FORM 2

THE PATENT ACT, 1970

(39 of 1970)

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The Patent Rules, 2003

COMPLETE SPECIFICATION

(See section 10 and rule 13)

1. TITLE OF THE INVENTION:

A technology for production of mesophase pitch from coal tar

2. APPLICANT:

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The following specification particularly describes the invention and the manner in which it is to be performed:

4. Description:

Field of Invention:

[0001] The present invention relates to a process for producing mesopahse pitch from coal tar useful as a raw material for producing carbon fibers with high modules and other carbon materials. More particularly the invention is concerned with a process for separating insolubles components from coal tar prior to formation of coal tar pitch.

Background of the invention:

[0002] In general, carbon fibers are industrially produced by using rayon, PAN (polyacrylonitrile) or a pitch as a raw material. PAN is disadvantageous as it is expensive and the carbonization yield is low. In this respect, pitches are inexpensive and thus economically attractive. Extensive studies have been made to produce mesophase pitches as raw materials for high performance carbon filbers from heavy oils such as petroleum catalytic cracking residual oil, naphtha tar pitch, or coal tar pitch.

[0003] Coal tar pitch utilized as a raw material for carbon fibers requires removal of quinoline insolubles. Mesophase pitch is a highly structured pitch which is used in applications where strength or the ability to conduct heat or electricity is important. Significant work has been performed to produce mesophase pitch from coal tar pitch with limited success because of the quinoline insolubles content of the pitch.

[0004] At present, coal tar to remove quinoline insolubles purification methods include: hot filtration, centrifugal separation, electrostatic separation method. Hot filtration separation is good to meet industrial requirements, but more stringent requirements for the filter, large industrial investment costs. High separation efficiency centrifugal separation, but a small amount of processing on the devices requires high energy consumption. High separation efficiency electrostatic separation, electrostatic separation voltage but low-impact separation efficiency, increased multi-stage electrostatic separation costs.

[0005] It has been shown that the quinoline insolubles particles in coal tar pitch hinder coalescence of the mesophase spheres causing a poor quality mesophase to be formed. Known methods of producing mesophase from coal tar pitch involve a filtration or

centrifugation step for removing the quinoline insolubles. While these processes work quite well and allow for production of a high quality mesophase, they result in a very high cost of the mesophase product.

[0006] Coal tar is derived from coal. It is a byproduct of the production of coke, a solid fuel that contains mostly carbon, and coal gas. While coal-tar pitch is a thick black liquid that remains after the distillation of coal tar. Prior methods of production of mesophase pitch include duistillation of coal tar to obtain coal tar pitch. Treatment of pitch with suitable solvent to remove insolubles and further distillation of the obtained insolubles free pitch. This method involves more energy input first for distillation of coal tar to obtain pitch and subsequent stages of distillation to obtain mesophase.

[0007] Further, these known processes of removing quinoline insolubles are extremely expensive, subject to breakdowns and connected with very great losses. Also these are complex operation with equipment requirements. Also known process for removal of insolubles components are performed after formation of coal pitch with insolubles and then subsequent removal.

[0008] Prior methods of removal of insolubles from coal tar include subjecting coal tar with solvent mixture to centrifugal separation which is costly. The ease to maintain temperature in centrifugal separation for aiding separation of insolubles from coal tar is very complicated to maintain given the losses. Further separation of insolubles through gravity settling although is considered but consumes a lot of time. Also the solvents used for separation of insolubles cannot be recovered completely. Low boiling point solvents often escape from the vents and as a result recovery is affected. The solvent need to be added freshly for every batch of mesophase pitch formation which is not economical.

[0009] Therefore, there is a need for an economical process for removal of insolubles from the coal tar prior to the formation the coal tar pitch. There is a need for suitable solvents that can be completely recovered without losses. There is also a need for a means to optimize settling process to separate insolubles in shorter time.

Objectives of the invention:

[0010] The objective of the invention is to produce mesophase pitch from coal tar with high modules that can be used as a precursor for carbon fibres and other carbon material.

[0011] Another objective of the invention is to produce mesophase pitch with a mesophase content of atleast 65%.

[0012] Another objective of the invention is to produce mesophase pitch with a softening point of 250˚C- 300˚C.

[0013] Another objective of the invention is to yield zero quinoline insoluble coal tar pitch by treating coal tar.

