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PROCESS OF TREATING TAR

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This invention relates to the treatment of tar, and more particularly to the distillation of coal tars of low free carbon content to obtain pitch products of higher free carbon content and which have low boiling oils removed therefrom so that the resultant products are of improved stability. By coal tar I intend to be understood as meaning coke oven, gas retort, vertical retort or low temperature tars, or mixtures of any thereof.

10 The process of this invention is also applicable to the treatment of water gas tar or mixtures of water gas tar and coal tar, or any tarry residuals resulting from the distillation of coal, peat or shale.

15 Heretofore coal tar pitch products such as road materials having preferred melting points and free carbon contents or those frequently specified in the trade could not be obtained by simply distilling or otherwise treating coke oven tar alone, i. e. not admixed with other tars. Coke oven pitch products such as the various grades of road materials and fluxing oils, etc., have been produced by distilling coke oven tar admixed with gas works or gas house tar until the pitch product of the desired consistency was obtained. The gas works or gas house tar which has a free carbon content in the neighborhood of 18% by weight because of its high free carbon content was admixed with the coke oven tar, the free carbon content of which is approximately 7% by weight and distilled, the resultant pitch product obtained from the mixture having a correspondingly higher free carbon content than the pitch product obtained from the distillation of the coke oven tar alone. In order to produce a road or other material having a free carbon content of approximately 12% or above and a melting point of from 100 to 110° F. it has been customary to distill a mixture of coke oven tar and gas house tar until a residual pitch of the desired melting point and free carbon content is formed.

Further, the pitch products resulting from the usual distillation of coal tar were often objectionable because they were readily susceptible to temperature changes encountered in the uses for which these products are adapted, i. e., changes in atmospheric temperature from day to day and seasonal changes in temperature would deleteriously affect these products.

One object of this invention is to provide a process for treating tars of relatively low free carbon content, such as coke oven tar or vertical retort tar, alone, i. e., not admixed with other tars of relatively high free carbon content, to obtain pitch products of preferred softening or

melting point and definite relatively high free carbon content, which products are suitable for use as road materials and are not deleteriously affected by the temperature changes normally encountered in the customary use of these products. Other objects and advantages will appear from the following description.

According to this invention coke oven tar or other tar is distilled to a pitch residue and the distillation is carried to the point where decomposition and free carbon formation takes place thereby increasing the free carbon content of the pitch, the distillate oils are collected, and thereafter the distillate oils or only the heavier fractions of these oils are oils corresponding thereto are admixed with the residue in the still. During the latter part of the above described distillation, the distillate vapors may be passed to a reflux condenser and part or all of the condensate returned to the still. The resultant mixture of increased free carbon content may be and preferably is further distilled to form a pitch product of the desired softening or melting point and having the desired free carbon content. By proper control of the distillation operation and of the specific gravity of the distillate oils returned to the pitch residue the low boiling oils are eliminated from the pitch product and a product of very satisfactory stability toward temperature changes is obtained.

In carrying out the present invention a battery or series of successive stills or an individual still of any well known type adapted for the distillation of coal tar may be utilized. Preferably stills of the fractionating type are used, the coke oven or other tar being charged into the still and the various fractions of distillate oils being separately collected. The distillation operation is carried to a temperature where decomposition and carbon formation takes place in the pitch, thus building up the free carbon content of the pitch residue in the still. To this residue is then added all or any part of the distillate, depending upon the nature and characteristics of the desired product and the resultant mixture is preferably distilled to obtain the end product. If desired, additional crude or dehydrated tar may be added to the mixture of pitch and distillate oil and the resultant mixture distilled to produce the desired end product.

In accordance with one specific embodiment of this invention to produce a pitch of approximately 108° F. and not less than 104° F. softening point and a free carbon content of approximately 27% by weight, a crude coke oven tar

having a specific gravity of 1.183 at 25° C. and a free carbon content of 7.2% by weight was distilled in the usual type still until a residual pitch having a softening point of 300° F., determined by the cube-in-air method given on page 821, vol. 10, 1918 issue of the Journal of Industrial & Engineering Chemistry, was formed. This distillation operation resulted in a removal of approximately 52% by volume of distillate oil from the crude tar and in a pitch having a free carbon content of approximately 46% by weight. All the distillate oil was returned to the still producing a tar mixture having a specific gravity of 1.202 at 25° C. and a free carbon content of 23.5% by weight. The resultant mixture was distilled until a pitch of 108° F. softening point (cube-in-water method) having a free carbon content of approximately 27% by weight, was obtained. The second distillation operation resulted in a removal of 14.3% by volume of distillate oil.

