July 19, 1932. S. P. MILLER 1,868,394

METHOD FOR DISTILLATION OF TAR

Filed June 4, 1927 2 Sheets-Sheet 1

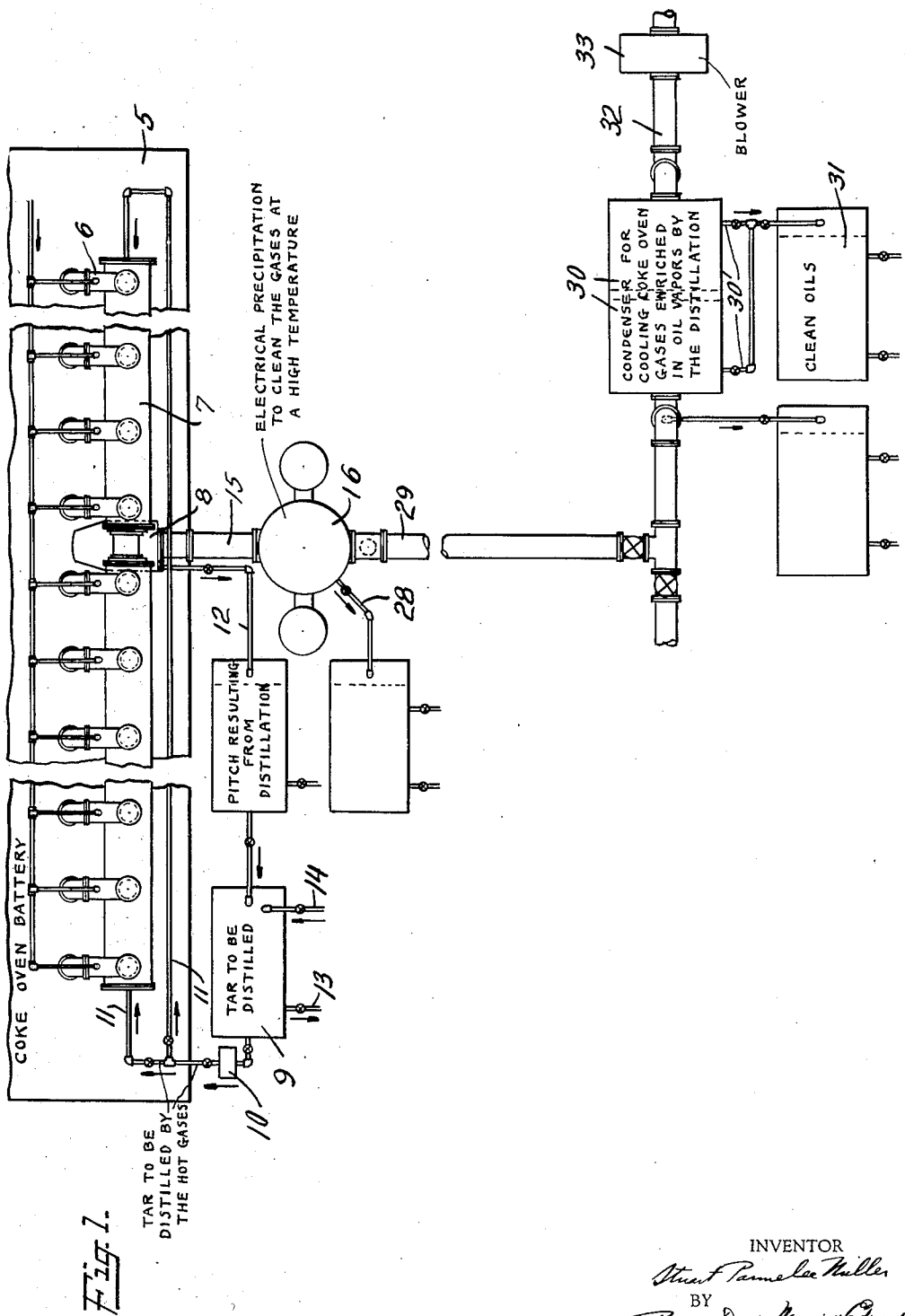


Fig. 1.

INVENTOR
Stuart Pamela Miller
BY
Pennie Davis Marvin & Edmonds
ATTORNEYS

July 19, 1932. S. P. MILLER 1,868,394

METHOD FOR DISTILLATION OF TAR

Filed June 4, 1927 2 Sheets-Sheet 2

Fig. 2.

