

Nov. 10, 1936.

K. SCHOENEMANN

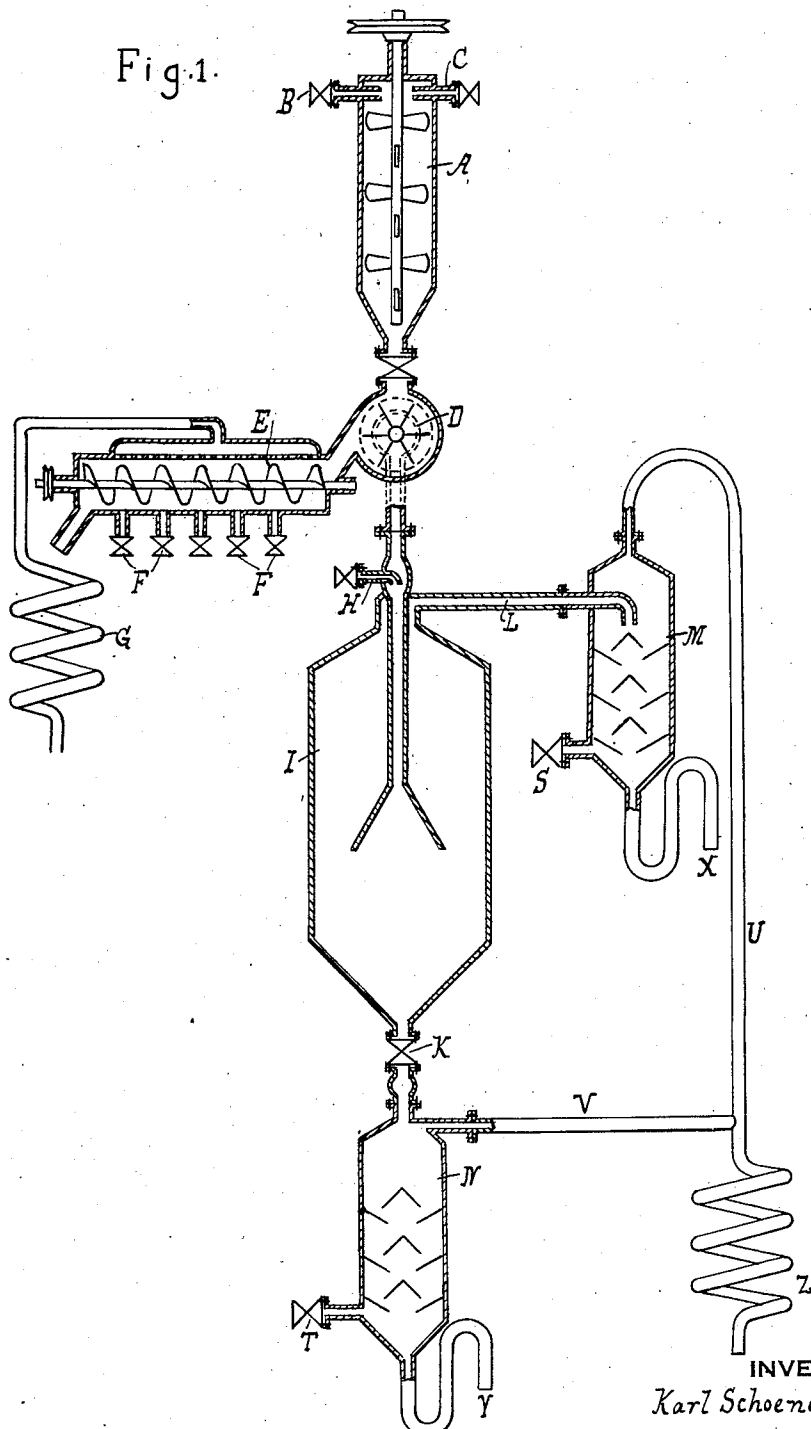
2,060,447

WORKING-UP OF CARBONACEOUS MATERIALS

Filed May 25, 1931

2 Sheets-Sheet 1

Fig. 1.



