March 30, 1954 A. H. RADASCH 2,673,833

DISTILLATION OF COAL TAR

Filed April 3, 1951 4 Sheets-Sheet 1

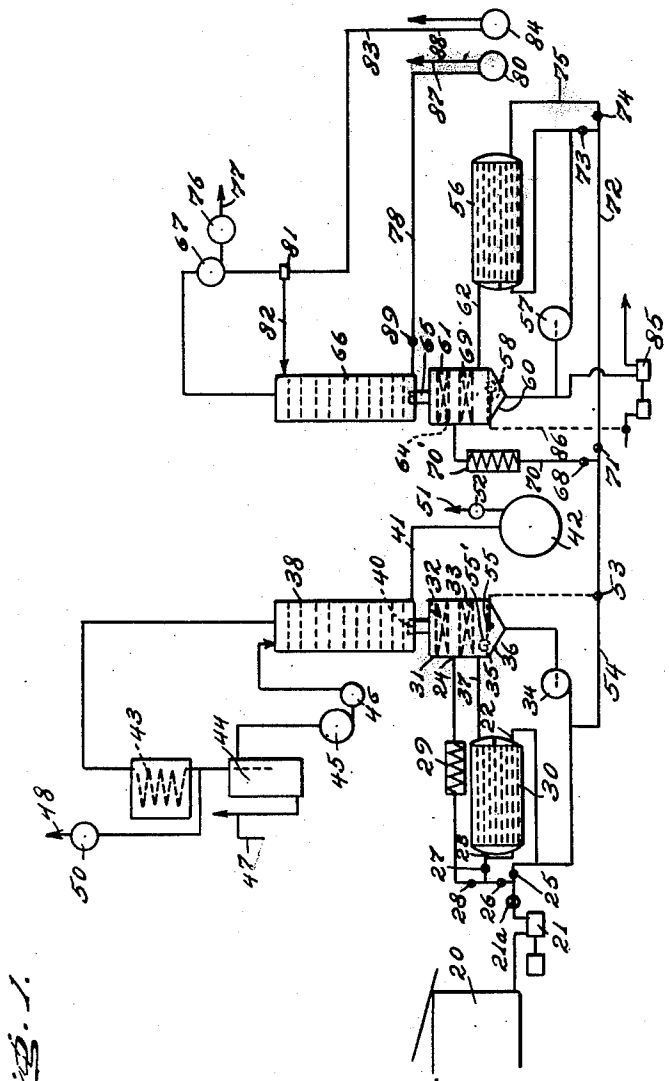

Fig. 1.

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DISTILLATION OF COAL TAR

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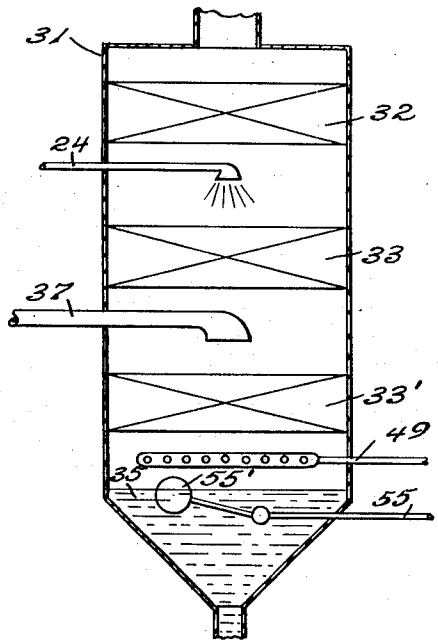
Fig. 1a.

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DISTILLATION OF COAL TAR

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Fig. 2.

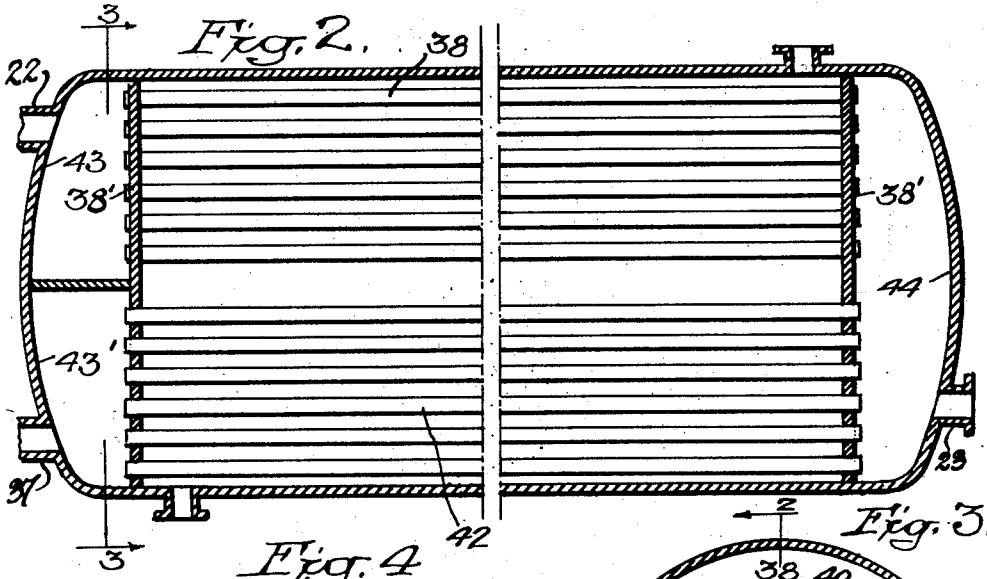



Fig. 3.

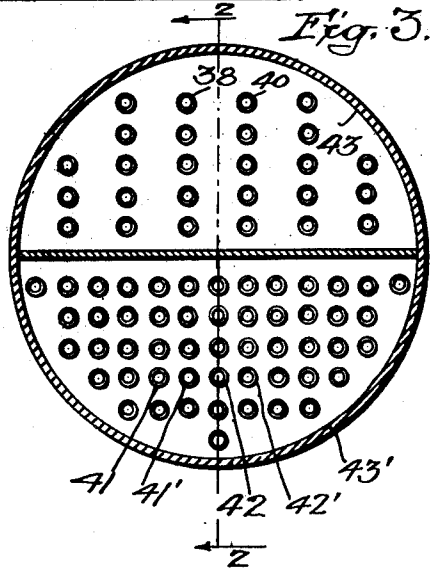
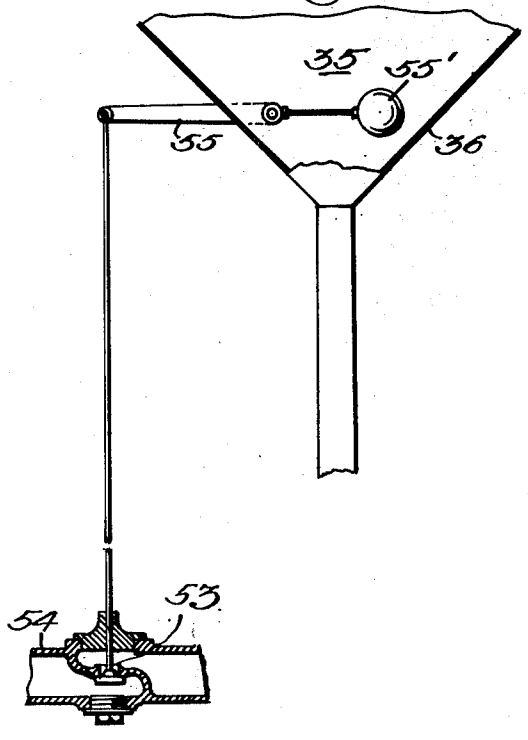



Fig. 4







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DISTILLATION OF COAL TAR

Filed April 3, 1951 4 Sheets-Sheet 4

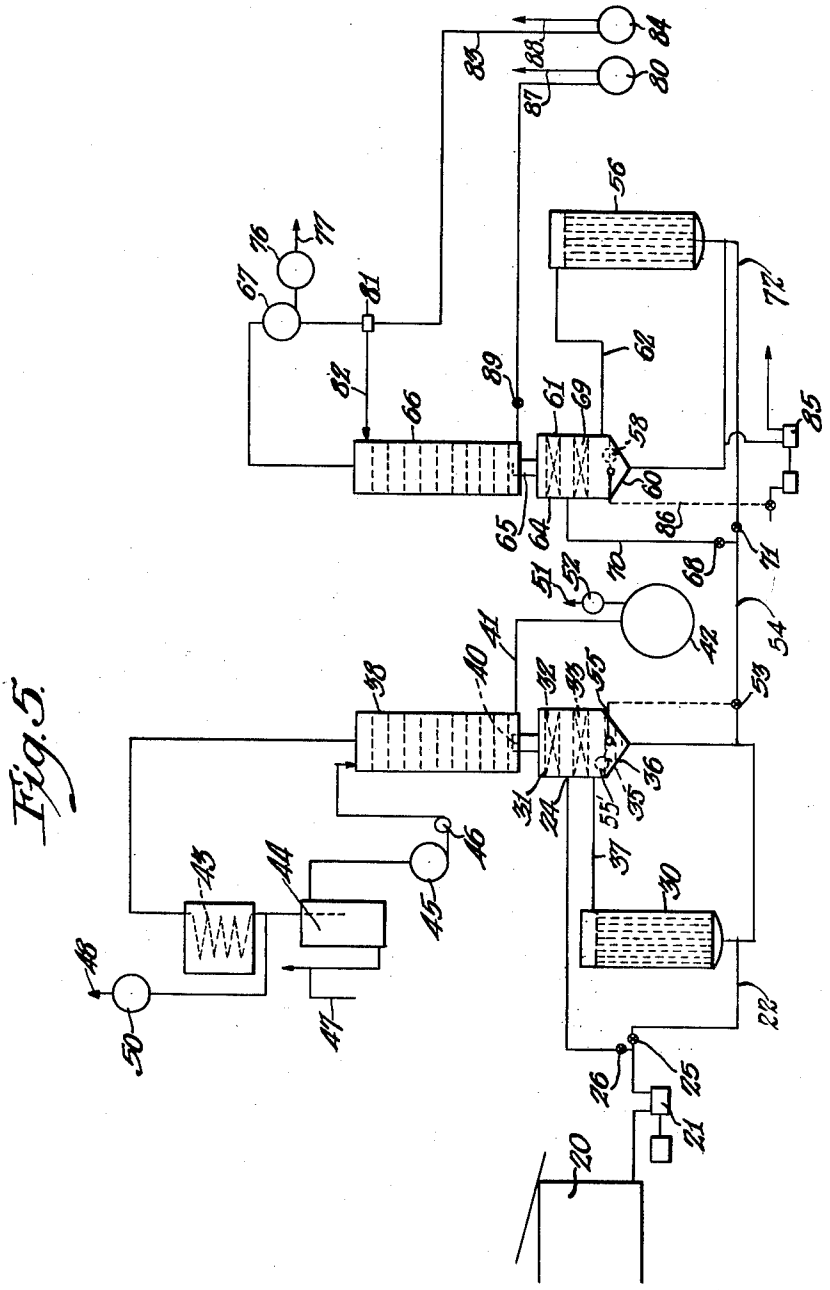

Fig. 5.