[0014] Another objective of the invention is to remove insolubles, in particular quinoline insolubles prior from coal tar prior to formation of coal tar pitch.

[0015] Another objective of the invention is to treat coal tar to remove insolubles, in particular quinoline insolubles economically.

[0016] Another objective of the invention is to separate insolubles from coal tar using gravity settling in a shorter time period.

[0017] Further objective of the invention is to decrease the time required for separation of insolubles from coal tar compared to the separation process preformed at the room temperature.

[0018] Further objective of the invention is to use suitable solvents that can be efficiently recovered after the separation of insolubles form coal tar.

Summary of the Invention:

[0019] The invention proposes a technology for production of mesophase pitch from coal tar by dehydrating coal tar at a temperature of 100°C C to 120°C to remove moisture from the coal tar pitch and treating the obtained dehydrated coal tar with coal derived solvent.

The coal derived solvent is selected from the group of high boiling point aromatics such as residual oils from cracking reactions of petroleum fractions rich in aromatics or especially from coal tar distillate fractions, such as wash oil, naphthalene oil, heavy oil, anthracene oil, debased and dephenolized light oil and the like.

[0020] The ratio of coal tar to solvent may be in a range from 0.2: 1 to about 3: 2 to separate insoluble components from the coal tar solvent mixture in a separation vessel maintained at temperatures in the range of 90˚C to 110˚C for a predetermined time. The insoluble components are separated from coal tar and are settled at the bottom of the vessel with the supernatant comprising of coal tar with zero insoluble components. Further distilling the coal tar with zero insoluble components is performed at a temperature range of 340°C to 380˚C for 5 to 10 hrs under inert atmosphere by means of inert gas purging to yield coal tar pitch with zero insoluble components having a softening point of about 60°C -85°C.

[0021] The solvent used in the process is recovered after first stage distillation. The solvent is continuously replenished after the first stage distillation of coal tar, as a result of fraction left after first stage distillation and reused for another cycles. The purified pitch for further distillation at a temperature of 430°C to 450°C at 1.1 to 1.5 bar pressure along with inert purging for 6 to 10 hrs to obtain mespohase pitch having 63% to 68% mesophase content with a softening point of 250°C - 300°C.

Description of Drawings:

[0022] The accompanying drawings, which are incorporated in and constitute a part of the specification, illustrate an embodiment of the invention, and, together with the description, serve to explain the principles of the invention.

[0023] FIG. 1 schematic flow diagram which illustrates a technology for production of mesophase pitch from coal tar according to an embodiment of the invention.

Detailed description of Drawings:

[0024] An exemplary embodiment of the present invention will be described in reference to the accompanying drawings. Wherever possible, same or similar reference numerals are used in the drawings and the description to refer to the same or like parts or steps.

[0025] Accordingly, the present invention relates to a technology for production of mesophase pitch from coal tar with a mesophase content of atleast 65% and a softening point of 250˚C- 300˚C. According to the present invention, the coal tar feed is heated to a temperature sufficient to remove substantially all water from coal tar to produce a heated dehydrated tar. In accordance to an aspect of the invention, the coal tar may be heated to the temperature in the range of 100°C to 160°C for a predetermined time to remove substantially all water.

[0026] It is a feature according to an aspect of the present invention to provide a process for the separation of components insoluble in quinoline by treating the coal tar with a suitable coal derived solvent and separating the insoluble components.

[0027] A coal derived solvent as used in the present invention is intended to mean a solvent with aromatic characteristics of which are paramount. Therefore, preferably high boiling point aromatic solvents are used, particularly those obtainable from residual oils from cracking reactions of petroleum fractions rich in aromatics or especially from coal tar distillate fractions, such as wash oil, naphthalene oil, anthracene oil, debased and dephenolized light oil or cuts from it.

[0028] As illustrated in Fig. 1, coal tar feed is first heated to a temperature sufficient to remove therefrom substantially all water to produce a heated dehydrated tar at step 101. The coal tar is heated in a temperature range of at a temperature 100°C to 120°C. The coal tar after dehydration remains hot having a temperature in between 100°C and 120°C. This temperature of the coal tar is taken advantage of in further process treatment of the coal tar.