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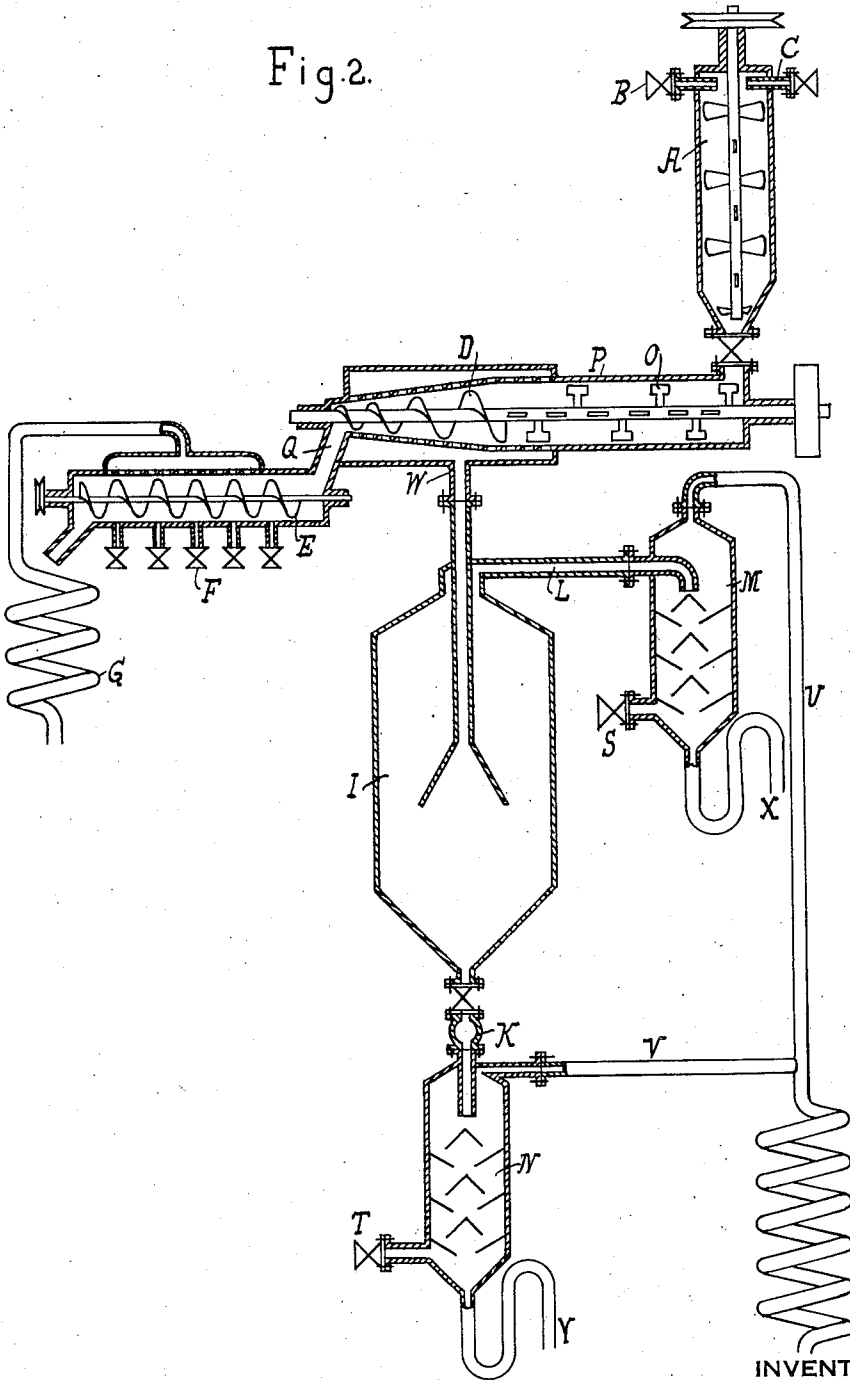
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Filed May 25, 1931

2 Sheets-Sheet 2

Fig. 2.



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2,060,447

WORKING-UP OF CARBONACEOUS
MATERIALS

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Application May 25, 1931, Serial No. 539,765
In Germany May 26, 1930

6 Claims. (Cl. 196—13)

The present invention relates to improvements in and apparatus for the working up of substances containing asphalt, such as tars, petroleums, destructive hydrogenation products of coals and the like.

In the working up of liquid or fusible carbonaceous substances containing asphalt, such as tars, petroleums, cracking products of these, destructive hydrogenation products of coals, tars, petroleums and the like, the asphalt contained therein may be precipitated together with the insoluble constituents, such as solid particles of coal, if desired continuously, by the addition of small amounts of liquid aliphatic hydrocarbons as for example distillation products of mineral oils such as benzines, petroleum, gas oil or products obtained therefrom by hydrogenating or cracking processes or mixtures containing substantial amounts thereof.

I have now found that the said methods of working up carbonaceous substances which are liquid at a temperature between 0° and 250° C. and which contain asphalt may be carried out in an advantageous manner by continuously precipitating the asphalt from the said substances in more than one stage. In this manner it is attained that large amounts of the asphalt contained in the initial materials are obtained free from dust, since the dust is practically completely carried down by the asphalt precipitated in the first stage. The least valuable, most thermally sensitive hard asphalt having the highest melting point is first precipitated together with the dust and the remaining asphalt precipitated in the subsequent stage or stages is a very valuable soft asphalt free from dust.

The materials freed from the readily decomposable asphalts may then be distilled without danger and are suitable for example for the preparation of lubricating oils. The precipitants may be recovered by distillation. In cases when it is desired not to recover the precipitant but to make use of it together with the material freed from asphalt, hydrocarbons of very high boiling point, as for example molten paraffin wax, may also be employed for the precipitation.