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# UNITED STATES PATENT OFFICE

2,673,833

## DISTILLATION OF COAL TAR

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Application April 3, 1951, Serial No. 218,951

16 Claims. (Cl. 196—76)

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The present invention relates to distillation of coal tar for the purpose of dehydrating it and of separating it into distillate oil fractions and a distillation residue.

The present application is a continuation in part of my copending application Serial No. 85,143, filed April 2, 1949, for Distillation of Coal Tar.

A purpose of the invention is to distill coal tar at unusually low temperatures, thereby economizing in the cost of equipment needed for the distillation and in the cost of carrying out the process.

A further purpose is to heat the tar with steam or other condensing vapor, thereby eliminating the hazard of fire always present in fire heated stills.

A further purpose is to recirculate coal tar residue continuously from and to a pool in a vapor box or flash chamber, to introduce tar to be distilled into the recirculating stream, and to heat the stream as it flows back to the pool.

A further purpose is to accomplish the heating of the recirculating stream by passing the coal tar and/or residue therefrom in parallel through a multiplicity of heating paths at a Reynolds number in excess of 2100 and preferably below 25,000, most desirably in the range between 7,000 and 25,000.

A further purpose is preferably to increase the number of multiple paths in parallel in the heating of recirculating tar residue at an intermediate point between the beginning and end of the heating.

A further purpose is to employ a vapor box or flash chamber for tar distillation as above set forth having a chamber size between 0.3 and 1.5 cubic foot per cubic foot of tar through-put per hour.

A further purpose is to produce naphthalene oil containing a comparatively high concentration of naphthalene and to produce other oils containing low concentrations of naphthalene.

A further purpose is to avoid difficulty in vacuum distillation and condensation of naphthalene in the presence of moisture and/or volatile oil, by eliminating water in a first stage to between 0.00 and 0.20 percent and preferably to not over 0.05 percent, and by eliminating volatile oils which boil at atmospheric pressure at a temperature below 200° C. such as will be done when the lightest two to five percent by volume of the tar is removed by distillation.

A further purpose is to distill from the tar and then fractionate a naphthalene oil in a second vacuum stage after removal of water and light oil, condensing the naphthalene oil in a condenser operating at a temperature above 50° C. and below 100° C.

A further purpose is to increase the recovery

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of heat-polymerizable constituents of coal tar in the form of distillate oil.

A further purpose is to produce a tarry residue from coke-oven tar which is of low polymer content, having a specific gravity in excess of 1.23 at 15.5° C. and a softening point of between 90 and 120° F., as determined by the ring and ball method, by heating coke-oven tar to a temperature below 200° C. and removing oils therefrom while the tar is maintained at a pressure not exceeding 60 millimeters of mercury absolute.

A further purpose is to introduce steam in the flash chamber of the first stage.

Further purposes appear in the specification and in the claims.

In the drawings I have chosen to illustrate a few only of the numerous embodiments in which my invention may appear, choosing the forms shown from the standpoints of convenience in illustration, satisfactory operation and clear demonstration of the principles involved.

Figure 1 is a diagrammatic illustration of a tar distillation system to which the invention has been applied.

Figure 1*a* is a variation of Figure 1, showing the change in the first stage flash chamber to admit steam. This can be used with or without the second stage.

Figure 2 is a diagrammatic longitudinal sectional elevation of a heat exchanger which may be employed in accordance with the invention, taken on the line 2—2 of Figure 3.

Figure 3 is a section of Figure 2 on the line 3—3.

Figure 4 is a fragmentary vertical central section of a float-controlled discharge valve for either stage.

Figure 5 is a diagrammatic illustration of a modification of the invention as applied to tar distillation in which the tar is heated in a naturally induced circulating stream. This view is not to be taken as showing the relationships of level required for gravity flow from one point to another in the system.

Describing in illustration but not in limitation and referring to the drawings:

In the prior art numerous efforts have been made to improve the distillation of coal tar, which is commonly carried out in fire-heated stills. Continuous tube stills give some improvement over batch stills but continuous fire-heated stills are comparatively expensive to construct and retain the hazard common to all fire-heated equipment handling hot oils. In general the recovery of naphthalene and other products of high value from the tar has been comparatively incomplete and a considerable amount of polymerization and even pyrolytic destruction of

valuable compounds has occurred in such prior art distillation processes.

The utilization of heat in many of such prior art systems has been comparatively inefficient, and even where extensive heat-exchange equipment has been installed the recovery of heat has often been accomplished at high cost. In an effort to improve the heat economy of tar distillation processes, distillation devices have been based on utilization of waste heat available in hot coke-oven gases but these systems have been comparatively difficult to operate.

Efforts have been made in the prior art to utilize vacuum in coal tar distillation, but these have by no means been completely satisfactory. The problem of vacuum utilization is complicated by the fact that the tar obtained from coke ovens in many cases contains a considerable amount of moisture, which has either necesitated troublesome separate steps for dehydration, or has added to the difficulty of condensing vapors from distillation and limited the degree of vacuum that could be employed.

In accordance with the present invention coal tars are distilled at relatively low temperatures, which preferably will not exceed 200° C. (although in the second stage the temperature may go as high as 275° C.). The minimum distillation temperature normally will be 140° C. Temperatures up to 200° C. are readily obtainable from high pressure steam which is available in most tar producing plants, or, in the case of the higher temperature ranges, from Dowtherm (diphenyl or diphenyl oxide or a mixture thereof in any proportions), or from other vapors used for industrial heating. The temperatures given refer to the maximum temperature to which the liquid tar is heated.

Where such materials are polymerized during the tar heating operation, the resulting larger molecules have far higher boiling points, if indeed they have any boiling points at all. Such polymerized substances therefore remain behind in the residue, and since they are not capable of separation by any practical means, they have low commercial value.

At the lower temperatures at which the present invention is carried out, these constituents are in considerable measure prevented from polymerizing during the heating, so that their boiling points remain in the range of the tar oils that are being vaporized, and so that they are recovered in the distillate oil. In this form they have high commercial value since they can be polymerized effectively later under controlled conditions to make useful products that can be separated from the other constituents of the oils that do not polymerize.

The coal tar of this invention refers to any tar formed from high temperature carbonization of coal and thus refers to "gas-house" or "gas-retort" tar, "vertical-retort" tar, and "coke-oven" tar. While the illustrative examples are given in terms of coke-oven tar, it will be understood that the process of this invention is the same for all of these coal tars as respects temperature and pressure of operation, moisture content of residues, mode of heating and general equipment used to carry out the process, but that differences may exist in the relative yields of fractions and in their precise compositions.

The degree of distillation contemplated by this invention covers the first oil fractions that are distilled from coal tar and which generally amount to not more than the first 30 percent of the tar by volume but may in the case of some light tars run as high as 40 percent of the tar by volume.

Operation in accordance with the present invention under vacuum at low temperatures is very advantageous for several reasons. At high temperatures such as occur during the heating of tar in prior practice, substantial amounts of valuable constituents of the tar are lost from a commercial standpoint by undesirable polymerization.

A further important advantage of the low temperature distillation is that the temperature needed to distill the oil can be produced by using steam as a source of heat, so that the equipment can be efficiently designed and constructed at relatively low cost. The control of the distillation is relatively simple under the low temperature conditions, and the reduction or elimination of fire hazard is a further important advantage.

A further advantage of the low temperature distillation is that all fractions, especially the tar residue, which is the product of greatest bulk, are discharged from the process at correspondingly low temperatures. Consequently less heat energy is stored up as waste heat in the hot products as compared with distillation processes at atmospheric pressure and even those carried out at moderate vacua. In order to obtain a thermal efficiency equivalent to that of the process of the present invention, an atmospheric distillation would require the use of extensive and costly heat-exchange equipment, which is entirely unnecessary in the present process because of the low temperatures employed.

Figure 1 illustrates by way of example one apparatus useful in the present invention. Crude tar from a source **20** is withdrawn by a pump **21** and fed to the distillation system. The rate of feed of the tar is controlled in any suitable manner, as by employing a steam pump in which the steam supply can be adjusted, or by using an automatic pressure actuated control valve, **21***a* as well known. The tar at this point may for example be at a temperature of 50 to 60° C. having a viscosity of 20 to 200 centipoises and a specific gravity of about 1.2, the latter being measured at 15.5° C.