[0029] According to an aspect of the invention, at step 102, the heated dehydrated coal tar is treated with a coal derived high boiling point aromatic solvent. The heated dehydrated coal tar may be mixed in a solvent in a ratio of coal tar to solvent in the range of 0.2: 1 to

about 3:2. The solvent is selected to have to have high boiling point aromatics in order to facilitate easy recovery.

[0030] The ratio of coal tar used to the solvent ranges generally from 1:5 to 3:1, preferably from 1:2 to 2:1. Effectively the ratio is selected at the same time such that for achieving good results, as little solvent as possible is required.

[0031] According to an aspect of the invention, the solvent coal tar blend is fed into a separation vessel at step 103. According to an aspect of the invention, the apparatus may be a decanter. The mixture is allowed to stand in the vessel for a predetermined time to allow settling of insoluble components from the coal tar. According to an aspect of the invention, the predetermined time is chosen to facilitate completer separation of insoluble components, wherein the predetermined time may be 70-72hrs.

[0032] According to an aspect of the invention, the separation vessel is insulated to prevent temperature losses. The coal tar solvent mixture is maintained within the vessel at a temperature of 90°C to 110°C to enhance the settling of insoluble components. It is to be understood the dehydrated coal is at a temperature in between 100°C and 120°C, as a result additional temperature input is not required in the separation vessel. The temperature of the dehydrated coal can be maintained in the vessel through proper insulation.

[0033] The insoluble components, especially the quinoline insolubles are settled at the bottom the decanter. The layer above the insolubles comprises of zero quinoline coal tar and solvent mixture. The top layer according to an aspect of the invention is removed after draining the quinoline insolubles from the bottom of the vessel.

[0034] According to an aspect of the invention, at step 104, the obtained supernatant which is zero QI (quinoline insolubles) coal tar is separated and fed into a high temperature reactor. The high temperature reactor facilitates distillation of the supernatant. The high temperature reactor may be an apparatus capable of operating at temperatures of 200° C. to 500° C. At step 104, first stage distillation of zero QI (quinoline insolubles) coal tar is carried out in the high temperature reactor at 355˚C to 365˚C for 5 to 10 hrs under inert atmosphere by inert gas purging. The distillation process yield coal pitch free from insoluble

components. According to an aspect of the invention, at step 104, the first stage distillation of coal tar yields coal tar pitch having a softening point of about 60˚C-85˚C.

[0035] According to an aspect of the invention, at step 105, the obtained coal pitch free from insoluble components is subjected to second stage distillation, in a temperature range of 430˚C-450˚C at 1.1 to 1.5 bar pressure, along with nitrogen purging at a flow rate of 1.5 to 2.0 litres per minute, for 6 to 10 hrs. The second stage distillation according to an aspect of the invention yields mesophase content with a softening point of 250˚C- 300˚C.

[0036] According to an aspect of the invention, after the first stage distillation, the solvent used in the recovered. Also, it is to be understood, the distillation of coal tar also obtains residual oils that can be used as the solvent for separation of quinoline, the solvent is fed into the separation vessel. The solvent is replenished at every cycle of the process.

[0037] As the subsequent example given by way of comparison shows, these results which may be achieved with the process according to the invention. If not stated otherwise, preceding and succeeding ratio and percentage data relate to parts by weight or percent by weight, temperatures relate to degrees centigrade and data for the boiling point relate to 1 bar pressure. QI designates quinoline insolubles.

[0038] EXAMPLE 1

[0039] A coal tar feed is preheated to a temperature of 110˚C to remove water content from the coal tar. The coal tar is then mixed with heavy oil in 1:1 ratio and allowed to stand in a decanter. The decanter is insulated to maintain at a temperature of 100˚C for 72 hrs. The insoluble components are tapped from the vessel, while the supernatant is separated and fed into a high temperature reactor. The supernatant is distilled at a temperature of 360˚C for 7 hrs yield zero Q.I pitch. Further distillation of the zero Q.I pitch is carried out at 430˚C at 1.1 bar pressure, along with nitrogen purging at a flow rate of 1.5 litres per minute, for 7 hrs.

[0040] The obtained zero Q.I pitch after first stage distillation is found to be having a softening point of about 60˚C. The zero Q.I pitch after second stage distillation is found to have softening point of 250˚C with mesophase content of 65. 70%.