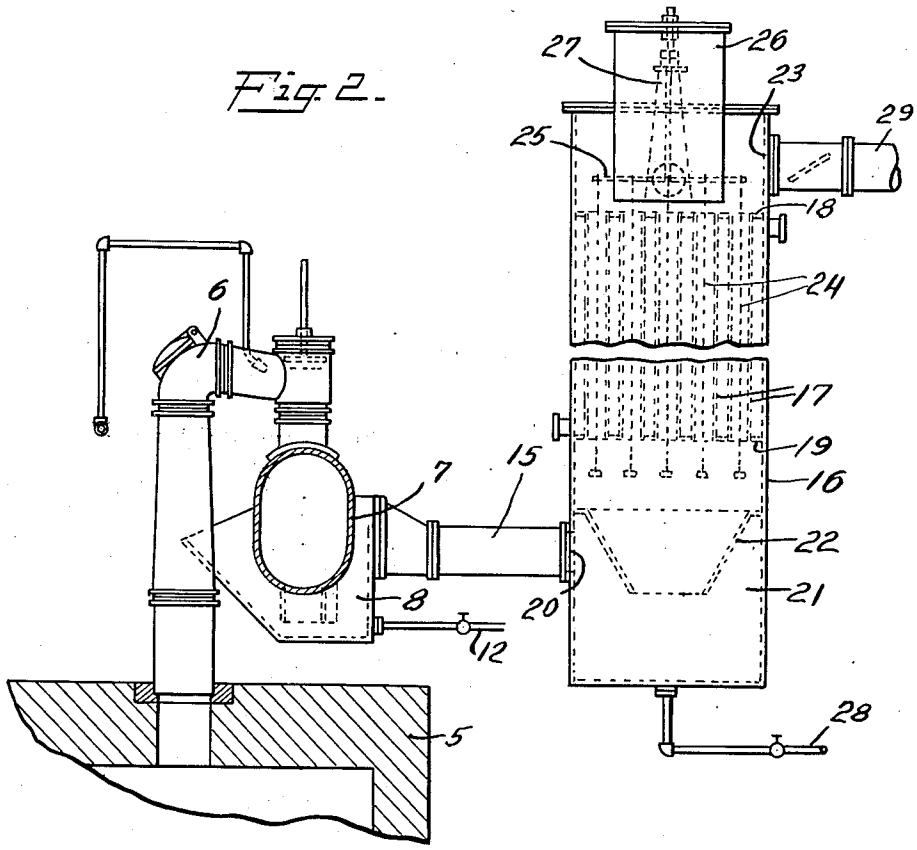
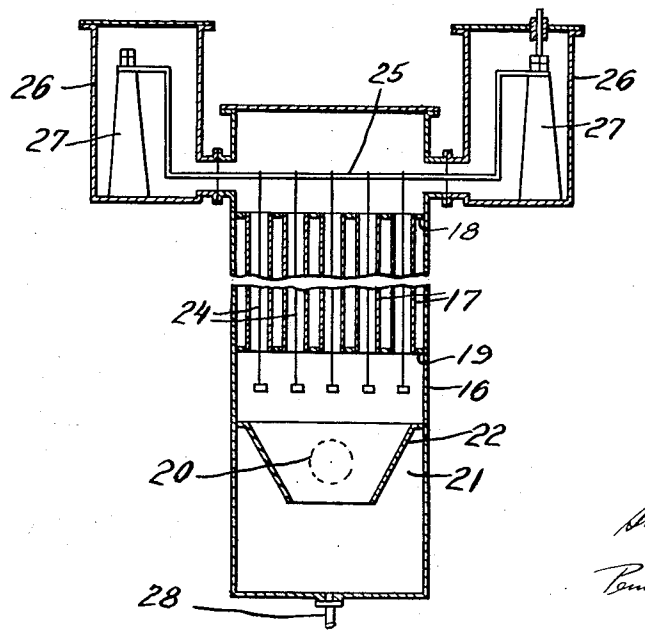


Fig. 3.



INVENTOR
Stuart Pamelee Miller
BY
Pennie Davis Marvin & Edmonds
ATTORNEYS

Patented July 19, 1932

1,868,394

UNITED STATES PATENT OFFICE

STUART PARMELEE MILLER, OF TENAFLY, NEW JERSEY, ASSIGNOR TO THE BARRETT COMPANY, OF NEW YORK, N. Y., A CORPORATION OF NEW JERSEY

METHOD FOR DISTILLATION OF TAR

Application filed June 4, 1927. Serial No. 196,486.