From the feed pump **21**, the tar may enter the first distillation stage through piping at one of several points, typically indicated as **22**, **23** and **24**, the flow to each being controlled by valves **25**, **26**, **27** and **28** in the piping. Point **22** is at the inlet end of a heat exchanger **30**, point **23** is at an intermediate point in the flow through heat exchanger **30** and point **24** is at an intermediate point between the bottom and the top of a vapor box or flash chamber **31**.

When the raw tar is admitted to the vapor box at **24** without passing through the heat exchanger **30**, it may flow in at storage temperature or it may be heated in a preheater **29** before it reaches the vapor box. In either case, it is preferably distributed over packing or baffle plates **33** below its point of entry, to effect a further heating and partial vaporization by the hot vapors rising through the vapor box. Regardless of how the tar enters the vapor box the residue from the new tar feed will join a pool of residue **35** that has been produced from tar previously fed and which is being recirculated through the heat exchanger. The residue of distillation in this first stage is designated as "dehydrated tar" to distinguish it from the tar residue later produced in the second stage, and sometimes it is referred

to simply as "tar" when simply a material of a tarry nature is meant.

Heat required to effect the desired degree of distillation is supplied to the system in the heat exchanger 30, through the tubes of which a stream of dehydrated tar is circulated. The force of gravity may be relied on to cause such circulation as in Figure 5 where tar flowing inside tubes in a vertical position is heated to the point where some of it boils. If the column of mixed liquid and vapor in the tubes is connected with a column of equal height of all liquid outside the tubes the unbalance produced by the difference in weights of the two columns will cause the liquid inside the tubes to flow upwards and the liquid in the column outside the tubes to flow downwards thus causing a circulation as is well known.

It is preferred, however, to produce circulation of the tar by means of the pump 34. This pump is connected on the suction side to the pool of dehydrated tar 35 collected in the depressed bottom 36 of the vapor box or flash chamber 31, so that the pump is constantly drawing off dehydrated tar from the vapor box 31 to recirculate it.

It has been found to be decidedly advantageous to employ a heat exchanger having tar passing through a multiplicity (normally a large number, in excess of 5) of heating paths in parallel. The paths consist of tubes which are surrounded by the heating medium, suitably steam, passed into the shell or housing around the tubes. Each parallel path of the tar may take one or more passes through the tubes in the heat exchanger. Figures 2 and 3 illustrate the heat exchanger diagrammatically, showing tube sheets 38' having tubes 38 and 40 (of a large number of tubes in parallel) on the first pass and tubes 41, 41', 42, and 42' (of a similar large group in parallel) on the return pass, being surrounded by steam or other heating medium and each group being suitably connected together in an inlet header 43, a reverse flow header 44, and an outlet header 43' beyond the tube sheets at the respective ends, as well known. To allow for some vaporization in the heater, without developing excessive back pressure, the number of tubes on the return pass, when more than one pass is used, is desirably increased over those on the forward pass (preferably to about twice).

The rate of heat transfer is greatly improved, while the time of exposure to heat is cut down, and the danger of polymerization or other pyrolytic effects on the tar is still further reduced by constructing the heater so that flow of tar in the tubes occurs in the turbulent range throughout its passage through the heater. To assure turbulent flow the Reynolds number describing the flow in the tubes must be in excess of 2100, and for best results should be in excess of 7,000. While the flow may correspond to a high Reynolds number, say in excess of 100,000, with increasing benefit to heat transfer conditions, generally the maximum value will preferably not exceed 25,000, since if the Reynolds number exceeds 25,000 the pressure drop through the system will tend to be excessive. The Reynolds number is represented by

$$\frac{DG}{\mu}$$

in which D is the diameter of the tube in feet, G is the throughput per tube in pounds/(sq. ft.) (sec.) and $\mu$ is the viscosity (at the existing temperature) of the flowing fluid in pounds/(ft.) (sec.) All units must be consistent as noted.

To maintain turbulent conditions of flow through each tube when a multiplicity of tubes is connected in parallel requires that the tar be circulated and recirculated through the tubes many times. Thus the pumping rate of the circulating pump 34 will be many times the rate of flow of new tar into the system through the pump 21, and it is safe to say that the flow through the circulating pump 34 will always be at least five times, normally will be ten to thirty times the flow of new tar. As previously mentioned gravity may be relied on to cause such circulation although forced circulation from the pump 34 is preferable for the purpose.

Advantage may be obtained of some features of the invention by using a heater of the single continuous coil type as commonly employed in fire-heated tube stills, but the advantages of the multiple-path heater with recirculation of the tar are so marked that it is decidedly preferred in the present invention. Where a single coil heater has a coil long enough to accomplish the heating desired there is commonly an excessively high pressure drop through the heater. Furthermore, this undesirable condition is accentuated by vaporization in the coil which would still further increase the back pressure on the pump.

It should further be noted that in such a continuous coil heater especially when crude tar is run through without recirculation there is a manifold change in viscosity over the course of the heating because of the temperature change from the cold inlet to the hot outlet, and since viscosity is a factor in determining the Reynolds number, a very wide variation in Reynolds number will occur. Thus if the Reynolds number at the inlet were in excess of 2100 so that the flow there would be turbulent, the velocity required to obtain this condition would cause excessively high pressure drop in a single coil heater. On the other hand if the flow at the outlet is reasonably well within the turbulent range the flow at the inlet is likely to be viscous or streamline which will give very poor heat-transfer conditions near the inlet. If we take an example of a tar having a viscosity of 50 centipoises at the inlet and 1 centipoise at the outlet, there will be a 50-fold change in viscosity and a 50-fold change in Reynolds number from inlet to outlet, and if the flow at the inlet were at a Reynolds number just slightly above 2100 and therefore in the turbulent range, the Reynolds number at the outlet would be about 100,000 and the velocity required to attain these conditions would be excessive and would correspond to an excessive pressure drop, whereas if a degree of turbulence corresponding to a Reynolds number of say 10,000 were produced at the outlet of a single coil heater the Reynolds number at the inlet would be not more than 200 and a large part of the flow would be in the streamline range with poor heat transfer.

In the present invention this difficulty is overcome by recirculating a large quantity of heated dehydrated tar through the multiple-path heater. Due to the fact that a large amount of tar mass is already up to temperature, the temperature rise in the circulating tar stream from inlet to outlet of the heat exchanger 30 is small and consequently there is a very small change in Reynolds number due to viscosity change from inlet to outlet, resulting in uniformity of heat transfer conditions from one end to the other of the heater.

Furthermore, in the multiple-path tubular heater described, the circulated tar flows through many tubes in parallel and flows only a comparatively short distance in a single one of the tubu-

lar paths. Thus the pressure drop required to force the tar through the tubes is relatively low. Furthermore, if vaporization occurs within the tubes there is less tendency to build up back pressure as compared with flow of tar through a single coil since the formation of a given amount of vapor based on the new tar feed becomes a proportionately smaller percentage of the total fluid flowing in the tubes when the quantity of circulating tar is many times the quantity of tar feed. Thus heating in multiple parallel paths overcomes several difficulties which have previously been evident in tar heating systems.

It will be possible and advantageous in many cases to supply some of the heat required to the tar while it is in the pool 35 as by jacketing the depressed bottom of the vapor box and even by inserting tubes within the pool and supplying the heating medium to the jacket and to the tubes and this invention does not preclude the use of such auxiliary heat when supplied in this manner but the main heat supply will be obtained as the tar is circulated through the tubes in heat exchanger 30.

The tar which enters the vapor box or flash chamber 31 by any of the routes described will be heated to a minimum temperature that permits vaporization of substantially all the water that is present in the tar. Thus, after the treatment in the vapor box, the residual water in the dehydrated tar will vary between 0.00 percent and 0.20 percent by volume, and preferably will be not more than 0.05 percent by volume. It will be evident to those familiar with the practice of tar dehydration that these moisture limits are very low as compared with industry standards in which a tar containing up to 0.5 percent water by volume is considered to be a dehydrated tar. Many of the chemical compounds contained in tar dissolve substantial amounts of water, and it may be expected, therefore, that tar itself, which is the aggregate of all such compounds, has the capacity to hold some water in true solution. Accordingly there will remain in the dehydrated tar the equilibrium percentage of water corresponding to the partial pressure of water vapor above the tar at the operating temperature in the vapor box.

While the literature states that the dehydration of tar can be accomplished at about 150° C. it has been found necessary in some cases according to the present invention to heat the tar to from 150° C. to 200° C., if the dehydration is accomplished at atmospheric pressure. To accomplish dehydration to the required degree it is decidedly preferable to employ temperatures between 140 and 200° C., and to accompany these temperatures with vacuum, employing an absolute pressure between 50 and 150 mm. of mercury in the vapor box. Along with vaporization of water, light and medium tar oils will be vaporized owing to the steam distillation effect in the vapor box.

While some vaporization of water and oil may occur in the tubes of the heat exchanger 30, especially where new tar is mixed with the circulating tar at the inlet to the heat exchanger or at an intermediate point therein, this vaporization is minimized by the pump pressure and by the small proportion of new tar, and it is desirable that most of the vaporization shall occur in the vapor box 31, which is provided for this purpose. It will be evident that there is a reduction of pressure in the vapor box as compared with the pressure on the tar in the tubes of the heat exchanger 30, and this release in pressure allows the desired amount of vaporization to be completed in the vapor box.

The heated tar together with any vapor therein enters the vapor box preferably tangentially through a pipe 37 at the circumference. The vapor box is normally of a size between 0.3 and 1.5 cubic feet per cubic foot of tar throughput per hour. In other words, if there is a tar throughput of 100 cubic feet per hour, the volume of the vapor box will be of the order of 30 to 150 cubic feet. This relatively large volume serves to allow liquid-phase material to separate from vapor-phase material under the force of gravity and the whirling motion due to the tangential entrance. To further aid in separation of vapor from liquid, baffles or packing 32 are located near the top of the vapor box and above the point 24 if new tar feed is admitted at this point.