[0041] The invention is advantageous in eliminating additional energy input by separation of QI insolubles from coal tar prior to formation of coal tar pitch by maintaining the dehydrated coal temperature. The dehydrated heated coal tar temperature can be maintained in the insulation vessel, during the separation process with solvent blend, which is useful in eliminating additional heat input for separation. According to the invention, the insolubles can be separated using gravity settling in a shorter time period with the aid of temperature conditions maintained within the separation vessel. Also the separation is faster compared to the separation process that is performed at room temperature. The invention eliminates the need of centrifugal separation which is costly and also cumbersome to maintain temperature that aid in the separation of QI insolubles.

[0042] It will readily be apparent that numerous modifications and alterations can be made to the processes described in the foregoing examples without departing from the principles underlying the invention, and all such modifications and alterations are intended to be embraced by this application.

5. Claims

We Claim

1. A technology for production of mesophase pitch from coal tar, comprising the steps of:

   Dehydrating coal tar by heating to a temperature of 100°C to 120°C to remove substantially all water from coal tar;

   Treating the obtained dehydrated coal tar with coal derived solvent, wherein the ratio of coal tar to coal derived solvent may be in a range of 0.2: 1 to about 3: 2 to separate insoluble components from the coal tar;

   Separating insoluble components from coal tar solvent mixture in a separation vessel maintained at temperature in a range of 90˚C to 110˚C for a predetermined time, wherein the insoluble components are settled at the bottom of the said vessel, with the supernatant comprising of coal tar with zero insoluble components;

   Subjecting the coal tar with zero insoluble components for first stage distillation in a temperature range of 340°C to 380˚C for 5 to 10 hrs under inert atmosphere by means of inert gas purging to yield coal tar pitch with zero insoluble components having a softening point of about 60°C to 85°C, wherein the solvent used is recovered and may be reused; and

   Subjecting the purified pitch distillate for second stage distillation at a temperature of 430°C to 450°C at 1.1 to 1.5 bar pressure, along with inert gas purging, for 6 to 10 hrs to obtain mespohase pitch having mesophase content greater than 60% with a softening point of 250°C - 300°C.

2. The technology for production of mesophase pitch from coal tar as claimed in claim 1, wherein the said coal tar based solvent may be high boiling point aromatic solvents, particularly those obtainable from residual oils from cracking reactions of petroleum fractions rich in aromatics or especially from coal tar distillate fractions, such as wash oil, naphthalene oil, anthracene oil and the like.

3. The technology for production of mesophase pitch from coal tar as claimed in claim 1, wherein said solvent is added to the coal tar at a ratio of coal tar to coal derived solvent in a range of 0.2: 1 to about 3: 2 to separate insoluble components from the coal tar.

4. The technology for production of mesophase pitch from coal tar as claimed in claim 1, wherein said solvent being a coal derived solvent, is continuously replenished after the first stage distillation of coal tar, as a result of fraction left after first stage distillation and reused for another cycles.

5. The technology for production of mesophase pitch from coal tar as claimed in claim 1, wherein the said dehydrated coal tar & solvent mixture is fed into a separation vessel and maintained at a temperature range of 90˚C to 110˚C for about 70 to 72 hours, wherein the insolubles separate from the coal tar and settle at the bottom of the vessel which are tapped, while the supernatant comprises of coal tar with zero insoluble components.

6. The technology for production of mesophase pitch from coal tar as claimed in claim 1, wherein said separation of insoluble components from coal tar and solvent mixture may be accomplished by simple gravity settling, wherein said gravity settling may be performed in a decanter, which is insulated to prevent heat loss from the dehydrated coal.

7. The technology for production of mesophase pitch from coal tar as claimed in claim 1, wherein the said obtained zero quinoline insoluble coal tar is subjected to distillation in a high temperature reactor at a temperature range of 340°C to 380˚C for 5 to 10 hrs under inert atmosphere by means of inert gas purging to yield coal tar pitch with zero insolubles having a softening point of about 60°C -85°C.

8. The technology for production of mesophase pitch from coal tar as claimed in claim 1, wherein said purified pitch contains greater than 60% mesophase content with a softening point of 250°C - 300°C.

9. The technology for production of mesophase pitch from coal tar as claimed in claim 1, wherein the solvent is distilled to obtain a purified pitch having a softening point of about 60°C to 85°C, wherein the distilled solvent may be recovered for re-use.