This invention relates to improvements in the distillation of tar and the production of pitches and distillate oils therefrom, and includes a new method of tar distillation. More particularly the invention relates to the recovery of desirable products from coal-carbonization and distillation plants such, for example, as by-product coke ovens.

In the ordinary operation of by-product coke ovens the gases produced by the coking operation pass from the individual ovens through uptake pipes and goose-necks to a collector main common to the ovens of the battery. The coke-oven gases which leave the ovens at temperatures from 600° to 700° C. or higher are cooled in the collector main and in condensers connected therewith to separate tar constituents therefrom. The separated coal tar is commonly shipped to tar-distillation plants where it is subjected to distillation to produce pitches and various other products.

Owing to the high temperature of the gases from the coke ovens, and particularly because of the enormous amount of heat carried thereby, the successful operation of the collector main in connection with these ovens has always been a serious problem. The heat carried by the gases is from twenty to thirty-five times that required for the distillation of all of the tar contained in the gases. A certain amount of heavy tar or pitch may separate in the collector main and such tar or pitch is subjected to the distilling effect of the gases at high temperature with the resulting formation in the main of solid hard pitch which can in some cases be removed only by shutting down the plant and scraping or digging it out. It is customary to flush the main with a mixture of tar and ammonia liquor or with ammonia liquor alone, but this practice does not always avoid the difficulty. In fact, the use of water or ammonia liquor aggravates the trouble in some cases because the pitches formed in the main are insoluble in water or ammonia liquor and hence are not flushed out unless an extremely large amount of the liquor is employed.

The use of a large quantity of ammonia liquor to flush the collector main introduces a new difficulty because of the resulting lowered temperature of the gases. Thus, by spraying ammonia liquor into the collector main in sufficient quantity to prevent accumulations of pitch therein the temperature of the gases may be so reduced that a considerable proportion of the valuable oil constituents which are present in the gases is condensed and thrown down with the tar and can be removed therefrom only by subsequent distillation.

It is the object of the present invention to provide an improved method of and apparatus for tar distillation directly at the coal-carbonization plant such as a by-product coke-oven plant, and especially an improvement in the operation of the collector main whereby advantage can be taken of the heat carried by the coke-oven gases to distill tar and oils and to separate volatile products therefrom so that the latter can be recovered independently of the pitch which then constitutes a separate product of the operation.

A further object of the invention is the separation from the hot gases of all solid and tarry constituents, leaving only the clean oil vapor therein which, upon condensation, yields clean oils which may be utilized in the trade without further distillation or other treatment.

According to the present invention the hot coke-oven gases as they come from the coke ovens are utilized for the distillation of tar and the separation of vaporizable oils therefrom by passing the gases through the collector main and by maintaining therein a large and constantly changing body of the tar to be distilled. This tar can be introduced to the collector main and circulated therethrough in sufficient volume to prevent overheating thereof and the consequent formation of solid hard pitch in the main. In circulating through the main the temperature of the tar is raised owing to its contact with the hot coke-oven gases flowing therethrough and distillation of the tar is effected so that the more volatile constituents are separated therefrom and are carried away with the hot gases. The pitch resulting from

the distillation may be withdrawn from the main and recirculated therethrough for further heating and distillation. The amount of distillation effected may be regulated by the extent to which the tar and pitch are subjected to the heating effect of the gases, and consequently the character of the pitch produced can be modified as desired to produce saleable pitch products.

The tar and pitch flowing through the collector main will be heated by the hot gases and their temperature may be raised considerably. Part of the heat is employed, however, in vaporizing oil constituents of the tar without corresponding increase in temperature thereof so that the maximum temperature of the tar and of the resulting pitch may be considerably below that of the gases. To avoid pitching of the main it is important that a large volume of tar or pitch be introduced thereto and that the tar or pitch be circulated actively through the main so as to prevent any accumulation of hard solid pitch therein. The heat available in the gases is such as to ensure the heating of the tar or pitch to a temperature at which it is thinly fluid so that it can be circulated easily. Moreover, the hot tar or pitch readily dissolves any deposits of solid hard pitch in the main and accumulation thereof is thus avoided.