Although it is possible to conduct the vapors from the vapor box directly to a condenser, it is preferred, whenever the naphthalene content of the tar is comparatively high to employ a fractionating column 38 above the vapor box and connected thereto by a channel 40. The fractionating column 38 may be a plate, packed or other type column operating in the usual manner with reflux supplied at the top and producing an overhead fraction and a bottoms fraction that are substantially different from each other in composition. The degree of difference between the fractions is determined by the number of equivalent plates in the column and by the ratio of reflux supplied to the vapors passing through. The bottoms fraction is withdrawn from the column through a pipe 41 to a receiver 42.

When tars containing substantial quantities of naphthalene are processed in the manner described, the vapors in the first stage will contain naphthalene in substantial amount even though the total quantity of vapors does not exceed 2 to 5 percent of the tar by volume. When these vapors are condensed to liquids they may tend to deposit crystals if the temperature falls below 60° C. or even below 65° C. in some cases. This may cause serious operating difficulties when the process is under a vacuum because of the necessity to approach these temperatures, or even to cool below them, in order to obtain substantially complete condensation of vapors. Under these circumstances a fractionating column on the first or dehydrating stage is desirable and may be very important since most of the naphthalene in the vapors of the first stage can be collected in the bottom stream of the column and can be withdrawn with this stream into the receiver 42, leaving light oils, that is, oils having boiling points at atmospheric pressure that are mostly below 200° C., and which do not deposite crystals at ordinary temperatures, that is, at 0° to 40° C., to be collected in the overhead stream.

The vapors of water and oil passing overhead from the fractionating column, or leaving the vapor box if the column is not used, are condensed in a condenser 43 cooled in any suitable manner. The condensate passes from the condenser to a decanter 44 from which the oil, usually being the lighter phase, passes to a receiver 45 where a portion of it is picked up by a reflux pump 46 to be returned to the top of the fractionating column as reflux. The water fraction overflows at 47 and may flow to a sewer or may receive further treatment. The detail regarding the treatment of the vapors and condensate may be varied in

accordance with good plant practice without departing from the spirit of the invention. For example the condenser 43 may be composed of a partial condenser and a final condenser, which respectively produce the reflux and condense the "make" of oil and water. In the latter case the pump 46 may prove to be unnecessary since the reflux may flow to the column by gravity. Likewise the separation of the condensate into light oils and water may take place in the receiver 45.

It will be understood of course that if the oil should be heavier than the water the connections in the decanter will be reversed.

A vent is provided at 48 to discharge any non-condensible gas such as may be released from the raw tar when it is heated or which may come into the system by leakage. When the dehydration is carried out under a vacuum, a vacuum pump 50 is employed at this point. It will be evident that the decanter 44 and the receiver 45, if not sufficiently displaced vertically from the rest of the equipment to permit operation at atmospheric pressure, may be equipped with proper seals and vents to enable either or both of them to operate under vacuum. Similarly, receiver 42 is provided with a vent 51 and this has a vacuum pump 52 where vacuum is employed.

In a preferred method of operating the present invention the crude tar flows directly into the vapor box 31. The tar flowed in may be at storage temperature, say 50 to 80° C., or it may be further heated in a preheater 29 before it reaches the vapor box. If preheater 29 is used it should be designed to contain a small volume of tar so that the time that the tar remains in the preheater is short. The preheater will be a "once-through" type in which the tar is heated progressively from inlet to outlet temperature rather than the recirculation type in which the tar receives its heat at practically maximum temperature. While the continuous coil type has some of the disadvantages at discussed above, in the case of the preheater these are not too serious since only about one-third of the total heat required in stage one is added to the tar in the preheater. Furthermore there will be little vaporization in the preheater to cause excessive back pressure. It is not necessary or desirable to preheat the tar excessively since some oils contained in it may polymerize or resinify, and it has been found that 80° to 100° C. is usually sufficient, but higher temperatures may be used if the time of heating is short.

When the tar reaches the vapor box at 24 it is flowed on to baffles or packing material 33 therein and comes into direct contact with vapors arising from the heated recirculated tar which enters the vapor box at 37. Under conditions of temperature and pressure prevailing where the fresh tar flows in, vaporization of water and light oils will commence, if this is not already in process. Vaporization increases as the tar flows down over the packing, being accomplished by thermal interchange with the hot vapors arising from below. When the tar reaches the bottom of the vapor box it joins the pool 35 of dehydrated tar therein, part of which is being continuously withdrawn and recirculated by the pump 34 through the heat exchanger 30 after the manner previously described. The heated tar returns to the vapor box at a point 37 which is above the surface of the pool 35 and which is below the packing 33 over which the fresh tar is distributed. This heating completes vaporization of oils from the tar in the first distillation stage.

It has been found desirable in the first distillation stage to distill an amount of oil that is considerably in excess of the amount that will distill incident to dehydration so that an even greater percentage of the polymerizable and resinifiable constituents of crude tar can be vaporized and removed quickly from a zone of high temperatures and recovered as distillate oil instead of remaining behind in the residue. If the heat-sensitive constituents are not vaporized in this stage some of them may never be vaporized at all, since by the time they reach a later stage of distillation, where temperature and pressure conditions are favorable for their vaporization, they may have become polymerized to form larger molecules, the volatility of which is much lower and may be practically zero. Consequently, to effect a high degree of distillation in the first stage it is desirable to operate at a low pressure and a high finishing temperature which shall be consistent with other requirements and limitations of the process.

In this stage, therefore, the circulating tar will be heated to a temperature between 140 and 200° C. The pressure of distillation depends on the temperature of the cooling water that is available and therefore it may fluctuate with the season of the year. It depends also on the water content of the crude tar. For example, if dry tar be available the pressure can be kept low, say 5 to 20 mm. of mercury, since the oil vapors by themselves can be condensed in this pressure range. Since most coal tar as prepared for distillation contains one or two percent and sometimes more of water, it will generally be necessary to cool the vapors of distillation to the point where the water vapor condenses because if water vapor is not condensed some light oil vapors will not be condensed and will represent a loss to the process.

The relationship between pressure of distillation and cooling-water temperature can be shown by following the vapors released in the vapor box through the fractionating column, in which the rising vapors meet oil reflux, and from thence into the condenser. To condense the water vapor and rectified oil vapors it is necessary to operate at a pressure greater than the sum of the water and oil vapor pressures prevailing at the condenser outlet.

For example, to illustrate numerically, it may be assumed that cooling water is available at 35° C. under extreme warm weather conditions. If the condenser is an indirect, or surface condenser, the distillate vapors may be cooled only to 40° C., at which temperature water has a vapor pressure of 55 mm. The oil vapor associated with the water vapor may be assumed at this point (in order to simplify the illustration) to have the properties of meta-xylene, which has a vapor pressure of 19.5 mm. at 40° C. Thus, the total pressure of the two vapors is about 75 mm. and a pressure greater than 75 mm. will be needed to condense them. Allowing for a pressure drop of 5 mm. in going through the apparatus the pressure in the vapor box where the vapors are released will be about 80 mm.

Similarly, if the condenser temperature is 30° C. the pressure in the vaporization zone will be about 48 mm.; at 20° C. it will be about 29 mm.; and at 10° C. it will be about 18 mm. At temperatures nearer the ice point and with low oil-vapor pressure it will be possible to operate at about 10 mm. With dry tar even lower pressures

can be used as described later in connection with a second stage of distillation.

As previously described the first stage will be operated at temperatures of 140–200° C. in the vapor box, preferably accompanying this temperature with a vacuum, employing as a maximum an absolute pressure between 50 and 150 millimeters of mercury. As shown above, the pressure of the vapors where the tar enters the vapor box at **24** will be at a minimum of about 80 millimeters for unfavorable warm weather conditions. These conditions of temperature and pressure serve adequately to meet the minimum requirements of the first stage distillation, which are to dehydrate the tar and to distill from 2 to 5 percent of the more volatile oils. Somewhat more oil will be distilled when pressures approaching the low of 50 millimeters are combined with temperatures approaching 200° C. However, in the more preferred operation when even lower pressures are used, that is, in the range from 10 to 50 millimeters, it is easily possible to distill more than 5 percent of oil in the first stage and even as much as 20 percent or more. Allowing for unfavorable weather conditions, the pressure range in the first stage may be 10 to 80 millimeters of mercury. For higher pressure operation the pressure may go up to 150 millimeters of mercury, as previously stated.

Furthermore, this can be done without deleterious effect on the tar even though temperatures in the upper part of the range, that is, from 160 to 200° C. are used. This is explained as follows:

In the preferred method of operation the pressure will be in the range from 10 to 50 millimeters which will be possible during about eight or nine months of the year without the use of any special equipment. When the temperature in the lower part of the vapor box is 160–200° C. the temperature at the point where the crude tar enters may be around 140° C. Under these conditions, practically all of the water and from 3 to 8 percent of oil by volume of the crude tar will be flashed off as vapors as soon as the tar enters the vapor box at **24**. In a time interval of less than one second these vapors will pass out of the vapor box into the fractionating column in which the temperature is lower or, if no column is used, they will pass directly to the condenser. Thus, the heat-sensitive constituents in this more volatile 3 to 8 percent or so of the tar will be quickly removed from a high temperature zone to a zone of lower temperatures where the rate of polymerization is slowed down so that the oil fraction containing these constituents can be withdrawn from the equipment before polymerization has taken place.

