H. W. ROBINSON.

TREATMENT OF COAL TAR OR ITS PRODUCTS FOR THE REMOVAL OF THE PROPERTIES THEREIN TENDING TO INDUCE PITCH CANCER.

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**1,114,045.**

Patented Oct. 20, 1914.

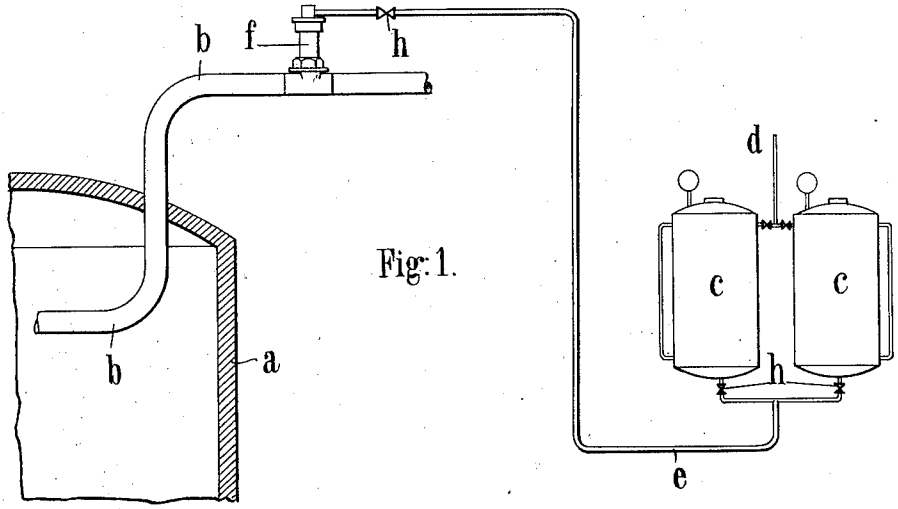



Fig: 1.

Fig: 2.

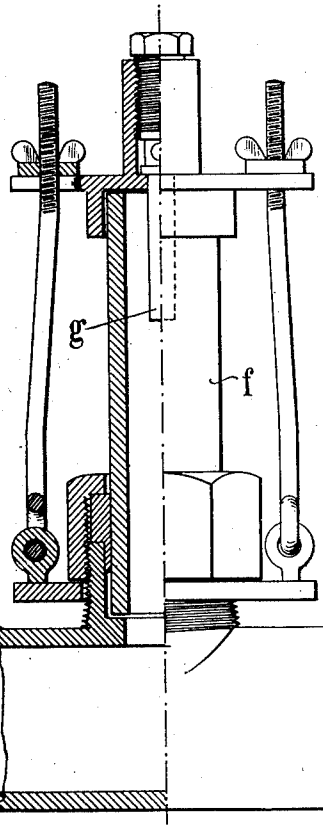
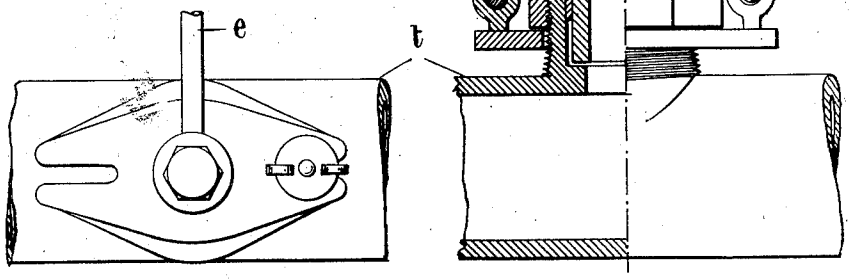



Fig: 3




Witnesses

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# UNITED STATES PATENT OFFICE.

HERBERT W. ROBINSON, OF SEDGLEY, ENGLAND.

## TREATMENT OF COAL-TAR OR ITS PRODUCTS FOR THE REMOVAL OF THE PROPERTIES THEREIN TENDING TO INDUCE PITCH-CANCER.

1,114,045. Specification of Letters Patent. **Patented Oct. 20, 1914.**

Application filed February 11, 1914. Serial No. 817,992.