While as above indicated the first distillation operation may be carried on until a pitch of 300° F. softening point is formed, this distillation operation may be conducted only until an appreciable decomposition of the pitch residue sets in, resulting in the formation of substantial amounts of free carbon in the pitch residue in the still in addition to the free carbon originally present in the tar. Appreciable decomposition of the pitch usually begins after the softening point of the pitch has reached approximately 175° F. Thereafter at any desired point depending on the softening point and free carbon content desired in the final product, distillation may be discontinued and the distillate oil or preferably only the heavy oil fraction thereof having a specific gravity of at least 1.10 at 25° C. is introduced into the still and the resultant mixture again distilled to form the desired end product. If desired additional crude or dehydrated tar may be added to the mixture of pitch and distillate oil and the resultant mixture distilled to produce the end product.

If desired, the first distillation operation may be conducted as above described and the various fractions of distillate oils separately collected. The heavier or higher boiling fraction, i. e., fractions having a specific gravity above 1.10 at 25° C., preferably above 1.13 at 25° C. and coming off from the still for the most part between 300° C. and 400° C. may be combined with some or all of the residual pitch of 300° F. softening point, preferably in the proportion by volume of 48% distillate oil to 52% pitch. A product having a softening point of 107° F. and a free carbon content of approximately 23% may thus be obtained. This pitch product is formed by mixing the pitch with the oil and suitably agitating the mixture until a substantially homogeneous pitch is obtained.

Instead of carrying on the first distillation operation until a pitch of 300° F. softening point is formed, the operation may be continued only until a pitch having a softening point of about 250° F. or a pitch having a free carbon content of 28% or higher remains in the still. This pitch residue may then be cut back with the necessary amount of heavy oil having a specific gravity of about 1.10 at 25° C. A pitch product having a softening point of 105° F. and free carbon content of 19% may thus be obtained. This product will be found to be very satisfactory as a road material, or, if desired, it may be mixed with a portion of low free carbon tar and then the mixture again

distilled to further increase the free carbon content thereof.

It will be understood that the above examples represent only illustrative instances of this invention and that various tars such as coke oven, vertical retort, and other tars of comparatively low free carbon contents, such as water gas tar, may be distilled in accordance with my invention to form residual pitches and these pitches admixed with distillate from the tar undergoing the distillation operation or other distillate tar oils or hydrocarbon material to form resultant pitch products of desired consistency, i. e., having higher free carbon contents.

It will be noted that this invention results in the production of a pitch product of definite softening point or melting point and of a definite free carbon content from coke oven tar alone and therefore eliminates the necessity for mixing coke oven tar with gas house tar or other tar of high free carbon content to produce such pitch products. This is an important advantage of the present invention for not only does it result in economies in the production of these pitch products due to the elimination of the necessity for using the more costly gas house tar but it also permits the production of pitch products of desired free carbon content at coke oven or other installations where gas house tar is not at all available or not available in sufficient quantities to be used for admixture with coke oven or other tar in the production of a pitch product of desired free carbon content.

The pitch product resulting from the practice of this invention consisting of coal tar pitch admixed with the distillate oils obtained from the distillation of coal tar has been found to be stable and not as susceptible to seasonal temperature variations as products of similar melting points produced directly by distillation to form a residue of the desired melting point. This may be attributed to the free carbon content of the product and also to the fact that the invention results in a product from which low boiling oils have been partially or substantially completely eliminated and thus the relative proportion of the high boiling oils has been increased.

While preferred embodiments of this invention have been disclosed herein, it will be understood that the invention is not limited thereto as various changes in the details thereof might be made by those skilled in the art.

I claim:

1. The process of producing a pitch of predetermined melting point and free carbon content, which comprises distilling tar from the group: coal tar, water-gas tar, peat tar and shale tar, to a point where substantial decomposition and a desired amount of free carbon formation take place, but not beyond the point where a pitch capable of being cut back with liquid tar products is formed, to produce as a distillation residue a pitch having a substantially increased amount of free carbon as compared with the amount in the tar initially distilled, collecting the distillate and mixing a portion of the distillate with the residue of the distillation operation, and thereafter distilling the resultant mixture until it has the desired melting point and free carbon content.

2. The process of producing a pitch product of predetermined melting point and free carbon content, which comprises distilling tar from the group coal tar, water-gas tar, peat tar and shale tar to a point where substantial decomposition and a desired amount of free carbon formation take

place, but not beyond the point where a pitch capable of being cut back with liquid tar products is formed, to produce as a distillation residue a pitch having a substantially increased amount of free carbon as compared with the amount in the tar initially distilled, collecting the distillate, admixing the distillate with the residue from the distillation operation, and then distilling the resultant mixture to obtain a pitch product having a free carbon content substantially in excess of that obtained by a single distillation, without substantial decomposition, of such tar to produce pitch of the same melting point.

