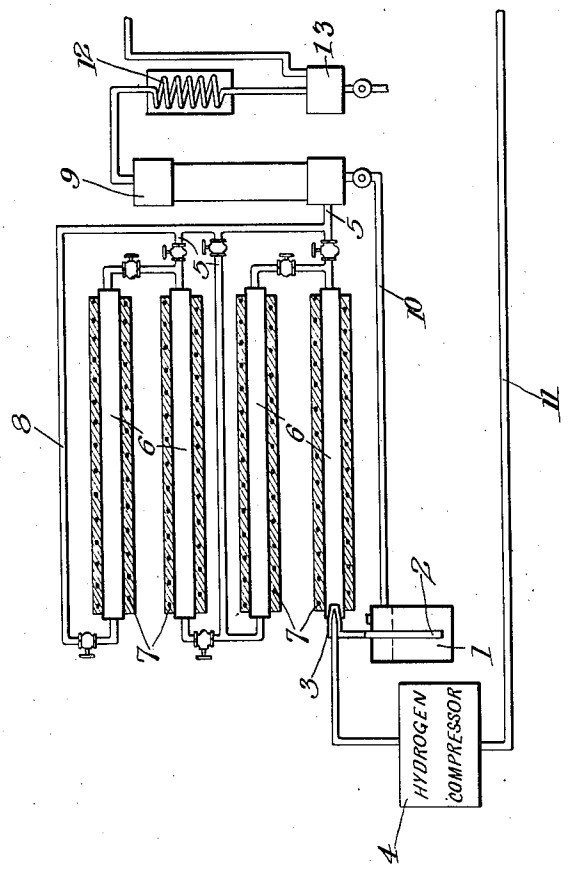
A. S. RAMAGE.

PROCESS OF PRODUCING PHENOLS.

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1,208,833.

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Witnesses

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UNITED STATES PATENT OFFICE.

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PROCESS OF PRODUCING PHENOLS.

1,208,833. Specification of Letters Patent. Patented Dec. 19, 1916.

Application filed April 20, 1916. Serial No. 92,469.

To all whom it may concern:

Be it known that I, ALEXANDER S. RAMAGE, a subject of the King of Great Britain, residing at Detroit, in the county of Wayne and State of Michigan, have invented certain new and useful Improvements in Processes of Producing Phenols, of which the following is a specification.

This invention is a process whereby phenols (phenol proper and its homologues, including cresol) are prepared by a treatment by hydrogenation, in presence of a suitable catalyst, of certain distillation products containing high boiling phenolic bodies of the creosol type.

It is known that the oils from the low-temperature distillation of coal consist chiefly of olefins, with variable proportions of so-called high-boiling tar acids, such as creosol and its homologues, boiling above 210° C. Such oils contain only very small proportions of cresol (cresylic acid) and practically no phenol, and are therefore valueless for creosoting purposes or for phenol-recovery. According to the present invention such high-boiling tar acids from the above or any other source are converted into phenol and cresol by hydrogen acting in presence of a suitable catalyst. It is probable that by this treatment the oxymethyl groups, together with a part of the methyl groups, are substituted by hydrogen, thereby producing a mixture containing phenol and cresol. The catalyst preferably used is nickel, reduced from nickel hydroxid at temperatures below 325° C. and distributed over long fibered asbestos. This is placed in tubes adapted to be heated by means of a resistance coil or otherwise. The oils containing the higher boiling tar acids are sprayed into the heated tubes containing the catalyst, together with and preferably by means of an excess of hydrogen gas. The oil-fraction employed for this purpose is preferably that collected between the temperatures of 210° and 300° C. The product is condensed, and if desired purified, in any usual manner, and the excess of hydrogen is collected for re-use if so desired.

Preferably the catalyst is heated to a temperature of about 600°–650° C., in which case I have been able to convert more than 50 per cent. of the contained high-boiling tar acids into phenol and cresol in the course of a single passage through a tube four inches in diameter and having a heated zone three feet, six inches in length. The product from the hydrogenation is fractionated, the fraction boiling between 175° and 208° C. being collected, and the higher boiling residue re-processed under the same conditions to continue or complete the conversion.

The described reaction proceeds, although relatively very slowly, at much lower temperatures than those mentioned above. For example, operating at about 300° C., ten passages of the material through a catalyst in a tube of the dimensions above mentioned were required to effect a forty per cent. conversion. It is therefore economical to operate at the higher temperature ranges mentioned above.

One form of apparatus which may be used for carrying out my invention is disclosed in the accompanying drawing, wherein the figure is a diagrammatic vertical sectional view.

In said drawing, 1 represents a tank for containing the material to be hydrogenized, from which tank this material is delivered through pipe 2 to an injector 3 which may be of the Schütte-Koerting type. Hydrogen is supplied to this injector under a pressure of about forty pounds, or sufficient to establish a flow of the vapors through the system. This hydrogen may be derived from any source whatever, illustrated in the present case as a compressor 4. The hydrocarbon thus atomized is sprayed into a series of interconnected tubes 6 containing the catalyst, each tube heated by a resistance coil 7. From the tube 6 a valved pipe 8 leads to a condenser 9, operating as a reflux, whereby the high-boiling or unconverted material is returned through pipe 10 to the tank 1 for reprocessing. Suitable valved connections 5 are provided whereby any desired number of tubes 6 may be operated in series according to the temperature and other conditions. The converted product passes in the form of vapor to the condenser 12, the condensate flowing to the receptacle 13 from which it may be withdrawn as desired. The uncondensed gases consisting largely of hydrogen, are carried through any suitable scrubbing or purifying system, and thereafter returned

by pipe 11 to the hydrogen compressor 4, together with such addition of hydrogen from extraneous sources as may be required.

The process carried out under the conditions described above proceeds without appreciable separation of carbon, and no tendency toward the conversion of the high-boiling tar acids into paraffins and olefins has been observed.

I claim:—

1. The process of producing phenols from high-boiling tar acids, which consists in subjecting a material containing such acids, at a suitable temperature, to the action of hydrogen in presence of a catalyst.

2. The process of producing phenols from high-boiling tar acids, which consists in introducing a material containing such acids in the form of a spray intimately mixed with hydrogen into a chamber containing a catalyst maintained at a suitable temperature, and condensing and collecting the resulting phenols.

3. The process of producing phenols from high-boiling tar acids, which consists in spraying a material containing such acids by means of a jet of hydrogen into a chamber containing a heated catalyst, and condensing and collecting the resulting phenols.

4. The process of producing phenols from high-boiling tar acids, which consists in spraying a material containing such acids by means of a jet of hydrogen into a chamber containing a catalyst maintained at a temperature of about 600°–650° C. and condensing and collecting the resulting phenols.

5. The process of producing phenols from high-boiling tar acids, which consists in subjecting such acids at a suitable temperature to the action of hydrogen in presence of a nickel catalyst.

6. In a process of producing phenols, the step which consists in converting high-boiling tar acids into phenols by reacting thereon with hydrogen.

In testimony whereof I affix my signature in presence of two witnesses.

ALEXANDER S. RAMAGE.

Witnesses:
CHARLES FENN,
FLORENCE JACOBI.