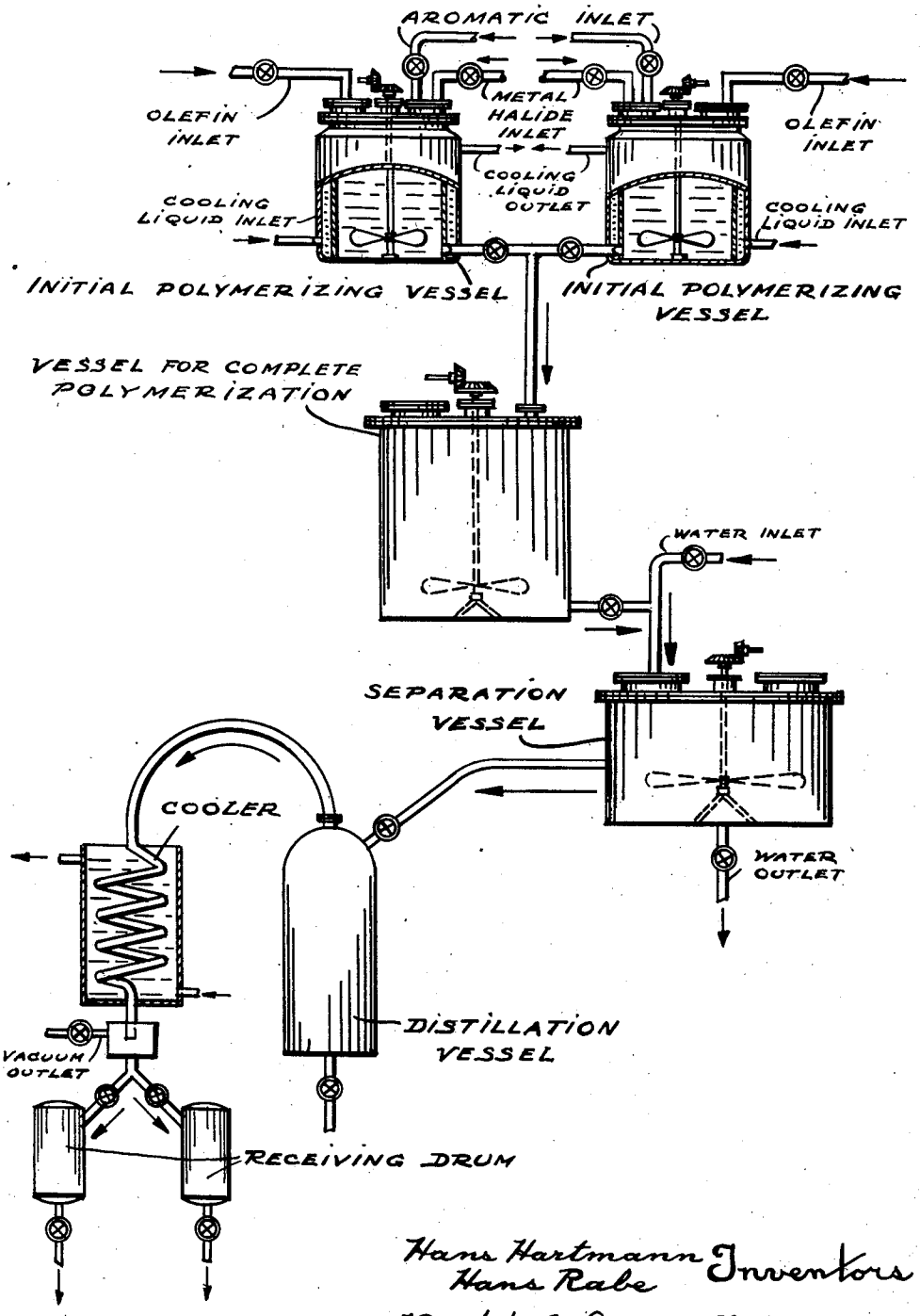
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PRODUCTION OF FLUORESCENCE PRODUCING SUBSTANCES SOLUBLE IN HYDROCARBON OILS

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PRODUCTION OF FLUORESCENCE PRODUCING SUBSTANCES SOLUBLE IN HYDROCARBON OILS

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5 Claims. (Cl. 196—78)

The present invention relates to a process for the preparation of fluorescence producing substances which are soluble in hydrocarbon oils and to the products obtainable thereby.