3. The process of producing a pitch product of predetermined melting point and free carbon content which comprises distilling with substantial but limited decomposition a tar of the group consisting of water gas tar and tar resulting from the distillation of coal, shale, or peat until a resultant pitch residue having a desired substantially increased amount of free carbon as compared with the amount of free carbon in the tar initially distilled but yet capable of being cut back with liquid tar products is obtained, admixing a quantity of oil distillate of a tar of the group consisting of water gas tar and tar resulting from the distillation of coal, shale, or peat with the pitch residue and thereafter distilling the resultant mixture until a pitch product of desired melting point and free carbon content is produced.

4. The process of producing a coal tar pitch product of predetermined melting point and free carbon content, which comprises distilling a coke oven tar with substantial but limited decomposition until a distillation residue is produced having a softening point of above about 175° F. and containing a substantially increased amount of free carbon as compared with the amount of free carbon in the tar initially distilled and the temperature of the vapors coming off from the still is not more than 400° C., collecting the distillate, admixing the distillate with the residue from the distillation operation, and then distilling the resultant mixture to obtain a pitch having a free carbon content substantially in excess of that obtained by a single distillation, without substantial decomposition, of such tar to produce pitch of the same melting point.

5. The process of producing a coal tar pitch product of predetermined melting point and free carbon content, which comprises fractionally distilling coal tar with substantial but limited decomposition until a liquid pitch having a softening point of at least 250° F. and a substantially increased amount of free carbon as compared with the amount of free carbon in the tar initially distilled, is obtained as a distillation residue, separately collecting the fractions, and admixing the heavy oil fractions obtained from the distillation of said tar with the residue of said distillation operation to form a pitch product having the desired melting point and free carbon content.

6. The process which comprises fractionally distilling coal tar with substantial but limited decomposition to liquid pitch having a softening point of above about 175° F. and a substantially increased amount of free carbon as compared with the amount of free carbon in the tar initially distilled, but not beyond the point where a pitch capable of being cut back with liquid tar products is produced as a distillation residue, separately collecting distillate fractions so as to obtain an oil having a specific gravity of at least 1.1 at 25° C., admixing said oil with the residue

of the distillation operation, and thereafter distilling the resultant mixture until it has the desired melting point and free carbon content.

7. The process which comprises distilling coal tar with substantial but limited decomposition until the temperature of the vapors coming off from the still is not more than 400° C. and a pitch having a softening point of approximately 300° F. and a substantially increased amount of free carbon as compared with the amount of free carbon in the tar initially distilled is obtained as a distillation residue, collecting the distillate and mixing the residual pitch with distillate oil, and distilling the resultant mixture until a pitch of approximately 108° F. softening point is formed.

8. The process which comprises distilling coal tar with substantial but limited decomposition until a liquid pitch having a softening point above about 175° F. and not more than 300° F. and a substantially increased amount of free carbon as compared with the amount of free carbon in the tar initially distilled is obtained as a distillation residue, collecting the distillate, admixing the residual pitch with crude coal tar and with an oil of the group consisting of coal tar distillate oil and water gas tar distillate oil, and distilling the resultant mixture to form a pitch product.

9. The process which comprises distilling coal tar with substantial but limited decomposition until a residual pitch having a softening point of above about 175° F., and having a substantially increased amount of free carbon as compared with the amount of free carbon in the tar initially distilled, and capable of being cut back with liquid tar products, is obtained as a distillation residue, collecting the distillate, mixing the residual pitch with an oil of the group consisting of coal tar distillate oil and water gas tar distillate oil, and with crude coal tar, and distilling the resultant mixture to form a pitch product having a softening point of at least 104° F.

10. The process which comprises distilling with substantial but limited decomposition coke oven tar of a specific gravity of approximately 1.18 at 25° C. and a free carbon content of approximately 7 per cent to a residual pitch having a softening point of approximately 300° F. and having a substantially increased amount of free carbon as compared with the amount of free carbon in the tar initially distilled, collecting the distillate, admixing the residual pitch with the distillate, and distilling the resultant mixture until a pitch of approximately 108° F. softening point and having a free carbon content of approximately 27 per cent is formed.

11. The process which comprises distilling with substantial but limited decomposition coal tar to a residual pitch having a softening point of at least 250° F. and a substantially increased amount of free carbon as compared with the amount of free carbon in the tar initially distilled and which is capable of being cut back with liquid tar products, and admixing said residual pitch with coal tar oil having a specific gravity of at least 1.1 at 25° C. to produce a pitch product.