Although indiscriminate cooling of the gases in the collector main as heretofore practiced is to be avoided, it may be desirable to introduce water or ammonia liquor with the tar or pitch for the purpose of cooling the gases to a predetermined and regulated extent. The regulation of the cooling effect can be accomplished more readily with water because of the higher specific and latent heats of water as compared with those of tar and oils. It is possible, therefore, to add water or ammonia liquor in the proportion required to remove all heat except that needed for the distillation of the tar. When harder pitches are desired less water or none at all can be added. Cooling of the gases to a regulated extent can be accomplished also by the introduction of volatile oil with the tar. Thus, a proportion of the oil recovered from the gases can be returned to the collector main and mingled with the tar or pitch therein. The distillation of this oil in the collector main will absorb heat from the gases and thus permit the maintenance of the desired temperature therein.

The distilling capacity of the gases from a single coke-oven battery is greatly in excess of that required for the distillation of the amount of tar which a single coke-oven battery produces so that the tar from a considerable number of coke-oven batteries can be distilled by the hot gases from a single battery. Consequently to maintain the operation as hereinbefore described, it is ordinarily necessary to provide an amount of tar very much in excess of that produced by the single battery which is so operated. The tar employed may be that produced by another coke-oven battery or it may be tar derived from other sources such as gas retorts, vertical retorts and water-gas sets. Such tar can be mingled with the tar from the battery or it may be introduced independently and circulated through the main for the purpose of producing pitches having different characteristics from those derived wholly from coke-oven tar. If tars from different sources are mingled and introduced to the collector main for distillation in the manner described, composite pitch products will be produced and in this manner products having a greater or smaller proportion of free carbon than normal coke-oven pitches can be prepared.

The hot gases flowing through the collector main will be cooled partially by transfer of heat to the tar or pitch undergoing distillation, and the gases will be enriched in oil constituents derived from the distillation of the tar or pitch. The gases may, however, leave the collector main still at relatively high temperature and will carry in addition to the oil vapors a certain proportion of tar in the form of "tar fog", together with solid particles of coal, coke, etc., which are carried over from the coke ovens. The dew points of the gases for the several constituents carried therein as vapor are lower than the normal boiling points of these constituents, and by suitable regulation of the temperature of the gases during the distillation and especially during the subsequent treatment of the gases, it is possible to retain the desired oils therein. Thus, by sufficiently cooling the gases in the collector main all or substantially all of the tarry constituents can be converted into the liquid phase while the desired oils remain as vapors therein. The temperature of the gases should, to accomplish the intended purpose, be maintained above the dew point of the gases for the most readily condensable oil constituent desired so that substantially all of the oils will be retained and will be carried over with the gases from the collector main.

Substantially all of the undesirable constituents, including "tar fog" and solid particles, can be separated from the gases by passing them through an electrical precipitator such, for example, as the well known Cottrell precipitator, the precipitator being operated at substantially the temperature required to maintain the desired oil constituents in the vapor phase, that is to say, at a temperature higher than the dew point of the gases for the most readily condensable oil constituent therein.

The gases which are thus cleaned will upon

total condensation yield a mixture of oil constituents. The dew points of the gases for these several constituents are different and it is possible by fractional condensation to recover oils having varying characteristics. These oils, being substantially free from tarry constituents and other impurities, are in condition for immediate utilization for the purposes to which oils commonly obtained by distillation of tar are adapted.

The invention can be applied to ordinary coke-oven plants such as Semet-Solvay, Koppers plants, etc., without substantial modification thereof, the usual standard equipment being employed with the addition of the electrical precipitator and of means for circulating tar and pitch through the collector main. Thus, the ovens of the battery may be connected in the usual manner by uptake pipes and goose-necks to the collector main and the gases may be conveyed therefrom through the electrical precipitator and thence through a cross-over main to the ordinary condensing system. A suitable storage tank for the tar or pitch to be circulated through the collector main and a pump or pumps with connections to the collector main will be provided to permit the maintenance of the required volume of tar or pitch in the main during the operation thereof.

The gases escaping from the collector main at the temperature requisite for recovery of desired oil constituents following cleaning of the gases are delivered at such temperature to the electrical precipitator which is disposed preferably closely adjacent to the collector main in order that the temperature of the gases may not be reduced unduly before precipitation of the tarry constituents is accomplished. The electrical precipitator should be constructed so as to be capable of withstanding the temperatures to which it is subjected in the operation as described. It may be insulated advantageously to prevent loss of heat from the gases.

