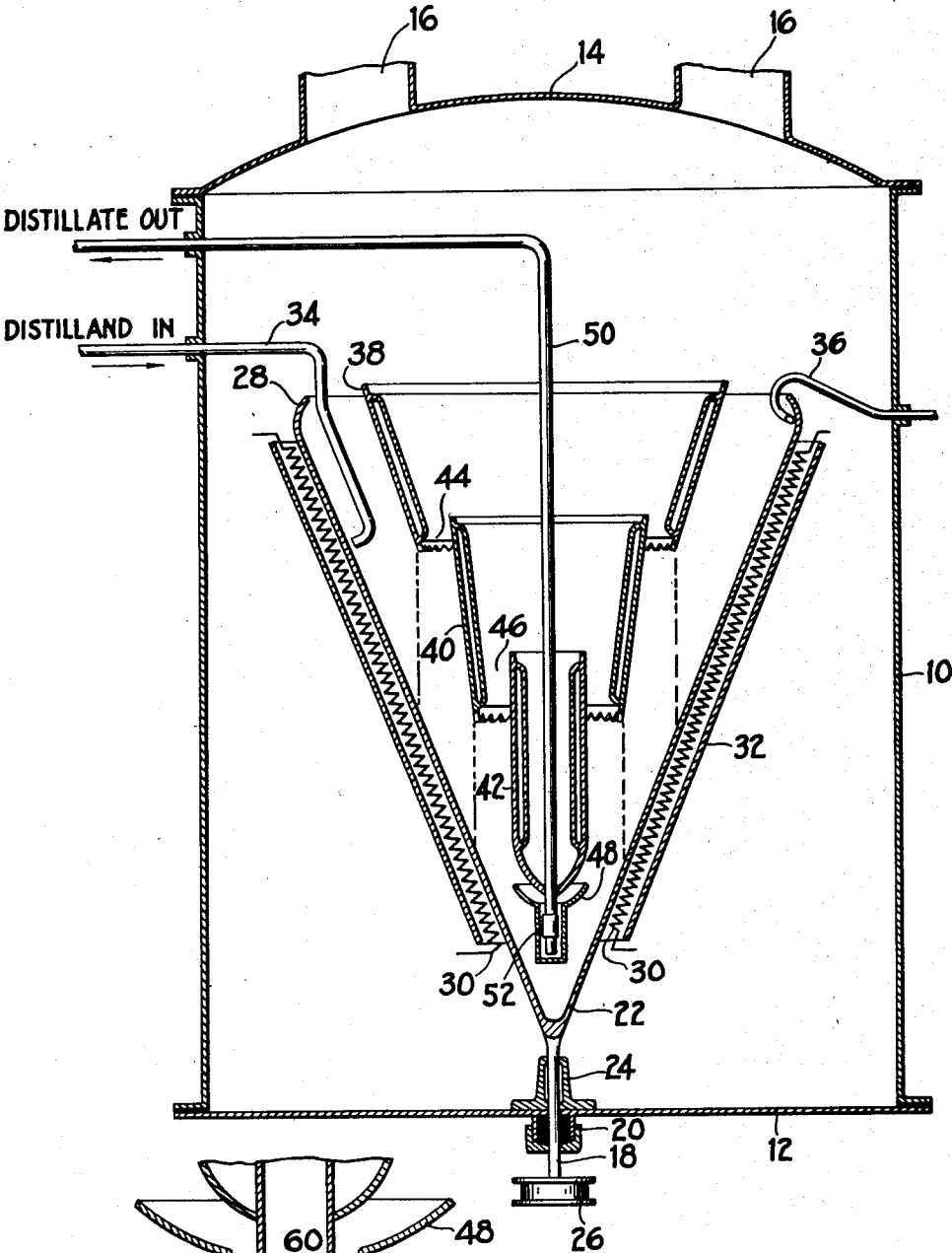
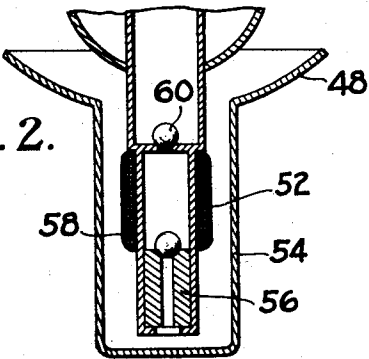


Fig. 2.