The single stages of precipitation are carried out in separate containers. The materials treated in each container are freed from the precipitated substances, for example by passing them through a drum filter. As a rule the precipitation of the asphalt according to the present invention is carried out in two stages and a valuable asphalt free from dust is obtained in this way.

If, however, it is desired to obtain asphalts of different kind, each kind having specific properties, the process is carried out in more than two stages.

I have further found that it is of great advantage to effect the single stages under gradually stronger precipitating conditions. These conditions may be made stronger for example by prolonging the duration of precipitation or by lowering the temperatures or by supplying further amounts of precipitants to the treated materials or by a combination of these methods.

The requisite amount of the precipitant depends upon the kind of the initial material and may be readily determined by a preliminary test. As a rule the requisite amount of precipitant increases with the following order of sequence of initial materials: Producer tar, products from the destructive hydrogenation of mineral coal, residues from asphalt base mineral oils, products from destructive hydrogenation of brown coal, residues from low temperature carbonization tar from brown coal.

According to my invention the materials are subjected to the precipitating treatment during their passage through the several containers. The process may be carried out for example as follows:

The initial material, as for example a topped product of the destructive hydrogenation of coal, is led in the molten state at 200° C. through several containers. The time occupied by its passage through the first chamber which serves for mixing it with the precipitant is regulated by the choice of the dimensions of the chamber so that it is just sufficient to precipitate a certain part of the asphalt together with the dust. By selecting a long cylindrical shape for this vessel the mixing of the contents of the vessel throughout its length is obviated. The asphalt remaining in the material freed from dust is then precipitated in a second container which is so large that the time occupied by the passage of the material there-through is sufficient for the precipitation of the asphalt. This may be effected in different ways, as for example by considerably increasing the time of precipitation by an appropriate increase in the volume of the second precipitating vessel, or by lowering the temperature, or by adding further amounts of precipitants, or by a combination of these methods. The precipitation of the asphalt in stages is of particular advantage if it is intended to use it as hard pitch. The withdrawal of the asphalt containing dust is effected in different ways depending on its consistency. Solid

granular precipitates which are infusible at the temperature of precipitation, such as are obtained when employing small amounts of precipitants or a short time of precipitation or high temperatures, may be continuously separated by filtration, as for example by means of a drum filter, sieve centrifuge or the like. Pasty asphalt capable of being kneaded is obtained when a sufficient amount of asphalt which is liquid at the precipitating temperature is precipitated with the granular hard asphalt. It is also removed continuously by kneading or pressing out, as for example by means of spiral presses. Fused asphalt may be separated for example by sedimentation and withdrawn in a liquid state.

If desired the precipitant may be distilled off and recovered in suitable apparatus, as for example distilling columns.

Considerable amounts of liquid products can be obtained from the hard asphalt containing ashes, and which otherwise has scarcely any industrial value, by low temperature carbonization.

The asphalts may be continuously precipitated in the same manner from asphaltic substances other than products of destructive hydrogenation, as for example from petroleum, tars, pitches, cracking products of the same and the like. The aliphatic hydrocarbons may also be wholly or partly replaced by other asphalt precipitants, as for example alcohols, ketones, acids, or esters.

The advantages of this method of working in stages consists mainly in the fact that there is an improvement in the precipitation and separation of the asphalt. Since the operation is carried out continuously the process may be very readily combined with a subsequent or previous process (as for example a destructive hydrogenation process) and greater throughputs and a better utilization of heat are made possible. Furthermore the continuous operation has the advantage that the conditions in the single stages, such as temperature, duration of treatment, amount of precipitant, may be maintained and controlled much more easily than when working discontinuously. Thus in the latter case the duration of precipitating the asphalts in a stage varies by the time required for separating the precipitate from the liquid.

Also the precipitation and the separation themselves take place in a better way by the fact that the asphalt first precipitated in the form of spongy flocks is much more readily conglomerated by stirring and kneading and is much more rapidly separated in a state in which it is much poorer in oil by filtration or pressing than by a discontinuous sedimentation and decantation.

The nature of the invention will be further described with reference to the accompanying drawings which illustrate arrangements of apparatus suitable for carrying out the process according to the present invention but the invention is not restricted to these arrangements.