The vapor box **31** will contain baffles or packing **33**, such as Raschig rings, to the depth of one to five feet, onto which the entering tar is spread. It has been found that hot tar takes about four seconds to travel through one foot of 1-inch rings when they are showered with tar at the rate of two to four gallons per sq. ft. per minute so that the average time that the tar is in contact with these rings is from four to twenty seconds. During its descent through the packing the tar becomes heated and vaporizes an additional 5 to 10% of oil (calculated in terms of the volume of liquid evolved) by the time it reaches the pool of hot tar **35** in the bottom of the vapor box. The tar from which this fraction of oil is distilled will have an average temperature of about 150° C. in the packing section of the vapor chamber and the oils will have been kept at this average temperature for approximately half the time of descent, or from two to ten seconds. Even though the average temperature is such as to cause a rapid rate of polymerization of heat-sensitive constituents in the fraction of tar being vaporized, the time is so short that a substantial percentage of these constituents is able to escape into the cooler zone of the fractionating column, or into the condenser, before polymerization takes place. Thus, the purpose of using the lowest pressure of distillation that will at the same time permit condensation of the vapors in a later step and using also a relatively high temperature in the range from 140 to 200° C. is to distill as much oil as possible in a period of time that is measured in seconds.

When the process of this invention is operated to yield less than ten percent by volume of oil distillate in the first stage the vapors passing into the fractionating column will generally be separated into two fractions as described. One of these will be a low naphthalene oil which does not deposit crystals at condenser temperatures and the other will be a high naphthalene oil having a setting point, which may be in the range from 45° to 65° C., and which is withdrawn from the base of the fractionating column at a temperature between 75° and 145° C.

When the vapors comprise more than ten percent of the tar they will contain substantial quantities of oils boiling above naphthalene and which are collectively known as creosote. It may be advisable in this case to separate the oil vapors into three fractions, one being the light, low-naphthalene fraction that separates in the condenser at a temperature of 40° C. or below, the second being a high naphthalene oil having a setting point between 45 and 65° C. taken off at some midpoint of the column where the temperature is between 50° and 125° C. and the third being a creosote fraction withdrawn from the base of the column where the temperature is at least 30° above that where the naphthalene is withdrawn. The operation of a fractionating column to yield more than two fractions is well-known standard practice and the details are not shown in the drawings. The naphthalene and creosote may be stripped in auxiliary columns, if desired, and the stripped vapors returned to the main fractionating column as is common practice in the operation of columns producing multiple fractions.

The percentage of oil distilled in the first stage may desirably be further increased by adding steam to the vapor box **31** as at **49** in Figure 1*a* where it will have the effect of decreasing the oil vapor pressure, thus permitting more oil to be distilled under any given conditions of temperature and total pressure prevailing in the vapor box. While this steam may be added by injecting it through an open or perforated pipe placed in the pool **35** of dehydrated tar, it will preferably be added as shown to the space in the vapor box directly above the pool of tar, or at some point in the circulating tar stream where it will mix intimately with the tar as it flows through the heater, or part thereof, and be discharged into the vapor box along with the circulating tar and oil vapors.

The steam should secure intimate contact with the hot tar in order to be effective and if, for example, it is admitted to the space above the tar pool, contact may be secured by having the tar from the heater **30** enter the vapor box at a

point somewhat higher up and be distributed over baffle plates or packing 33' similar to 33. An arrangement for accomplishing this result is shown in Figure 1a although other means will occur to persons who are skilled in the art upon becoming familiar with the process.

Steam added in this way will be condensed in condenser 43 along with the steam vaporized from the water originally present in the tar provided that the pressure on the system at this point is more than about 1.4 times the vapor pressure of water at the temperature in the condenser.

The use of added steam in the amount of approximately 0.3 to 1.0 pound per gallon of tar fed to the system may increase the total oil distilled to 20% or more by volume so that a second stage of distillation may be unnecessary in many cases.

When steam is added in the first stage vapor box its partial pressure over the residual tar will be increased over what would otherwise be the case and this will tend to raise the concentration of water left in the residue from distillation but so long as this does not exceed 0.2% by volume of the residue it will have no serious effect on a later stage of distillation. Of course, if all of the desired distillation takes place in a single stage, the concentration of water in the residue becomes of little significance since its only effect is to diminish slightly the heating value of the residue as fuel, or to alter in a minor way some physical characteristic of the residue.

The "make" of dehydrated tar, which corresponds to the raw tar feed minus the vaporization of water and oil, collects in the pool 35 at the bottom of the vapor box 31. In order to prevent accumulation of material at this point, it is conveniently removed automatically. Desirably a valve 53 in a transfer pipe 54 is operated by pivoted float lever 55 having a float 55' in the pool 35, to permit withdrawal of the "make" from the circulating system, as shown in Figure 4. It will be understood of course that any other suitable procedure or mechanism such as a separate pump can be employed to withdraw the "make" from the first distillation stage.

If no further distillation is desired than that which has been accomplished in this stage, the dehydrated tar will be removed from the system. It can be disposed of as fuel, as road tar, as a base for tar paint, or in any other desired manner.

The characteristics of the residue from the first stage of distillation after 10 to 18 percent by volume of oil has been distilled are shown in the following tabulation, which also gives the characteristics of the tar distilled:

| Run No. | 8 | 13 | 15 | 18 |
|---|---|---|---|---|
| Tar Used: | | | | |
| Spec. Grav., 15.5° C. | 1.188 | 1.197 | 1.186 | 1.184 |
| Percent Free Carbon by wt. | 7.36 | 11.07 | 6.77 | 5.92 |
| Percent Water by vol. | 1.5 | 3.0 | 2.7 | 1.8 |
| Percent by Wt. Dist. to 300° C. | 21.59 | 22.46 | 25.14 | 25.22 |
| Engler Spec. Visc., 50° C. | 16.4 | 10.2 | 6.5 | 6.2 |
| Oil Removed by Distillation | 9.9 | 10.0 | 11.6 | 17.9 |
| Residue made: | | | | |
| Spec. Grav., 15.5° C | 1.214 | 1.238 | 1.224 | 1.233 |
| Percent by Wt. Dist. to 300° C | 16.2 | 12.73 | 13.67 | 9.43 |
| Engler Sp. Vis., 70° C | 14.8 | 13.9 | 13.1 | 17.7 |
| Percent Free Carbon, by weight | 7.71 | 11.67 | 8.23 | 8.57 |
| Percent Free Carbon calc. from tar for no increase by weight | 8.31 | 12.70 | 7.90 | 7.38 |
| Increase in percent by weight of free carbon | −0.60 | −1.03 | 0.33 | 1.19 |



The difference in the nature of this residue from that obtained from high temperature processes is shown in the percentage of "free carbon." This refers to the matter insoluble in some solvent, which in this case is benzene. In high temperature distillation the free-carbon content of the distillation residue almost invariably shows an increase over that in the original tar when calculated to the same basis, assuming that the original free carbon in the tar is concentrated owing to removal of oil by distillation. In the present invention in runs 8 and 13, for example, there were decreases respectively of 0.60 and 1.03 in the percent by weight of the free-carbon of the residue. In runs 15 and 18 there were increases of 0.33 and 1.19 percent by weight respectively, but the average for the four runs is a decrease of 0.03 percent, indicating that, on the average, there is no build-up of free carbon when operating according to this invention.

If further distillation is desired, the surplus of tar in the first stage flows into a second stage of distillation where removal of chemical oils such as naphthalene and any constituents boiling below it may be completed and where some creosote may be distilled. If chemical oils have been removed in large part in the first stage, the total oil distilled in the second stage may be collected as a single creosote fraction.

In the second stage, a heat exchanger 56 is employed that is similar structurally to the heat exchanger 30, and heated in the same way (by steam or Dowtherm or other vapor around the tubes). In an actual illustration, it was found that the heat exchanger 56 might have 103 tubes, of which 31 are in the first pass and 72 in the return pass, the number of parallel heating paths therefore increasing more than double in the progress from the inlet to outlet. This permits some vaporization in the heater without developing excessive back pressure. A circulating pump 57 passes tar residue through a circuit including a pool 58 of the tar residue in the depressed bottom 59 of a second stage vapor box or flash chamber 61, from which tar is withdrawn and passed to the suction side of the recirculating pump 57, then passes to the inlet of the heat exchanger 56, and then returning from the outlet of the heat exchanger by a pipe 62 to the vapor box. The vapor box desirably has packing or baffle plates 64 in its upper portion and at the top is connected directly to a condenser or it may, as shown, be connected by a vertical channel 65 with a fractionating column 66. The overhead vapors from the fractionating column, or from the vapor box if a fractionating column is not used, pass to a condenser 67 suitably cooled by means not shown.

The feed to the second stage is withdrawn from the first stage circulating tar stream as by the pipe 54, and may enter the vapor box directly through valve 69 and pipe 70. Alternatively the feed may pass through valve 71, pipe 72 and valve 73 to the inlet to the heat exchanger 56, or may pass through part only of the exchanger as by flowing through valve 71, pipe 72, valve 74 and pipe 75, or it may be heated in any other suitable manner desired as by a heat exchanger 70' placed in the line 70. Where the amount of distillation in the second stage is not over 3 to 5 percent by volume of the tar, this heater may furnish the only heat supply to the second stage and heat exchanger 56 may not be required. When the tar enters the vapor box through the line 70 is may flow on to pack-

ing 69 located in the vapor box 61 below the inlet from pipe 70.

The heat exchanger 56 is desirably of the parallel tube type similar to that shown in Figure 2, and as described in relation to heat exchanger 30. While the single continuous coil type as used in fire-heated stills can be employed, this type is disadvantageous as explained in connection with the heat exchanger 30.

Since the distillation in the second stage is to be carried out at a very low pressure, it is especially desirable to use a heater in the second stage in which the pressure required to overcome fluid friction is small.