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2 Sheets-Sheet 2

Fig. 3.

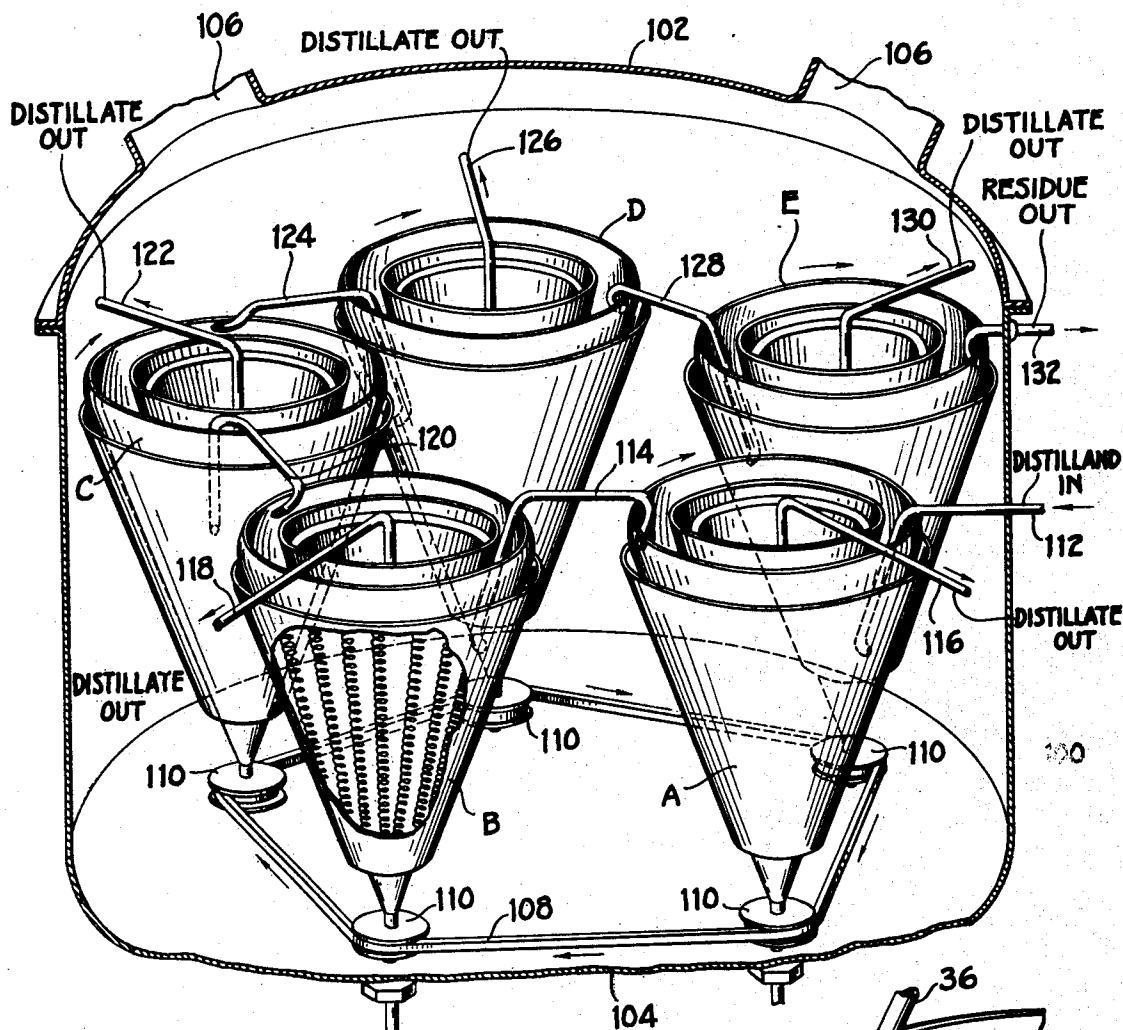


Fig. 5.