Referring to Figure 1, 1000 kilograms per hour of a mineral coal destructive hydrogenation product freed from constituents boiling below 300° C. and containing 25 per cent of dust enter a stirring tower A at B at a temperature of about 200° C. and at the same time 150 kilograms of kerosene (boiling point from 200° to 250° C.) the tower A at C. The temperature in the stirring tower A is maintained at about 180° C. Each part of the product remains about 10 minutes in the said tower. The two products are thus intimately mixed. The hard asphalt containing dust precipitated during the passage of the mixture through the stirring tower A is separated by a

drum filter D and is treated in the spiral E with superheated steam entering through inlets F. The distillate is condensed in G. The filtrate is then mixed with a further 80 kilograms of kerosene which flows in through a nozzle H. The precipitation and sedimentation of the remaining asphalt take place in the vessel I which is maintained at a temperature of about 160° C. and through which the materials pass during the course of about 50 minutes and the asphalt is allowed to flow out at K in the liquid state. The kerosene is expelled from the oil free from asphalt flowing through the pipe L in the distilling column M, and from the soft asphalt in the column N by means of steam introduced at S and T respectively, and flows by way of pipes U and V respectively into the condenser Z. The asphalt is withdrawn at Y and the oil free from asphalt at X. The initial product is thus split up into 35 per cent of hard asphalt containing dust, 45 per cent of soft asphalt free from dust and 20 per cent of oil free from asphalt.

Referring to Figure 2, 200 kilograms of kerosene are added to 1000 kilograms of a hydrogenation product in the stirrer A in the same manner as described with reference to Figure 1. The materials remain in this mixer, which is maintained at a temperature of about 180° C., for about 30 minutes. The asphalt which separates in the form of spongy soft flocks is kneaded together into a coherent mass in the vessel P by a slow stirrer O, pressed by means of the spiral press R, forced out through the pipe Q and treated in the spiral E with steam entering through the inlet F. The distillate is condensed in G. The oil flowing through W is allowed to remain in the sedimentation vessel J which is kept at a temperature of from 120° to 150° C., about four times as long as in the mixing and stirring vessels. During this time the remainder of the asphalt is precipitated. The asphalt and oil are withdrawn and the precipitant distilled off in the manner described with reference to Figure 1. The yields amount to 40 per cent of asphalt containing dust, 30 per cent of asphalt free from dust and 30 per cent of oil.

What I claim is:—

1. A process for working up a carbonaceous material which is liquid at a temperature between 0° and 250° C. and which contains dissolved asphalt, which comprises precipitating said asphalt in more than one stage by the addition to said material of a liquid aliphatic hydrocarbon as precipitant while continuously passing through each stage the mixture of said material and said hydrocarbon, and separating the asphalt precipitated therefrom in each stage.

2. A process for working up a topped product of the destructive hydrogenation of coal which contains dissolved asphalt, which comprises precipitating said asphalt in more than one stage by the addition to said product of a liquid aliphatic hydrocarbon as precipitant while continuously passing through each stage the mixture of said product and said hydrocarbon, and separating the asphalt precipitated therefrom in each stage.

3. A process for working up a carbonaceous material which is liquid at a temperature between 0° and 250° C. and which contains dissolved asphalt, which comprises precipitating said asphalt in more than one stage by the addition to said material of a liquid aliphatic hydrocarbon as precipitant under successively stronger precipitating conditions comprising duration of exposure to the precipitant, amount of the precipitant and

temperature, while continuously passing through each stage the mixture of said material and said hydrocarbon, and separating the asphalt precipitated therefrom in each stage.

5 4. A process for working up a carbonaceous material which is liquid at a temperature between 0° and 250° C. and which contains dissolved asphalt and solid constituents, which comprises precipitating the dust and part of the asphalt by
10 the addition to said material of a liquid aliphatic hydrocarbon as precipitant, while continuously removing the mixture of liquid and precipitate and separating the latter from the liquid, and precipitating in a second stage the remainder of
15 asphalt contained in said liquid under stronger precipitating conditions comprising duration of exposure to the precipitant, amount of the precipitant and temperature, while continuously passing the said liquid through this stage and
20 separating from the liquid the asphalt precipitated in this stage.

5 5. A process for working up carbonaceous material, which is liquid at a temperature between 0° and 250° C. and which contains dissolved asphalt, which comprises passing said material in
25 a continuous stream through a series of chambers,

adding a precipitant for asphalt to said material in the first chamber, removing the precipitated asphalt from the carbonaceous material in the course of its passage from each chamber to the next and adding further quantities of a precipitant for asphalt in each succeeding chamber. 5

6. A process for working up carbonaceous material, which is liquid at a temperature between 0° and 250° C. and contains dissolved asphalt and suspended solid impurities, which comprises
10 passing the said material in a continuous stream through a series of chambers, adding to the material in the first chamber a precipitant for asphalt to precipitate sufficient asphalt to carry
15 down the solid impurities, removing the precipitate of asphalt and solid impurities from the carbonaceous material in the course of its passage from the first chamber to the next, precipitating pure asphalt in each of the remaining
20 chambers by adding to the material in each of said chambers further quantities of a precipitant for asphalt and recovering pure asphalt from the carbonaceous material in the course of its passage from each chamber to the next. 25

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