The second stage is operated under a suitably high vacuum from a vacuum pump 76 connected to a vent 77. The absolute pressure at the vacuum pump should be in the range from 5 to 50 millimeters of mercury and preferably below about 10 millimeters of mercury. The absolute pressure in the vapor box 61 will be from about 2 to 10 millimeters of mercury higher than that at the inlet to the vacuum pump 76, so that in the vapor box 61 the absolute pressure will range from 7 to 60 millimeters of mercury and preferably be about 12 to 20 millimeters, in order to carry out the distillation. The temperature in the vapor box 61 will be maintained in the range from 160 to 275° C., preferably from 160 to 200° C. With a pressure of 10 to 15 millimeters of mercury and a maximum temperature of 200° C. in the vapor box it has been found that most of the valuable coal-tar chemicals remaining in the dehydrated tar, that is those having boiling points at 235° C. or below at atmospheric pressure, will be vaporized from the tar in the second stage and that the oil recovered from both stages will be 18 to 25 percent of the tar by volume. If a temperature between 200 and 275° C. be used in the second stage, along with an absolute pressure of 15 millimeters of mercury, higher percentages of oil, say 30 to 35%, may be distilled, and a harder residue (similar to roofing pitch) may be obtained. Or, if desired, the chemical oil fraction can be removed at temperatures of about 225° C. so that distillation in the second stage can very well be conducted at 50 millimeters of mercury absolute, instead of 10 to 15 millimeters of mercury.

Oils boiling above 235° C. at atmospheric pressure will also be vaporized to a greater or less extent in the second stage and may be condensed with lower boiling oils as a single fraction if the vapors pass directly from the vapor box to a condenser without use of a fractionating column. However, if the total oils vaporized are collected as one fraction the higher boiling oils will dilute the lower boiling oils and make recovery of chemicals more difficult and more expensive in later operations. Particularly is this true of naphthalene which frequently is isolated from other oils in semi-purified condition by cooling, crystallizing and mechanically separating the naphthalene from the other oils. The latter always retain some naphthalene in solution that is unrecoverable by this method of operation, since even at 5° C. coal-tar oils of this type hold about 20 or 25% naphthalene in solution.

It is the purpose of the fractionating column 66 to effect as far as is practicable a reasonable separation between the two classes of oil before they are withdrawn from the system. The column 66 should be of a design that gives a low pressure drop, and therefore while other designs may be used, a packed column is preferred. Oils boiling above 235° C. at atmospheric pressure are concentrated in the fraction leaving the bottom of the column by pipe 78 and flowing to receiver 80. Generally, from one-third to three-quarters of the total oil vaporized in the second stage may be collected in the fraction withdrawn from the bottom of the column and this fraction contains less than 25 percent by weight, and generally less than 15 percent by weight of naphthalene, thus making unnecessary any further processing for its naphthalene content. Of course, if it is desired to avoid even this small loss of naphthalene, this heavy fraction may, instead of being withdrawn at 78, be allowed to overflow into the vapor box by closing the valve 89, whereupon the naphthalene content of the oil will become revaporized and eventually will work its way out of the system as part of the oil removed from the top of the column.

Oils produced in condenser 67 when a fractionating column is used, have boiling points at atmospheric pressure mostly below 235° C. and will include most of the naphthalene left in the dehydrated tar. Part of this oil may be returned to the fractionating column as reflux through a flow splitter 81 and pipe 82, while the remainder will flow through a pipe 83 to a receiver 84. Alternatively, all of the condensate produced in the condenser 67 may flow to receiver 84 and a portion of the condensate may be returned as reflux to the fractionating column from the receiver 84 in any suitable manner, if desired. As a further alternative method of operation, oil produced in the bottom of column 38 and collected in receiver 42 may be used to furnish all or part of the reflux to column 66.

The "make" of the distillation residue in the second stage, which would tend to accumulate in the pool at the bottom of the vapor box or flash chamber 61, is removed in the manner described for the first stage as by a pump 85 controlled by a float-operated device 86 as well known, responding to a pivoted lever and float in the pool in the vapor box, or by any other suitable mechanism.

A very desirable fuel, such as may be employed in an open hearth furnace, and also a very satisfactory material for road tar, can be produced by the process of the invention carried through the second stage at a temperature below 200° C. The residue processed in this manner is freed of most of its chemical oils, and can be further distilled for production of creosote and pitch. This residue will have a specific gravity at 15.5° C. of not less than 1.23 and a softening point as determined by the ring-and-ball method of between 87 and 120° F. as shown by a large number of tests of which the following are typical. All of these data were obtained when the temperature in the second stage vapor box was 163 to 182° C. and when the absolute pressure was maintained at 14 to 18 millimeters of mercury:

| Test No. | Tar Sample | Total Oil Removed, Percent by Volume | Residue | |
|---|---|---|---|---|
| | | | Specific Gravity, 15.5° C. | Softening Point, ° F. |
| 1 | A | 22.8 | 1.259 | 101.2 |
| 2 | A | 21.8 | | 116.8 |
| 3 | C | 20.9 | | 98 |
| 4 | D | 18.1 | | 90 |
| 5 | E | 18.5 | 1.247 | 91.8 |
| 6 | F | 19.8 | 1.230 | 105 |

In order to produce residue of these general physical properties, fire-heated stills have been used in the prior art which must heat the tar to around 300 to 330° C., in order to distill at atmospheric pressure. Even when using moderate vacuum, such as about 75 millimeters of mercury absolute pressure, the temperatures required are considerably in excess of 200° C. Consequently the prior art residues have had substantially higher polymer contents which are avoided by the residue prepared by using the process of the present invention.

There is no simple analytical method to measure this condition but the free-carbon content is customarily used to show the effect of thermal treatment of tar during distillation. In the present invention one tar used contained 7.36 percent by weight free carbon. The residue of distillation from the second stage contained 8.11 percent by weight free carbon, whereas concentration of the original 7.36 percent by weight owing to removal of oil by distillation would have given 9.35 percent by weight. Other tests have shown similar decreases and still others, slight increases, far less, however, than is obtained when distilling at higher temperatures as in past practice.

The present invention makes it possible to produce tarry residue suitable for use as fuel or as road tar and freed of most of the chemical oils by using steam as a heating medium and heating the tar to temperatures not exceeding 200° C.

My experiments have established that I can produce a partially "topped" tar by removing 10 percent or more of oil by volume and producing not more than 90 percent of distillation residue containing not more than 15 percent by weight distilled to 300° C., and that I can produce a completely "topped" tar by distilling off 15 percent or more of oil by volume, which will contain practically all the light oils and naphthalene, and the distillation residue amounting to 85 percent or less of the tar will contain not more than 7 percent by weight distilled to 300° C.

The importance of moisture control in the first stage and in the feed to the second stage will become apparent when it is realized that when the feed to the second stage contains 0.1 percent water by volume, 8.3 pounds of water are introduced into the second stage with every thousand gallons of feed. After vaporization, water is noncondensible at the temperature and pressure conditions normally prevailing at the outlet from condenser 67. Subsequently the vacuum pump or other vacuum producing means must be large enough to handle all this water vapor plus the unavoidable air leakage into the equipment of the second stage, and if the water in the feed to the second stage is in excess of 0.2 percent by volume, the size of the vacuum equipment may become excessively large.

For the same reason it is desired to remove as distillate in the first stage most of the light tar oils, that is, oils which boil at atmospheric pressure below 200° C., so that residual light oils passing into and vaporized in the second stage will be small in amount and their uncondensed vapors will not add greatly to the burden of the vacuum pump. Of course if a source of dehydrated tar is available, that is, containing 0.00 to 0.20 percent of water by volume and preferably not more than 0.05 percent by volume as defined in this invention, the use of a first distillation stage for dehydration would be unnecessary and the tar could be fed directly from its source into a single stage operation that would correspond substantially to second stage operation as described herein.

It will be evident that in case the receivers 80 and 84 do not themselves provide vacuum seals, vacuum seals or their equivalent will be supplied on the discharge pipes 87 and 88.

Most coal tars of 1948 production contain a large proportion of naphthalene under United States and Canadian practice. It is possible according to this invention to concentrate naphthalene in the oils condensed in the condenser 67 so that this fraction will have a setting point of 45° to 60° C. or even higher, denoting 50 percent by weight or more of naphthalene in the product. The condenser should not be operated below the temperature at which naphthalene will deposit as a solid phase from the oil since there would be danger of solidification of naphthalene which would block the equipment and hence allowing for a 5° C. safe operating margin, the temperature of 50° C. is the minimum at which the condenser should be operated when a naphthalene oil having a setting point of 45° C. is being fractionated from the vapors of the second stage.

Since at 70° C. the vapor pressure of naphthalene is about 4 millimeters of mercury, the naphthalene-oil vapor concentration in the gases leaving the condenser 67 will be 40 mol percent when the system is being operated at a pressure of 10 millimeters if the vapor pressure of the oil is assumed to be substantially the same as that of pure naphthalene. Similarly, at 100° C. the vapor pressure of naphthalene is 13.5 millimeters of mercury and if the system were operated at 50 millimeters the naphthalene oil vapor would amount to about 37 mol percent of the total vapors. Thus the condenser should operate at a minimum temperature of 50° C., desirably at a temperature not exceeding 70 to 80° C. when a naphthalene oil is being fractionated, and in any case at a maximum of 100° C. to prevent solidification of naphthalene on the one hand and excessive vapor losses on the other hand.

Most of the naphthalene that passes through the condenser as vapor can be removed from the gases by dissolving it in oil in a scrubber, which may be used in the present invention, but which forms no part of it. The oil used for this purpose may be petroleum oil as is used for scrubbing coal gas, for recovery of coke-oven light oils such as toluene, and is therefore readily available at coke plants, or it may be a coal-tar fraction of low naphthalene content such as the fraction collected in the receiver 80 (the bottom fraction in the second stage), or it may be the oil left after chilling oil containing naphthalene to crystallize and remove the naphthalene, and which is known as "drain oil."