It is already known that by polymerization of aromatic hydrocarbons containing more than one nucleus under appropriate conditions of temperature and in the presence of catalysts of the Friedel-Crafts type substances are obtained which when dissolved in a mineral oil impart to the latter a green fluorescence.

These products show a from tarry to asphaltic consistency, may be dissolved in lubricating oils while warming and have the disadvantage that the greater part of them is precipitated again, after allowing the solution to stand and cool for some time, and that in many cases they give only a slight opalescence. These products furthermore are not soluble in benzines but are precipitated by them.

We have now found that fluorescence producing substances which are readily soluble, even at low temperatures, in hydrocarbon oils (by which expression we understand heavy oils or middle oils or lubricating oils or liquid motor fuels, such as benzines, or other liquid hydrocarbon fractions—whether they be of naphthenic or paraffinic base—) are obtained, if olefines of purely aliphatic constitution, which are liquid at ordinary temperature, are subjected to condensation with polynuclear organic substances in which none of the nuclei is saturated with hydrogen, said condensation being carried out at an elevated temperature in the presence of condensing catalysts of the Friedel-Crafts-type, in particular aluminium chloride. Of particular advantage are those of the aforesaid polynuclear substances whose aromatic nuclei are devoid of substituents. Examples of such substances are unsubstituted polynuclear aromatic hydrocarbons, such as naphthalene or anthracene or phenanthrene, or polynuclear heterocyclic compounds, such as carbazol. As olefines suitable as initial materials for this conversion may be mentioned the liquid products obtained by cracking paraffinic hydrocarbons, such as hard or soft paraffin wax, petrolatum, or oils entirely or mainly consisting of paraffinic hydrocarbons; or olefines obtained by dehydration of alcohols, such as from octodecyl alcohol.

The said condensation of olefines and polynuclear aromatic hydrocarbons is effected at temperatures between 70° and 250° C., preferably between 125° and 200° C. In order to carry off the great heat evolved by the said condensation, the treated materials are preferably diluted with an indifferent diluent, such as, for example, a benzine fraction boiling between 150° and 200° C., or another oil boiling at the same or higher temperatures, and simultaneously externally cooled. The amount of polynuclear aromatic hydrocarbons employed should be higher than 20 per cent of the amount of olefines employed, but is advantageously not higher than from 60 to 70 per cent of the said amount of olefines, since a surplus does not take part in the reaction, but acts only as a diluent and would have to be distilled from the resulting condensation product. The intensity of the fluorescence caused by a definite amount of the condensation product is the greater the greater is the amount of aluminium chloride employed. Preferably from 20 to 60 per cent of aluminium chloride with reference to the amount of the initial olefines to be condensed are employed. With larger amounts of aluminium chloride the resulting condensation products produce a fluorescence having a slightly blue tinge. The products obtained according to the present invention ordinarily have a mean molecular weight of between 600 and 800.

The accompanying drawing shows a schematic layout of equipment suitable for carrying out the process, and the drawing is provided with appropriate legends to explain the functioning of the equipment.

The fluorescence producing substances thus obtained are thickly liquid, viscous products which at ordinary temperature may be dissolved in hydrocarbon oils of naphthenic or paraffin base, such as in lubricating oils or in benzine. No precipitation or resinification occurs in the oils mixed with the said substances. The oils containing these substances are entirely clear in transparent light. An addition of from 0.1 to 0.2 per cent is sufficient for imparting to the oil a red color in transmitted light and a green color in reflected light, so that this oil, in its appearance, is in no way different from that of a Pennsylvanian oil.

The following examples will further illustrate how the present invention may be carried out in practice; the invention is, however, not restricted to these examples.