Patent Agent Signature:

Patent Agent Name: Ganapathi S Naidu Konathala

6. Abstract:

Title: <u>A technology for production of mesophase pitch from coal tar</u>

The present invention proposes a technology for production of mesophase pitch from coal tar. In specific, the mesophase pitch is produced from coal tar by separation of quinoline insolubles from coal tar prior to formation of coal tar pitch. Dehydrated coal tar is mixed with a suitable coal derived solvent in an insulated separation vessel for a predetermined time. The coal tar free from insolubles is subjected to subsequent distillation stages in a high temperature reactor for production of mesophase pitch with a mesophase content greater than 60˚C having a softening point in the range of 250˚C- 300˚C. The solvent is continuously replenished in the first stage distillation and is recovered for reuse. The dehydrated heated coal tar temperature can be maintained in the separation vessel, during the separation process with solvent blend, which is useful in eliminating additional heat input for separation. The obtained mesophase pitch may be used as the raw material for carbon fibres and other carbon materials.

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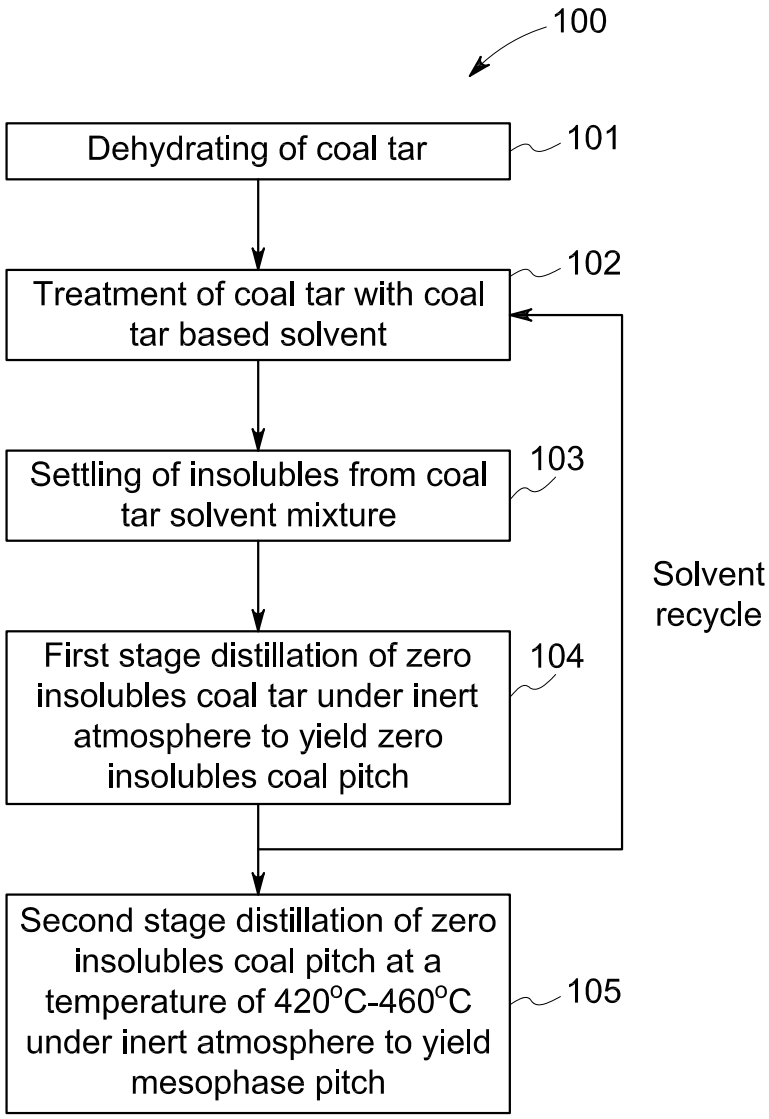

100
Dehydrating of coal tar
101
Treatment of coal tar with coal tar based solvent
102
Settling of insolubles from coal tar solvent mixture
103
First stage distillation of zero insolubles coal tar under inert atmosphere to yield zero insolubles coal pitch
104
Second stage distillation of zero insolubles coal pitch at a temperature of 420°C-460°C under inert atmosphere to yield mesophase pitch
105
Solvent recycle

FIG. 1