In operation it will be evident that the heater in the first stage which is operating on the recirculating stream drawn from the pool will heat to a temperature of about 200° maximum, the minimum temperature being about 140° C. Preferably a multiplicity of parallel paths is provided in both heaters through which the distillation residues are recirculated and both heaters operate at Reynolds numbers in excess of 2100 and preferably between 7,000 and 25,000 assuring turbulent flow with good heat transfer rates. In both heaters the number of parallel paths preferably increases as the flow takes place through the heater (except with gravity circulation), thus permitting some vaporization to occur in the heaters without developing excessive back pressure. The increase in the number of parallel

paths may occur at one point or at a plurality of points in each heater. The tar feed to either stage may be introduced to the recirculating stream in that stage at the inlet to the heater, part way through the heater, or in the vapor box of that stage and the tar feed to either stage may be separately heated before it enters either stage.

In both stages there is a reduction in pressure in the flash chamber as compared with the corresponding heater. While the first stage vapor box may operate at atmospheric pressure, it will desirably be at a maximum absolute pressure of between 50 and 150 millimeters of mercury and more desirably at an absolute pressure of between 10 and 80 millimeters. The second stage will have an absolute pressure in the vapor box between 7 and 60 millimeters of mercury and at the vacuum pump of between 5 and 50 millimeters of mercury, preferably not exceeding 10 millimeters of mercury.

The first stage will remove water very completely, so that the water content in the "dehydrated" tar withdrawn from the first stage will be between 0.00 and 0.20 percent by volume and preferably not in excess of 0.05 percent by volume, and the first stage will remove most of the light oils boiling at atmospheric pressure below 200° C. by distilling off as a minimum from 2 to 5 percent of the tar by volume in this stage. Desirably the first stage will distill more than 10 percent by volume of oil and up to about 20 percent or more by volume.

Both vapor box sizes will be large, and preferably in the range between 0.3 and 1.5 cubic feet of vapor box space per cubic foot of tar throughput per hour.

The temperature in the vapor box of the second stage will range from 160 to 275° C. and preferably from 160 to 200° C. The vapors from either stage will be condensed to form a single oil fraction in either stage or they will be fractionated to form more than one oil fraction in either stage. When producing a naphthalene oil, the condenser temperature in the second stage will be not less than 50° C. so that naphthalene will not solidify in the condenser.

In view of my invention and disclosure variations and modifications to meet individual whim or particular need will doubtless become evident to others skilled in the art to obtain all or part of the benefits of my invention without copying the process and structure shown, and I, therefore, claim all such insofar as they fall within the reasonable spirit and scope of my claims.

Having thus described my invention what I claim as new and desire to secure by Letters Patent is:

1. The process of dehydrating coal tar containing a substantial amount of naphthalene and separating it into fractions, which comprises continuously feeding coal tar into a flash chamber of a first stage maintained at an absolute pressure of 10 to 150 millimeters of mercury and there flashing water and oil into vapor and producing a pool of dehydrated tar, condensing at a temperature below 65° C. an oil fraction that does not deposit crystals at temperatures between 0° and 40° C., continuously withdrawing dehydrated tar from the pool, continuously heating the tar withdrawn to a temperature between 140 and 200° C. and reconducting the tar thus heated to the flash chamber, the dehydrated tar having a water content of between 0.00 and 0.20 percent by the volume, diverting accumulation of dehydrated tar from the pool and passing it into a flash chamber of a second stage maintained at a temperature of between 160 and 275° C. and at an absolute pressure between 7 and 60 millimeters of mercury, there vaporizing volatile components of the dehydrated tar into vapor, condensing at a temperature above 50° C. a portion of the vapor as a naphthalene fraction having a setting point of 45° C. or higher and removing the accumulation of residue from the second stage.

2. The process of dehydrating coal tar containing a substantial amount of naphthalene and separating it into fractions, which comprises continuously feeding coal tar into a flash chamber for a first stage maintained at an absolute pressure of 10 to 150 millimeters of mercury and there flashing water and oil into vapor and producing a pool of dehydrated tar, condensing at a temperature below 65° C. an oil fraction that does not deposit crystals at temperatures between 0° and 40° C., continuously withdrawing dehydrated tar from the pool, continuously heating the tar withdrawn in a multiplicity of parallel paths at a Reynolds number greater than 2100 to a temperature between 140 and 200° C. and reconducting the tar thus heated to the flash chamber, the dehydrated tar having a water content of between 0.00 and 0.20 percent by volume, drawing off accumulation of dehydrated tar and passing it into a flash chamber of a second stage maintained at a temperature of between 160 and 275° C. and at an absolute pressure of 7 to 60 millimeters of mercury and there vaporizing a portion of the dehydrated tar, condensing at a temperature above 50° C. a portion of the vapor as a naphthalene fraction having a setting point of 45° C. or higher, and removing the accumulation of the residue from the second flash chamber.

3. The process of dehydrating coal tar containing a substantial amount of naphthalene and separating it into fractions, which comprises continuously feeding coal tar into a flash chamber for a first stage maintained at an absolute pressure of 10 to 150 millimeters of mercury and there flashing water and oil into vapor and producing a pool of dehydrated tar, continuously withdrawing dehydrated tar from the pool, continuously heating the tar withdrawn in a multiplicity of parallel paths at a Reynolds number greater than 2100 to a temperature between 140 and 200° C. and reconducting the tar thus heated to the flash chamber, the dehydrated tar having a water content of between 0.00 and 0.20 percent by volume, conducting the vapor from the first stage into a fractionating column and there producing at a temperature below 65° C. an overhead fraction that does not deposit crystals at temperatures between 0° and 40° C., forming within the fractionating column at a temperature between 75 and 145° C. a naphthalene fraction, withdrawing this fraction from the column, drawing off accumulation of dehydrated tar from the first stage and passing it into a flash chamber of a second stage maintained at a temperature of between 160 and 275° C. and at an absolute pressure of 7 to 60 millimeters of mercury and there vaporizing a portion of the dehydrated tar, condensing the vapors formed, and removing the accumulation of the residue from the second flash chamber.

4. The process of dehydrating coal tar containing a substantial amount of naphthalene according to claim 3, which comprises introducing 0.3 to 1.0 pound of steam per gallon of tar feed into the distillation in the first stage.

5. The process of distilling coal tar containing a substantial amount of naphthalene, which comprises flowing the tar at a predetermined rate to a first stage of distillation in which the tar is to be separated into volatile fractions and dehydrated tar, continuously recirculating dehydrated tar from the first stage and continuously heating the dehydrated tar as it is recirculated in a multiplicity of parallel paths at a Reynolds number greater than 2100 and at a temperature between 140 and 200° C., flowing the heated material into a first zone for separating vaporized material from dehydrated tar while maintaining this zone at an absolute pressure of between 10 and 150 millimeters of mercury, condensing at a temperature below 65° C. an oil fraction that does not deposit crystals at temperatures between 0° and 40° C., flowing the accumulation of dehydrated tar, which contains from 0.00 to 0.20 percent of water by volume, from the first stage into a second stage containing a second zone for separating vapors from residue of distillation of the second stage, recirculating the residue from the second stage through a multiplicity of parallel heating paths and heating the residue to a temperature of between 160 and 200° C. while passing it through the multiplicity of parallel heating paths at a Reynolds number greater than 2100, flowing the heated residue into the second zone for separating vapors from distillation residue while maintaining this zone at an absolute pressure of 7 to 60 millimeters of mercury, condensing at a temperature above 50° C. a portion of the vapor as a naphthalene fraction having a setting point of 45° C. or higher, and removing the accumulation of residue from the second stage.

6. The process of distilling coal tar containing a substantial amount of naphthalene, which comprises flowing the tar at a predetermined rate to a first stage of distillation in which the tar is to be separated into volatile fractions and dehydrated tar, continuously recirculating dehydrated tar from the first stage and continuously heating the dehydrated tar as it is recirculated in a multiplicity of parallel paths at a Reynolds number greater than 2100 and at a temperature between 140 and 200° C., flowing the heated material into a first zone for separating vaporized material from dehydrated tar while maintaining this zone at an absolute pressure of between 10 and 150 millimeters of mercury, conducting the vapor from the first stage into a fractionating column and there producing at a temperature below 65° C. an overhead fraction that does not deposit crystals at temperatures between 0° and 40° C., forming within the fractionating column at a temperature between 75 and 145° C. a naphthalene fraction having a setting point in excess of 45° C., withdrawing this fraction from the column, flowing the accumulation of dehydrated tar, which contains from 0.00 to 0.20 percent of water by volume, from the first stage into a second stage containing a second zone for separating vapors from residue of distillation of the second stage, recirculating the residue from the second stage through a multiplicity of parallel heating paths and heating the residue to a temperature of between 160 and 200° C. while passing it through the multiplicity of parallel heating paths at a Reynolds number greater than 2100, flowing the heated residue into the second zone for separating vapors from distillation residue while maintaining this zone at an absolute pressure of 7 to 60 millimeters of mercury, condensing the vapor from the second stage, and removing the accumulation of residue from the second stage.