12. The process which comprises fractionally distilling coal tar with substantial but limited decomposition to a residual pitch capable of being cut back with liquid tar products, said residual pitch having a softening point of approximately 300° F. and a substantially increased amount of free carbon as compared with the amount of free carbon in the tar initially distilled, separately collecting the various fractions,

and admixing the heavier fractions distilled off between 300° and 400° C. and having a specific gravity of approximately 1.13 at 25° C. with a portion of the residual pitch to obtain a pitch product.

13. The process which comprises fractionally distilling coke oven tar with substantial but limited decomposition to a residual pitch having a substantially increased amount of free carbon as compared with the amount of free carbon in the tar initially distilled and a softening point of approximately 300° F., separately collecting the various fractions, admixing the fractions distilled off between 300° C. and 400° C. having a specific gravity of approximately 1.13 at 25° C. with a portion of the pitch in the proportions of about 48 parts distillate to about 52 parts pitch to form a pitch product.

14. The process which comprises distilling with substantial but limited decomposition a tar of the group consisting of water gas tar and tar resulting from the distillation of coal, shale, or peat until a pitch residue having a softening point of above about 175° F. and having a substantially increased amount of free carbon as compared with the amount of free carbon in the tar initially distilled and capable of being cut back with liquid tar products is obtained, admixing a quantity of heavy oil distillate of a tar of the group consisting of water gas tar and tar resulting from the distillation of coal, shale, or peat, said oil distillate having a specific gravity of at least 1.1 at 25° C. with the pitch residue, and thereafter distilling the resultant mixture until a pitch product having a free carbon content of at least 12 per cent by weight is obtained.

15. The process which comprises distilling with substantial but limited decomposition a tar of the group consisting of water gas tar and tar resulting from the distillation of coal, shale, or peat until a pitch residue having a softening point of above about 175° F. and having a free carbon content of not less than about 28 per cent by weight and capable of being cut back with liquid tar products is obtained and then admixing with said pitch residue a heavy oil distillate of tar of the group consisting of water gas tar and tar resulting from the distillation of coal, shale, or peat, said oil distillate having a specific gravity of not less than about 1.1 at 25° C. in quantities sufficient to produce a pitch product having a free carbon content of at least 12 per cent by weight.

16. The process of producing a coal tar pitch product of predetermined melting point and free carbon content, which comprises distilling a coke oven tar with substantial but limited decomposition until a distillation residue is produced having a softening point of above about 175° F. and containing a substantially increased amount of free carbon as compared with the amount of free carbon in the tar initially distilled and the

temperature of the vapors coming off from the still is not more than 400° C., collecting the distillate, admixing a portion of the distillate with the residue from the distillation operation, and thereafter distilling the resultant mixture until it has the desired melting point and free carbon content.

17. In the process of producing a coal tar pitch product of predetermined melting point and free carbon content, the steps which comprise distilling with substantial but limited decomposition coal tar to pitch having a softening point of above about 175° F. and then refluxing the residue until a pitch product of substantially increased amount of free carbon as compared with the amount of free carbon in the tar initially distilled and having a desired softening point of not more than 300° F. is produced.

18. The process of producing a coal tar pitch product of predetermined melting point and free carbon content, which comprises distilling coke oven tar to a point where substantial decomposition and a desired amount of free carbon formation take place but not beyond the point where a pitch capable of being cut back with liquid tar products is formed to produce as a distillation residue a pitch having a substantially increased amount of free carbon as compared with the amount in the tar initially distilled, collecting the distillate and mixing a portion of the distillate with the residue of the distillation operation, and thereafter distilling the resultant mixture until a pitch having a desired melting point and free carbon content is produced.

19. The steps which comprise distilling with substantial but limited decomposition coal tar to a pitch residue having a softening point of above about 175° F. and then refluxing the residue and accompanying the refluxing operation with the removal of a portion of the condensate to obtain a pitch product having a softening point of not more than 300° F. and a substantially increased amount of free carbon as compared with the amount of free carbon in pitch produced by a single distillation, without substantial decomposition, of coal tar to produce pitch of the same melting point.

20. In the process of producing coal tar pitch product of predetermined melting point and free carbon content, the steps which comprise distilling with substantial but limited decomposition coal tar to pitch having a softening point of above about 175° F. and then refluxing the residue until a pitch product is obtained having a substantially increased amount of free carbon as compared with the amount of free carbon in pitch produced by a single distillation, without substantial decomposition, of coal tar to produce pitch of the same melting point.

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