The operation of an electrical precipitator as commonly used consists of passing the gas to be treated between electrodes whose difference in electrical potential is very great. Experience has shown that it is best to use a rectified alternating current. The alternating current (the primary) is sent through a step-up transformer to produce a high potential current (the secondary) which is then rectified to an intermittent uni-directional current, for example, by means of a rotary converter. The rectified current is delivered from the converter to the electrical precipitator at practically the potential at which it leaves the transformer.

The electrical precipitator consists commonly of a group of vertical pipes with a wire or rod in the centre of each, the pipes being connected to proper headers for the introduction and discharge of the gases. The pipes generally constitute the positive electrodes and the wires or rods the negative electrodes. The size of the pipes may vary, but in general pipes of less than six inches in diameter are not used. Electrical precipitators with pipes six inches in diameter using secondary voltages from 35,000 to 50,000 volts are satisfactory for the purposes of this invention. It is generally best to operate with maximum potential difference (secondary current) between electrodes, this maximum being just below the break-down voltage at which arcing occurs.

The efficiency of the cleaning is dependent upon several variables. Satisfactory cleaning of the gas may be accomplished if the time of treatment is of the order of one second although this time may be varied widely depending upon the character of the oil to be recovered from the gases passing through the precipitator. In working with tubes nine feet long and six inches in diameter, for example, an oil substantially free from all tarry constituents has been obtained with a time of treatment of from one and one-half to two seconds, that is to say, with a gas velocity of from 6 to 4.5 feet per second. This has given an efficiency of cleaning of approximately ninety-nine per cent. If it is desired to collect oils containing small amounts of tar shorter treatment may be used, for example, a treatment of from 0.5 to one second, that is to say, a velocity of from 18 feet to 9 feet per second.

From the electrical precipitator the gases carrying condensable vapors can be conducted through suitable condensing apparatus including coolers, scrubbers, fractional condensers, fractionating columns, etc., designed to reduce the temperature of the gases and to cause the separation of oils therefrom. One total oil fraction may be collected if the gases are cooled in one step to the lowest desired temperature. Several oil fractions can be recovered if the gases are cooled by stages in coolers designed for separate collection of oils corresponding to the individual cooling steps.

Electrical precipitators of the type employed for the initial cleaning of the gases may be used to separate the condensed vapors. By passing the gases through such precipitators at predetermined temperatures the condensed constituents can be separated effectively and sharp cuts of oil can be obtained. The gases, after the successive condensations accompanied by separation of the oil constituents, can be conducted through the usual equipment provided for the purpose of cooling the gases and recovering other valuable constituents such as ammonia and light oil therefrom.

The present invention is of more or less general application to the distillation of tar and oils and the recovery of clean oils there-

from, and it will be further illustrated by the following more detailed description taken with the accompanying drawings which shows apparatus embodying the invention and adapted for the practice thereof. It is intended and will be understood that the invention is not limited to the specific details of the apparatus as illustrated in the drawings, in which

Fig. 1 is a plan view of a portion of a coke-oven plant illustrating the application of the invention thereto;

Fig. 2 is an enlarged sectional view illustrating the collector main and the electrical precipitator connected therewith; and

Fig. 3 is a cross section of the precipitator.

Referring to the drawings, 5 indicates a battery of coke ovens in which the individual ovens are connected by uptake pipes and goose-necks 6 to a collector main 7 having a centre-box 8. Tar or pitch to be distilled is supplied from a storage tank 9 by means of a pump 10 and a pipe or pipes 11 which may deliver the tar or pitch to the opposite ends of the collector main, for example, in sufficient volume to maintain a large supply of tar or pitch in the collector main during the operation thereof. The tar or pitch flows through the collector main to the centre-box 8 and may be withdrawn therefrom through a pipe 12 and delivered to the tank 9 from which it may be recirculated for further distillation. A portion of the tar or pitch may be withdrawn from the tank 9 through a pipe 13 and additional quantities of tar for distillation can be introduced to the tank through a pipe 14.

The hot gases flowing through the collector main, after contact with the tar and pitch therein and after enrichment as the result of distillation with oil constituents, are withdrawn from the centre-box 8 and are delivered through a pipe 15 to the electrical precipitator which may be of any suitable form or construction. Preferably it consists of a shell 16 enclosing a number of tubes 17 supported in heads 18 and 19 within the shell. An inlet 20 near the bottom of the shell communicates with a chamber 21 which is partially separated from the tube section by a baffle 22. An outlet 23 permits the escape of gases from the separator after the latter have passed through the tubes.