*To all whom it may concern:*

Be it known that I, HERBERT WILLIAM ROBINSON, a subject of the King of Great Britain, and resident of Turls Hill House, Sedgley, in the county of Stafford, England, have invented certain new and useful Improvements Relating to the Treatment of Coal-Tar or Its Products for the Removal of the Properties Therein Tending to Induce Pitch-Cancer, of which the following is a specification.

Recent experiments have been directed to the problem of ascertaining the causes of cancer among workers in the industries producing and using pitch, and particularly in the manufacture of briquets wherein the pitch is employed as a binding agent for the coal dust. A recent publication on this subject "*The Problem of the Gas Works Pitch Industries and Cancer*" issued under the auspices of the John Howard McFadden researches (John Murray, Albemarle Street, London, W. 1912) gives particulars of researches showing what results have been obtained in experiments with samples of pitch and oils obtained from coal tar; these experiments showed that cancer would only be induced when two different classes of substances were present, these being known under the names of auxetics and kinetics respectively. Either of these alone will not induce cancer, but when both are present cancer may be induced in workers who are constantly working in the presence of dust containing the substances. The auxetics are certain agents present in dead matter, and are exciters of reproduction which stimulate cell-multiplication; among them may be mentioned creatin, xanthin (which contain the amidin grouping N—C—N) methylamin (a primary amin) leucin and tyrosin (amino-acids). The kinetics are alkaloids which excite amœboid movements in the blood cells. Most alkaloids have this property.

Various ways were suggested of treating either the pitch or the tar in order to remove from it either the auxetics or the kinetics, and so to render it innocuous in so far as the inducing of pitch cancer was concerned. It was suggested for instance that the desired result could be obtained by mixing a considerable amount of water with the tar when the water would dissolve out some of the constituents of the tar. The aqueous solution was then to be separated from the tar by centrifugal action or otherwise. Experiments on the resulting pitch and oils showed that the kinetics could be removed in this way, and usually also the auxetics. Now this process is not a practicable one, for the present at any rate, from the point of view of the tar distiller. The expense is too great and valuable constituents of the tar are dissolved out by the water. I have therefore made some experiments with the object of ascertaining whether the addition of some chemical substance to the tar would produce the desired result at a comparatively small expense, and without deleteriously affecting the value of the tar and of the products obtained therefrom. In so far as these experiments have been carried up to the present, it has been found that an addition of even small quantities of formaldehyde to the tar at a convenient stage in the distillation process, produces at least beneficial results, the pitch obtained being free from auxetics, or containing only small traces so that it would be comparatively harmless in so far as the risk of inducing cancer is concerned. The action of the formaldehyde is doubtless due to its property of condensing organic nitrogen compounds, these being the compounds in the tar which by the research above referred to, are shown to have auxetic properties.

In place of formaldehyde, para-formaldehyde, or other aldehydes may be used, as it is found that the action in question is due to the presence of an aldehyde, and practically all aldehydes act, to some extent at least, to get rid of auxetics. For example, acetic aldehyde may be used, or other aldehydes of the fatty group. The amount of the aldehyde added and the stage in the process at which it is added, will vary according to particular requirements and to the condition of the tar or pitch to be treated. It is not essential for most purposes to add the aldehyde in a pure form, and substances containing a suitable aldehyde or capable of yielding an aldehyde during the subsequent treatment of the product, may be added to the tar or pitch. For example, sugar decomposes at a temperature below 250° C., with the formation of formaldehyde, and for some purposes it may be cheaper or better to add sugar or the like to the product before or during the heating process in order

to insure the presence of sufficient aldehyde to render the resulting pitch substantially free from auxetics.

When formaldehyde is to be added to the tar during the distillation, it is generally most convenient to effect the distillation in the ordinary way until the carbolic acid and the earlier creosote fractions have been taken off. When the temperature of the residue in the still reaches about 250° to 270° C., the distillation of further fractions consisting of heavy oils and so forth, is assisted by a current of steam introduced for example through a perforated pipe in the still, and the formaldehyde may be introduced conveniently with this steam. Alternatively, the temperature of the material in the still may be reduced if necessary, and the formaldehyde or a substance capable of yielding a suitable aldehyde may be simply added before heating further to continue the distillation. When the formaldehyde is added with steam, the means for effecting the addition may be as shown in the accompanying drawing.

In this, Figure 1 shows a portion of the apparatus, and Figs. 2 and 3 are detail views of a mixing jet used therein.