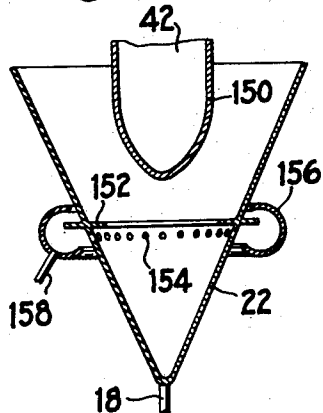


Fig. 6.

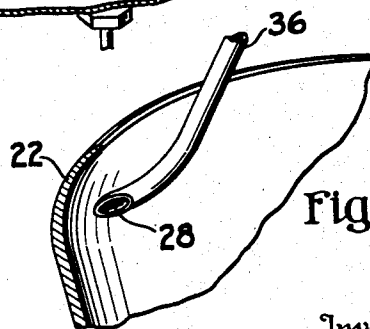
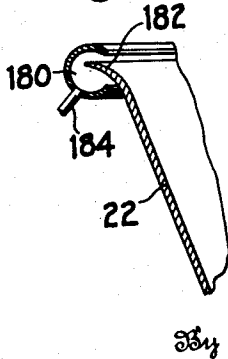


Fig. 4.

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UNITED STATES PATENT OFFICE

2,234,166

VACUUM DISTILLATION

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Application November 3, 1939, Serial No. 302,747

6 Claims. (Cl. 202—205)

This invention relates to the art of vacuum distillation and in particular relates to apparatus for high vacuum unobstructed path distillation from moving heated vaporizing surfaces.

5 In application 99,632, filed September 5, 1936, I have described vacuum distillation procedure wherein the substance to be distilled is introduced onto a moving heated surface over which it is caused to pass due to the movement of the surface at a rate substantially greater than would be obtained if the distilland were caused to flow by gravity. One of the methods of distilling in this manner involves introducing the distilland onto the approximate center of a rotating disk or plate which is heated to distillation temperature.

10 This invention has for its object to provide improved distillation apparatus of the nature described above. A further object is to provide high vacuum unobstructed path distillation apparatus for distilling from a moving surface whereby fractionating may be accomplished from one moving surface. A further object is to provide distillation apparatus whereby distilland can be introduced onto the inner surface of a heated revolving cup or cone and a plurality of fractions separated from the distilland during its passage over said surface. A further object is to provide improved high vacuum unobstructed path distillation apparatus. A still further object is to provide an efficient centrifugal still which avoids problems of pumping condensates and residues and/or avoids difficulties due to splashing of distilland or residue during its passage over the centrifugal surface. Other objects will become apparent from the following description and claims.

15 These and other objects are accomplished by my invention which includes apparatus whereby vacuum fractional distillation, i. e., separation of a plurality of fractions or repeated fractionation of a single fraction is accomplished by distillation from one, heated, cone or cup shaped, rotating vaporizing surface. A plurality of such multiple effect cone surfaces can be employed.

20 In the following description I have set forth several of the preferred embodiments of my invention. However, it is to be understood that they are given for the purpose of illustration and not in limitation thereof.

In the accompanying drawings in which like numbers refer to like parts.

25 Fig. 1 is a vertical section of a still adapted to repeatedly fractionate a component and separate it in a form more concentrated than could

be achieved in a single act of evaporation and condensation. In accordance with my invention the still is provided with a conical vaporizing surface from which distillation takes place from the inside surface thereof.

30 Fig. 2 is a fragmentary enlarged detail of the pump located at the base of the condensing units in Fig. 1.

Fig. 3 is a diagrammatic drawing of a plurality of multi-stage still units similar to that illustrated in Fig. 1 all of which are housed in a single still casing.

Fig. 4 is a fragmentary detail of the upper edge of the conical vaporizing surface and illustrates the manner in which the undistilled residue which collects in the turned edge thereof is removed.

Fig. 5 is a fragmentary detail of the lower portion of the cone vaporizing surface and illustrates an alternative method of removing condensate from the lowest portion of the condensing surface.

Fig. 6 is a detail of a modified method of removing residue from the upper edge of the cone and illustrates the edge of a conical evaporator which is surrounded by a stationary gutter to collect the undistilled residue.