A plurality of electrodes 24, preferably in the form of metal rods, extend through the tubes and are supported on a bus-bar 25 near the upper end of the tubes. The bus-bar 25 extends at both ends into casings 26 which enclose insulators 27 upon which the bus-bar is supported. The high tension current line extends into one of the casings and is connected with the bus-bar 25, thus supplying the necessary current from any suitable source of uni-directional current under high tension. The casing of the separator is grounded or otherwise connected to the source of current to complete the circuit. The casing and tubes form the positive electrode, the electrodes connected to the bus-bar being negative. The form and arrangement of conductors in the circuit can be varied. The arrangement should be such as to supply high tension uni-directional current to the electrodes, thereby permitting a continuous silent discharge betweeen the electrodes and the tubes through which the gases pass.

The gases carrying tar in the form of globules or "tar fog", together with solid materials such as carbon, etc., in finely divided form, enter the electrical percipitator from the collector main and pass through the tubes in the precipitator, being subjected therein to the electrical discharge which through ionization of the solid and liquid particles causes them to separate from the gases and condensable vapors. The separated liquids, together with solid particles, run down the inner walls of the tube into the chamber at the bottom of the precipitator and can be withdrawn through a pipe 28. The gases carrying condensable vapors escape from the outlet at the top of the precipitator and are delivered through a pipe 29 to a condenser 30. This condenser may be of any suitable form and in the present instance I have shown a condenser of the ordinary or wet type which is employed frequently in by-product recovery systems. The gases and vapors are cooled therein by contact with grid surfaces wet by sprays of ammonia liquor, for example, and the resulting condensate is withdrawn through pipes 30′ to a decanter 31 wherein the oil is separated from the ammonia liquor. The gases leave the condenser through a pipe 32 and are delivered to an exhauster 33 which maintains the pressure balance in the system. The gases may be conducted thence through the usual equipment (not shown) for the recovery of ammonia, light oils, etc.

The apparatus as described permits the distillation of tar and the utilization for this purpose of a portion of the heat in the coke-oven gases, and in particular it permits the operation of the collector main in a satisfactory manner, avoiding accumulation of hard pitch therein and the separation of the condensable oils which, in accordance with the present invention, are retained in the gases which, after cleaning, yield clean oil condensates. Thus, the invention permits the recovery of clean oils and of pitches having desired characteristics directly from the coke-oven gases with resulting saving in the cost of handling and of redistilling the ordinary products of a by-product recovery system.

It will be understood that the foregoing description is illustrative of the invention and that various changes may be made in the form and arrangement of the apparatus and

in the operation thereof without departing from the invention or sacrificing any of the advantages thereof.

I claim:—

1. The method of operating the by-product recovery system of a coke oven battery to recover clean tar oils, which comprises collecting hot fresh coke oven gases from the ovens of the battery in a substantially horizontal gas collector main and conducting them through the gas collector main over a body of tar continuously moving over the bottom thereof, thereby causing distillation of the tar from the surface thereof, withdrawing the gases and vapors resulting from the distillation without substantial condensation of desired tar oils, cleaning them in an electrical precipitator at a temperature above the dew point of the desired tar oils by subjecting them to an electrical discharge therein to separate entrained solid and liquid particles therefrom, and cooling the resulting clean gases and vapors to produce clean tar oils comprising constituents originally present in the gases and constituents distilled from the tar in the collector main.

2. The method of operating the by-product recovery system of a coke oven battery to recover clean tar oils, which comprises collecting hot fresh coke oven gases from the ovens of the battery in a substantially horizontal gas collector main, conducting them through the gas collector main over a body of tar continuously moving over the bottom thereof, thereby causing distillation of the tar from the surface thereof, bringing the hot gases used for the distillation in direct contact with a regulated amount of water to thereby regulate the temperature of the gases and the distillation of the tar, withdrawing the gases and vapors resulting from the distillation without substantial condensation of desired tar oils, cleaning them in an electrical precipitator at a temperature above the dew point of the desired tar oils to separate entrained solid and liquid particles therefrom, and cooling the resulting clean gases and vapors to produce clean tar oils.

In testimony whereof, I affix my signature.

STUART PARMELEE MILLER.