The steam is introduced into the still *a* (only part of which is shown) through a pipe *b* from a boiler capable of supplying steam at the required pressure. Commercial formalin is pumped through a pipe *d* into storage vessels *c* under a suitable air pressure, such as 20 to 30 pounds per square inch, and is supplied from these vessels as required through cocks *h* and a pipe *e* to a jet for mixing it with the steam. A suitable form of jet is shown in Fig. 2 in elevation, partly in section, and in Fig. 3 in plan. The pipe *e* terminates in a fitting at the top of the device and communicates with a jet *g* in a glass sight tube *f*. The lower end of this tube communicates with the steam pipe *b* near to its point of entry into the still *a*. With this construction, the feed of the formalin can be observed and the jet can be removed and replaced easily if it becomes choked, while the feed of the formalin can be regulated by the cocks *h* as required. The regulation can be effected also by varying the pressure in the vessels *c*, the size of the orifice of the nozzle *g*, the strength of the solution, or by any combination of these means. In the case of intermittent stills which are completely emptied after each distillation of a charge of tar, it has been found convenient to introduce about two-thirds of the total charge of formalin for each charge of tar, during the distillation as set forth above, and to add the remaining one-third after the withdrawal of the fire and while the pitch residue is cooling in the still. About one and a half gallons of commercial formalin (40% solution of formaldehyde) have been used in successful experiments for a charge of 2,000 gallons of tar, but the quantity can be varied to any extent according to the condition of the tar and the results shown by experiment. The object is to eliminate the auxetics as far as possible from the product, and to do this with the least expenditure of formalin or its equivalent.

In the case of continuous stills, the formalin is introduced into the compartments of the still where the heavy oil fractions are being distilled at temperatures above 250° C., and if jets are used as above described, the feed of formalin may be continuous in these compartments. The pitch drawn off from the last compartment will be found to be substantially free from auxetics, so that it would have little or no tendency to induce cancer among operatives working with it.

The invention is not limited to the particular ways of adding the formaldehyde or its equivalent as set forth above, but any suitable method may be adopted of adding it at any convenient stage in the distillation or to the pitch residue after the completion of the distillation.

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

1. A process for treating tar or pitch with the object of removing therefrom the properties tending to induce pitch cancer, the process consisting in mixing with the tar or pitch less than 1% of a substance yielding an aldehyde.

2. A process for treating tar or pitch with the object of removing therefrom the properties tending to induce pitch cancer, the process consisting in mixing with the tar or pitch less than 1% of a substance containing and yielding an aldehyde.

3. A process for treating tar or pitch with the object of removing therefrom the properties tending to induce pitch cancer, consisting in adding less than 1% of an aldehyde to the tar or pitch before the completion of the distillation.

4. A process for treating tar or pitch with the object of removing therefrom the properties tending to induce pitch cancer, consisting in adding less than 1% of formalin to the tar or pitch before the completion of the distillation.

5. A process for treating tar or pitch with the object of removing therefrom the properties tending to induce pitch cancer, consisting in adding less than 1% of a substance yielding an aldehyde to the tar during the distillation thereof at a stage after the separation of the carbolic acid fraction and before the separation of the heavy oil fraction.

6. A process for treating tar or pitch with the object of removing therefrom the properties tending to induce pitch cancer, con-

sisting in adding less than 1% of a substance yielding an aldehyde to the tar during the distillation thereof at a stage after the separation of the carbolic acid fraction and before the separation of the heavy oil fraction, and adding to the residue a further quantity of said substance after the completion of the distillation.

7. A process for treating tar or pitch with the object of removing therefrom the properties tending to induce pitch cancer, consisting in mixing a small amount of formalin with steam and spraying the mixture into the distillation still after the carbolic acid fraction has been separated and before the heavy oil fraction is separated.

8. A process for treating tar or pitch with the object of removing therefrom the properties tending to induce pitch cancer, consisting in spraying a small amount of formalin under pressure into steam, and supplying the mixture of steam and formalin into a still containing the tar at a stage after the separation of the carbolic acid fraction and before the separation of the heavy oil fraction.

In witness whereof, I have hereunto signed my name this 20th day of January, 1914, in the presence of two subscribing witnesses.

HERBERT W. ROBINSON.

Witnesses:
HUBERT A. GILL,
H. W. BLAKE.