Referring to Figs. 1, 2, and 4, numeral 10 designates a cylindrical still casing mounted upon base plate 12 in a gas-tight manner. The casing is provided with a closure 14 to which is connected evacuating conduits 16. At the approximate center of plate 12, there is provided a shaft 18 which passes through a packed gas-tight gland 20. Shaft 18 is integral with and terminates in cone shaped vaporizing surface 22. The shaft is rigidly mounted by means of support 24 so that cone 22 is maintained in the position illustrated when rotated by force transmitted through pulley 26. The upper edge or mouth of cone 22 is bent or turned inwardly to form an inverted gutter 28. Cone 22 is surrounded by an electrical heating element 30, the heat from which is reflected against the column by reflector 32. Numeral 34 designates a conduit which introduces the distilland onto the upper portion of the vaporizing cone at the point where it terminates near the cone. Numeral 36 designates a conduit, one end of which terminates inside gutter 28 and which serves to withdraw undistilled residue from the top of the cone vaporizing surface.

35 Numerals 38, 40, and 42 designate a plurality of double-walled condensing surfaces arranged in sequence within the cone 22 but in spaced relation thereto. Each of the condensing units 38, 40, and 42

40, and 42 are cooled by circulation of cooling fluid through the space present between the two walls thereof. Condensing units 38 and 40 are provided at the base thereof with a serrated edge. The base of condensing unit 38 is preferably located so that condensate dropping therefrom by gravity will strike the surface of cone 22 at a point approximately opposite to the lowest portion of condensing unit 40. The bottom edge of condensing unit 40 is so located that condensate dropping therefrom by gravity strikes the vaporizing cone at a point approximately opposite the lowest portion of condensing unit 42. The path of the condensate in dropping from condensing units 38 and 42 is shown in dotted lines. The condensing units 38, 40 and 42 are so designed as to leave gaps 44 and 46 for removal of air and gases from the distilling gap. Numeral 48 designates a cup integral with the bottom portion of condensing unit 42. The base of the cup is extended downward and withdrawal conduit 50 extends into said extension. At the base of conduit 50 and located in the extension of cup 48 is a magnetically operated pump 52 which is entirely located within the extension 54. The magnetic pump construction is shown in Fig. 2 and involves a hollow plunger 56 provided with a ball bulb 58 which is pulled upwardly by solenoid 52 and moved downwardly by gravity. Numeral 60 designates a ball valve at the top of the pump.

Referring to Fig. 3, numeral 100 designates a cylindrical casing provided with top and base plates 102 and 104. The upper or top plate is integral with evacuating conduits 106. Within the casing 100 are mounted 5 complete distilling units, A, B, C, D, and E, which are substantially identical with the distilling unit shown in Fig. 1. Alternatively they may be of progressively diminishing size. The distilling units are rotated by a common belt 108 which is held during operation by pulleys 110 at the base of each vaporizing cone. Numeral 112 designates the conduit through which the original distilland is introduced onto the first distilling unit A. Numeral 114 designates the conduit which removes undistilled residue from the vaporizing column of still A at the upper edge thereof and delivers it onto the upper portion of the vaporizing cone of still B. Numeral 116 designates the conduit through which the first fraction separated in still A is withdrawn from the still. Numeral 118 designates a conduit which serves to remove the second fraction fractionally distilled in still B. Numeral 120 designates a conduit which serves to remove undistilled residue from the vaporizing cone of still B and delivers it onto the upper portion of the vaporizing cone in still C. Numeral 122 designates a conduit through which the fractional distillate separated in still C is removed from the apparatus. Numeral 124 designates a conduit which serves to convey undistilled residue from still B onto the vaporizing cone of still D. Numeral 126 designates a conduit by which distillate fractionally distilled in still D is removed from the apparatus. Numeral 128 designates a conduit which delivers residue undistilled in still D onto the cone vaporizing surface of still E. Numeral 130 designates a conduit for removing the fraction separated in still E and numeral 132 designates a conduit from the top surface of vaporizing cone in still E by which final undistilled residue is removed.

Referring to Fig. 5, numeral 150 designates the lower end of the lowest vaporizing unit 42. The

end is brought to a blunt point and the liquid condensate thereon is permitted to drop by gravity as shown into the bottom of vaporizing cone 22. The base of the cone is provided with a flange 152 which extends an appreciable distance from the inside and outside wall of the cone. Immediately below the flange are provided a plurality of holes 154. Numeral 156 designates an annular gutter into which flange 152 protrudes at its outer edge. Numeral 158 designates a conduit for withdrawing liquid from gutter 156.