7. The process of distilling coal tar containing a substantial amount of naphthalene to separate moisture and volatile oil fractions, which comprises flowing the tar to a first stage of distillation maintained at an absolute pressure of 10 to 150 millimeters of mercury and there separating vapors and forming a pool of distillation residue, withdrawing dehydrated tar from the pool and flowing it through a multiplicity of parallel heating paths at a Reynolds number greater than 2100 and therein heating to a temperature of 140 to 200° C., fractionating the vapors from the first stage to produce at a temperature below 65° C. an overhead fraction which does not deposit crystals at temperatures between 0° and 40° C., flowing the accumulation of dehydrated tar from the first stage to a second stage and there separating vapors and forming a pool of distillation residue while maintaining an absolute pressure at the point of separation which is always between 7 and 60 millimeters of mercury, withdrawing distillation residue from the pool and flowing it through a multiplicity of parallel heating paths at a Reynolds number greater than 2100 and therein heating to a temperature of 160 to 275° C., fractionating the vapors from the second stage while under reduced pressure and concurrently supplying a flow of reflux liquid countercurrent to the vapors, condensing a naphthalene fraction at a temperature above 50° C. and which has a setting point of 45° C. or higher, withdrawing a heavier fraction containing less than 25 percent by weight of naphthalene, withdrawing noncondensible vapors, and removing the accumulation of residue from the second stage.

8. The process of removing water from coal tar containing a substantial amount of naphthalene and separating the latter into fractions, which comprises recirculating a stream of dehydrated tar in a first stage, heating the stream to a temperature of between 140 and 200° C. while it is progressed through a multiplicity of parallel heating paths at a Reynolds number greater than 2100, introducing the tar residue thus heated into a flash chamber maintained at an absolute pressure of 10 to 150 millimeters of mercury and having a volume of between 0.3 and 1.5 cubic foot per cubic foot of through-put of tar per hour, condensing at a temperature below 65° C. an oil fraction that does not deposit crystals at temperatures between 0° and 40° C., recirculating a stream of tar residue in a second stage, introducing the accumulation of dehydrated tar from the first stage into the recirculating stream from the second stage, heating the recirculating stream of tar residue in the second stage to a temperature of between 160 and 200° C. in a plurality of parallel heating paths at a Reynolds number greater than 2100, introducing the heated stream in the second stage into a flash chamber maintained at an absolute pressure of 7 to 60 millimeters of mercury and having a volume of between 0.3 and 1.5 cubic foot per cubic foot of through-put of tar per hour, condensing a naphthalene fraction from the second stage at a temperature above 50° C. and removing the accumulation of residue from the second stage.

9. The process of distilling coal tar containing a substantial amount of naphthalene and separating it into fractions, which comprises con-

tinuously feeding coal tar into a flash chamber maintained at an absolute pressure of 10 to 150 millimeters of mercury and there flashing oil vapors and producing a pool of distillation residue, continuously withdrawing distillation residue from the pool, continuously heating the distillation residue withdrawn in a multiplicity of parallel paths at a Reynolds number greater than 2100 to a temperature between 140 and 200° C. and reconducting all of the material making up the residue thus heated to the same flash chamber where it joins the pool of residue previously formed and vapors therefrom join the vapors from the tar previously fed to the flash chamber, conducting the vapors from the flash chamber into a fractionating column and there producing at a temperature below 65° C. an overhead fraction that does not deposit crystals at temperatures between 0° and 40° C., forming a naphthalene fraction within the fractionating column at a temperature between 75 and 145° C., withdrawing this fraction from the column, and drawing off accumulation of distillation residue from the flash chamber.

10. The process of distilling coal tar containing a substantial amount of naphthalene and separating it into fractions, which comprises continuously feeding coal tar into a flash chamber maintained at an absolute pressure of 7 to 60 millimeters of mercury and there flashing oil vapors and producing a pool of distillation residue, continuously withdrawing distillation residue from the pool, continuously heating the distillation residue withdrawn in a multiplicity of parallel paths at a Reynolds number greater than 2100 to a temperature between 160 and 275° C. and reconducting all of the material making up the residue thus heated to the same flash chamber where it joins the pool of residue previously formed and vapors therefrom join the vapors from the tar previously fed to the flash chamber, conducting the vapors from the flash chamber into a fractionating column and there producing at a temperature below 65° C. an overhead fraction that does not deposit crystals at temperatures between 0° and 40° C., forming a naphthalene fraction within the fractionating column at a temperature between 75 and 145° C., withdrawing this fraction from the column, forming a heavier fraction containing less than 25 percent by weight of naphthalene, withdrawing this heavier fraction from the column, and drawing off accumulation of distillation residue from the flash chamber.

11. The process of distilling coal tar containing a substantial amount of naphthalene according to the process of claim 9, which comprises introducing into the distillation 0.3 to 1.0 pounds of steam per gallon of tar fed into the flash chamber.

12. The process of distilling coal tar containing a substantial amount of naphthalene, which comprises flowing the tar at a predetermined rate to a first stage of distillation containing a first zone for separating volatile vapors from dehydrated tar and there flashing water and oil into vapor and producing a pool of dehydrated tar while maintaining this zone at an absolute pressure of 10 to 150 millimeters of mercury and at a temperature below 200° C., condensing at a temperature below 65° C. an oil fraction that does not deposit crystals at a temperature between 0° and 40° C., flowing the accumulation of dehydrated tar from the first stage into a second stage containing a second zone for separating vapors from residue of distillation of the second stage, recirculating residue from the second stage through a multiplicity of parallel heating paths and heating the residue to a temperature of between 160 and 275° C. while passing it through the multiplicity of heating paths at a Reynolds number greater than 2100, flowing the heated residue into the second zone for separating vapors from distillation residue while maintaining this zone at an absolute pressure of 7 to 60 millimeters of mercury and at a lower absolute pressure than the first zone, condensing at a temperature above 50° C. and below 145° C. a naphthalene fraction having a setting point of 45 C. or higher, condensing and withdrawing a heavier fraction containing less than 25 percent by weight of naphthalene, and removing the accumulation of residue from the second stage.

13. The process of distilling coal tar containing a substantial amount of naphthalene, which comprises flowing the tar at a predetermined rate to a first stage of distillation containing a first zone for separating volatile vapors from dehydrated tar and there flashing water and oil into vapor and producing a pool of dehydrated tar while maintaining this zone at an absolute pressure of 10 to 150 millimeters of mercury and at a temperature below 200° C., condensing at a temperature below 65° C. an oil fraction that does not deposit crystals at a temperature between 0° and 40° C., flowing the accumulation of dehydrated tar from the first stage into a second stage containing a second zone for separating vapors from residue of distillation of the second stage, recirculating residue from the second stage through a multiplicity of parallel heating paths and heating the residue to a temperature between 140 and 200° C. while passing it through the multiplicity of heating paths at a Reynolds number greater than 2100, flowing the heated residue into the second zone for separating vapors from distillation residue while maintaining this zone at an absolute pressure of 7 to 60 millimeters of mercury and at a lower absolute pressure than the first zone, condensing at a temperature above 50° C. and below 145° C. a naphthalene fraction having a setting point of 45° C. or higher, and removing the accumulation of residue from the second stage.

14. The process of distilling coal tar containing a substantial amount of naphthalene, which comprises flowing the tar at a predetermined rate to a first stage of distillation, containing a first zone for separating volatile vapors from dehydrated tar, and there flashing water and oil into vapor and producing a pool of dehydrated tar while maintaining this zone at an absolue pressure of 10 to 150 millimeters of mercury and at a temperature below 200° C., condensing at a temperature below 65° C. an oil fraction that does not deposit crystals at a temperature between 0° and 40° C., flowing the accumulation of dehydrated tar from the first stage into a second stage containing a second zone for separating vapors from residue of distillation of the second stage, while maintaining this zone at a temperature of between 160 and 275° C. and at an absolute pressure of 7 to 60 millimeters of mercury but at a lower absolute pressure than the first zone, condensing at a temperature above 50° C. and below 145° C. a naphthalene fraction having a setting point of 45° C. or higher, condensing and withdrawing a heavier fraction containing less than 25 percent by weight of naph-

thalene and removing the accumulation of residue from the second stage.

15. The process of distilling coal tar containing a substantial amount of naphthalene, which comprises flowing the tar at a predetermined rate to a first stage of distillation containing a first zone for separating volatile vapors from dehydrated tar maintained at an absolute pressure of 10 to 150 millimeters of mercury and at a temperature below 200° C. and there flashing water and oil into vapor and producing a pool of dehydrated tar, condensing at a temperature below 65° C. an oil fraction that does not deposit crystals at a temperature between 0° and 40° C., flowing the accumulation of dehydrated tar from the first stage into a second stage containing a second zone for separating vapor from residue of distillation of the second stage, while maintaining this zone at a temperature of between 140 and 200° C. and at an absolute pressure of between 10 and 80 millimeters of mercury, condensing at a temperature above 50° C. and below 145° C. a naphthalene fraction having a setting point of 45° C. or higher, and removing the accumulation of residue from the second stage.

16. The process of distilling coal tar containing a substantial amount of naphthalene, which comprises heating coal tar at a temperature below 200° C. and distilling off water and oil vapor and producing a residue of dehydrated tar, flowing



the accumulation of dehydrated tar from the dehydrating operation into a stage of distillation containing a zone for separating vapors from residue of distillation, recirculating residue from this stage through a multiplicity of parallel heating paths, heating the residue to a temperature between 160 and 275° C. while passing it through the multiplicity of heating paths at a Reynolds number greater than 2100, flowing the heated residue into the said zone for separating vapors from residue of distillation while maintaining this zone at an absolute pressure of 7 to 60 millimeters of mercury, condensing at a temperature above 50° C. and below 145° C. a naphthalene fraction having a setting point of 45° C. or higher, condensing and withdrawing a heavier fraction containing less than 25 percent by weight of naphthalene, and removing the accumulation of residue from the stage of distillation.

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**References Cited** in the file of this patent

UNITED STATES PATENTS

| Number | Name | Date |
|---|---|---|
| 2,029,883 | MacCubbin et al. | Feb. 4, 1936 |
| 2,066,386 | Bergeim | Jan. 5, 1937 |
| 2,260,072 | Wilton | Oct. 21, 1941 |
| 2,310,500 | Zavertnik | Feb. 9, 1943 |
| 2,366,900 | Weir | Jan. 9, 1945 |