Referring to Fig. 6 numeral 180 designates an annular gutter or hollow ring split on the inside to form an opening into which the outwardly turned edge 182 of cone 22 protrudes. Numeral 184 designates a withdrawal conduit.

During operation of the apparatus illustrated in Figs. 1, 2 and 4, conduits 16 are connected to a source of high vacuum and the system is evacuated to the required extent. Cone 22 is caused to rotate by force transmitted through pulley 26. Electrical heating unit 30 is put into operation so that vaporizing cone 22 is heated to distillation temperature. Distilland which is preferably in a degassed condition is introduced under the inside surface of cone 22 by way of conduit 34. This distilland is caused to flow upward from the point of introduction to the mouth of the cone in the form of a thin film. This is due to centrifugal force. During passage over the upper portion of cone 22, the film of distilland is heated to distillation temperature. Vapors thus produced condense on the cooled walls of condensing unit 38. Undistilled residue collects in gutter 28 at the top of the cone and due to the speed of rotation is thrown into the end of conduit 36 as illustrated in Fig. 4. The inertia of the liquid entering conduit 36 is sufficient to cause it to flow up hill the short distance required to get over the top of cone 22. This undistilled residue then flows from the still or is introduced onto the next still of the series if a plurality of such stills are employed.

Liquid condensing on the surface of condensing unit 38 flows to the base thereof and drops by gravity on to the surface of cone as illustrated by the dotted lines. The point of contact between these droppings and the cone is preferably approximately opposite the lowest portion of the next lowest condensing unit 40. This condensate dropping onto cone 22 is caused to flow upwards by centrifugal force. The cone is heated and distillation from this upwardly flowing film of condensate takes place. All low vapor pressure components which were accidentally separated and condensed on unit 38 are substantially rejected during this upward travel and these low vapor pressure fractions are immediately intermixed with the fresh distilland as it passes over the upper portion of the cone. The more volatile portions are vaporized and condense on condensing unit 40. This condensate flows by gravity to the bottom edge of 40 and drops onto the vaporizing cone at a point approximately opposite the base of condensing unit 42. These droppings are likewise caused to flow upwards in a thin film and vaporization of the lightest vapor pressure component takes place. Lower vapor pressure components remain undistilled and become intermixed with the droppings from condensing unit 38. The lightest and substantially pure fraction condenses on condensing unit 42, flows by gravity into cup 48 and is elevated from this point by magnetic pump 52 and is

withdrawn through the still by way of conduit 59.

It will be seen that a three-fold fractionation is directly accomplished by the one operation of introducing distilland onto the upper portion of the cone surface. Fractionation then automatically takes place with automatic return of condensate to the appropriate points on the vaporizing surface. No pumps or manual operations are required to effect the separation of a highly concentrated fraction by this apparatus.

The pitch of the cone can be varied substantially in order to permit variation in number of condensing surfaces employed. Also the contours of the cone need not be uniform throughout the length and any cup-shaped vessel may be used. Such variations are to be understood as being within the meaning of the word "cone" as used herein. Of course the number of condensing surfaces can be varied widely and in case three successive fractionations are insufficient to purify a fraction completely, the length or pitch of the cone can be altered to permit the use of a larger number of condensing units. Instead of utilizing free gravitational fall to return the various distillates to the appropriate portions of the vaporizing surface the return can be effected by pipes, gutters or wetted rods. These can be at fixed angles other than the vertical and the liquid distillate thus returned to any lower point. This variation permits the arrangement of the condensing surfaces to be varied. For instance, the condensing surfaces can be then located closer to the cone. Also, gravitational return of distillate to the cone is not a necessary feature of my invention. If desired the cone can be arranged upside down and the condensing surfaces provided with collecting gutters from which the distillates could be pumped through return pipes to appropriate higher portions of the cone.

In some cases the removal of the final distillate fraction by the method illustrated in Fig. 1 is inconvenient. Many other methods are available. The final fraction can be frozen and mechanically removed continuously or intermittently. Endless conveyor belts may be used to remove ~~the~~ either in liquid or solid form. Rod operated piston or gear pump may replace the magnetic pump. An alternative method of removal is illustrated in Fig. 5 during the operation of which condensate drops from condensing surface 42 into the lower portion of the cone 22. Due to centrifugal force, this liquid condensate is lifted as far as flange 152. However, instead of overflowing the inner edge thereof it is forced or flung through holes 154 into gutter 156. Any liquid which tends to travel from holes 154 up the outside surface of cone 22 is stopped by the outer edge of 152 and is flung by centrifugal force into gutter 156. The condensate collected in gutter 156 is removed from the still by way of conduit 158.

When operating apparatus provided with the gutter removal means shown in Fig. 6, the undistilled residue is thrown by centrifugal force from outwardly turned edge 182. This residue is caught by gutter 180 and flows into conduit 184 through which it is removed from the still.

The operation of each of the units illustrated in the apparatus of Fig. 3 is substantially the same as that described in connection with Fig. 1. In each unit the substance to be distilled is introduced onto the upper portion of the vaporizing cone and distillate thus obtained is thrice fractionated and the final fraction removed through conduits 110, 118, 122, 126 and 130. The

lowest fraction is removed in concentrated form in still A. The next highest boiling point fraction is removed in still B, the next highest in still C, etc. The number of stills should correspond to the number of fractions which are to be separated. The apparatus illustrated in Fig. 1 could be used in series, each unit of the series being housed in a separate casing. However, disposing all of the still units in a single casing greatly reduces pumping costs and other operating expenses and difficulties.

The vaporizing cone can be divided into a plurality of definite temperature zones. The passage of undistilled residue from one to the other of which can be easily accomplished without splashing by numerous devices, one of which is slight over-lapping of the different zones. Ordinarily I prefer to employ a single vaporizing cone and, if need be, heat the different vaporizing zones thereof to different vaporizing temperatures. The condensing zones likewise can be constructed as one continuous surface. At appropriate points flanges or similar drop-off leaves may be provided to direct downwardly flowing condensate from the condensing surface so that it will drop onto the vaporizing cone at the appropriate point. I prefer to use separate vaporizing units since the gaps between each unit, such as gaps 44 and 46, Fig. 1, provided additional space for removal of gases from the distilling zone. If desired evacuating pumps can be directly connected to the top edges of each such condensing unit. Very efficient removal of gases would then be possible. The conical surface has the decided advantage that direct introduction of liquid thereon at a plurality of points does not cause splashing. The introduced liquid is thrown against the conical surface and held tightly thereto.

The vapors derived from the different vaporizing surfaces or zones may become mixed to a certain extent. The mixing diminishes as the distance between the condensing and vaporizing zones is shortened. With a four foot cone and a distance of two inches mixing is not serious. The distance shown in the drawings are rather large in proportion to the plates, and were made artificially wide so that the details of construction could be more clearly seen.

The mode of recirculation to accomplish efficient fractionation can be varied greatly. Thus the distilland may be introduced onto the intermediate vaporizing areas of the cone instead of on the lower or upper segment. Condensate from the different condensing zones may be recirculated in a variety of sequences to improve fractionation or to accomplish a particular fractionating result. Attention is directed to U. S. Patents 2,073,202 and 2,128,223 for common methods of recirculation which may be used with advantage when fractionating with a particular point in view.

Although I find it useful procedure to heat the different vaporizing zones of a single still unit to different temperatures, this is not always necessary. If desired, the entire vaporizing surface may be at the same temperature but the areas of the different portions thereof may be varied. It is a characteristic of high vacuum unobstructed path or short path distillation that substances all distill over any temperature range, but the rate of distillation varies with the area exposed and the molecular weight, as well as with the temperature. If the area is increased the same result is obtained as if the temperature

were increased, but the area maintained the same or reduced. The area of course controls the time of heating of the distilland which is also affected by the speed of rotation. Therefore, the different zones can have equal areas and be heated to successively higher temperatures or they may be heated to equal temperatures with decreasing areas or any combination of the two.

In order to separate a larger number of fractions than those specifically described, any of the different forms of apparatus may be employed in combination by connecting them in series. The temperature or temperatures in each unit of the series will be appropriate for the component removed therein and will usually progressively increase from one unit to the other in the same order that the distilland passes therethrough.

My invention is applicable to distillation under vacuum distillation conditions. However, I have found it to be of particular value for distilling under high vacuum unobstructed path distillation conditions, i. e., at pressures of below 1 mm. such as for instance .1-.001 mm. and with condensing and vaporizing surfaces separated by substantially unobstructed space. When the distance between the surfaces is short such as a distance less than 12", and preferably a distance of about 1/2 to 6", the distillation is known as high vacuum, short path distillation. When the distance is less than the mean free path it is known as molecular. My invention is applicable to all such distillations.

Reference is made to my application No. 99,632 referred to above for further details of construction of stills having moving surfaces, their mode of operation and materials operated upon. In my co-filed application Serial No. 302,748, filed November 3, 1939, I have described other forms of fractionating centrifugal fractionating surfaces and reference is made thereto for further details of construction and operation of centrifugal stills.

What I claim is:

1. High vacuum, unobstructed path fractional distillation apparatus which comprises in combination a rotatable cone shaped vaporizing surface, a plurality of condensing zones arranged within the cone and separated therefrom by substantially unobstructed space, a conduit for introducing liquid to be distilled onto the inside of the vaporizing cone so that during operation it is caused to flow thereover by centrifugal force toward the mouth thereof, means for removing undistilled residue from the mouth of the vaporizing surface, means for separately removing condensate from each of the condensing zones and means for evacuating the space between the condensing zones and the vaporizing surface.

2. High vacuum, unobstructed path fractional distillation apparatus which comprises in combination a rotatable cone shaped vaporizing surface, a series of condensing surfaces arranged within the cone and separated therefrom by substantially unobstructed space, a conduit for introducing liquid to be distilled onto the inside of the vaporizing surface near to the mouth thereof, so that during operation it is caused to flow thereover toward the mouth by centrifugal force,

means for removing undistilled residue from the mouth of the vaporizing surface, means for returning condensate from each of the condensing surfaces, except the last, to the cone at a point farther from the mouth than it was vaporized from, means for evacuating the space between the condensing and vaporizing surfaces and means for removing from the still condensate which condenses on the last condensing surface of the series.

3. High vacuum, unobstructed path fractional distillation apparatus which comprises in combination a rotatable cone shaped vaporizing surface, a series of condensing surfaces or zones arranged in sequence within the cone but in spaced relation with the walls thereof, means for heating the cone surface, means for cooling the condensing surfaces, a conduit for introducing liquid to be distilled onto the inside of the vaporizing cone surface so that during operation it is caused to flow thereover from the point of introduction to the mouth of the cone vaporizing surface, means for removing undistilled residue from the mouth of the cone vaporizing surface, means for conveying condensate from each of the condensing surfaces or zones except the last, onto the vaporizing cone at a point approximately opposite to the portion of the next condensing surface or zone in the series, means for removing condensate from the still which collects on the last condensing surface or zone of the series and means for maintaining a high vacuum in the space between the condensing surfaces or zones and the vaporizing cone.

4. The apparatus of claim 3 in which the rotatable cone is approximately vertical with the mouth of the cone uppermost.

5. The apparatus of claim 3 in which the rotatable cone is approximately vertical and the condensing surfaces or zones are arranged so that the return of condensate to the cone takes place by gravitational force.

6. High vacuum, unobstructed path fractional distillation apparatus which comprises in combination a rotatable cone shaped vaporizing surface, a series of condensing surfaces or zones arranged in sequence within the cone but in spaced relation with the walls thereof, means for heating the cone surface, means for cooling the condensing surfaces, a conduit for introducing liquid to be distilled onto the inside upper portion of the vaporizing cone surface so that during operation it is caused to flow thereover in an upward direction from the point of introduction to the top of the cone vaporizing surface, means for removing undistilled residue from the top of the vaporizing surface, means for conveying condensate from each of the condensing surfaces or zones except the lowest onto the vaporizing cone at a point approximately opposite to the lowest portion of the next lowest condensing surface or zone in the series, means for removing condensate from the still which collects on the lowest condensing surface or zone and means for maintaining a high vacuum in the space between the condensing surfaces or zones and the vaporizing cone.

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