Example 1

1 kilogram of a fraction mainly consisting of olefines which boils between 90° and 150° C. under a pressure of 2 millimeters mercury gauge and which has been obtained by cracking soft paraffin wax is subjected to condensation with 1

kilogram of naphthalene in the presence of 0.4 kilogram af aluminium chloride. The reaction product having attained a temperature of 180° C. is allowed to cool down, then stirred with water and thereupon freed from the solid matter mainly consisting of the aluminium hydroxide formed. The reaction product is then heated to 150° C. under a pressure of 2 millimeters mercury gauge, 0.05 kilogram of oil thereby being distilled as first runnings. The liquid residue amounting to 1.9 kilograms represents an excellent fluorescence producing substance soluble in lubricating oils and benzine. If a mineral oil having a yellow color is mixed with 0.15 per cent of its weight of the said liquid residue the mineral oil obtains a remarkable green fluorescence and appears red in transmitted light.

Example 2

4 kilograms of a product obtained by cracking petrolatum, which boils above 100° C. under a pressure of 2 millimeters mercury gauge are polymerized with 1 kilogram of naphthalene, 0.5 kilogram of anthracene oil and 0.5 kilogram of carbazol in the presence of 1.2 kliograms of aluminium chloride at a temperature of 200° C. while stirring. The resulting products are worked up as described in Example 1. An oil is thus obtained in a nearly quantitative yield representing a fluorescence producing substance which when added to an oil causes that the latter appears red in transmitted light and green in reflected light.

Example 3

10 kilograms of a product obtained by cracking hard paraffin wax and which boils between 20° and 300° C. at atmospheric pressure is diluted with 5 kilograms of first runnings as obtained for example in the process described in Example 1. After adding 6 kilograms of naphthalene and 10 kilograms of aluminium chloride to the mixture the whole is polymerized for 2 hours at about 125° C. The resulting product is worked up as described in Example 1. In addition to 6 kilograms of first runnings 14.5 kilograms of a viscous product are obtained which when added to an oil showing no fluorescence in an amount of 0.2 per cent of the amount of oil, imparts to the oil a fluorescence which is in no way different from that of a Pennsylvanian oil.

Example 4

20 kilograms of a product obtained by cracking paraffin wax which has been diluted with 8 kilograms of a high boiling benzine fraction, are mixed with 5 kilograms of naphthalene and 7.5 kilograms of aluminium chloride added to the mixture in 3 portions amounting each to 2.5 kilograms, the second and third portions being added only after the reaction set up by the last preceding portion is finished (which may easily be determined by the decrease of the temperature). The whole of the aluminium chloride having been added to the reaction mixture, the latter is maintained for about 1 hour at 150° C. The reaction product, when worked up as described in Example 1 represents a fluorescence producing substance which when added to an oil in an amount of 0.1 per cent imparts to the latter a green fluorescence and a red appearance in transmitted light.

Example 5

10 kilograms of a product obtained by cracking paraffin wax are diluted with 10 kilograms of first runnings as obtained for example in the process described in Example 1 and, after an addition of 5 kilograms of aluminium chloride, polymerized while simultaneously introducing the cracking gases obtained in the cracking of paraffin wax. After the reaction temperature has attained about 100° C., 8 kilograms of naphthalene, 1 kilogram of anthracene and 0.5 kilogram of phenanthrene are added. The whole mixture is then heated to 150° C. and maintained at this temperature for one hour. The resulting products are subjected to distillation at 150° C. under a pressure of 1 millimeter mercury gauge, 18 kilograms of a substance being obtained as residue which when added to oils imparts to them a fluorescence as intensive as do the products obtained as described in the foregoing examples.

The addition of the said cracking gases does not materially increase the yield of the fluorescence producing substance but the amount of the portions which are separated by the distillation of the condensation products and which constitute oils having lubricating properties is considerably increased. In this manner the cracking gases are made use of at the same time for the preparation of lubricating oils.

In the appended claims by the term "condensation" (or "condensing") it is intended to include both condensation and polymerization.

What we claim is:—

1. A process for the production of a fluorescence producing substance which is soluble in hydrocarbon oils which comprises subjecting a mixture of an olefine of purely aliphatic constitution and liquid at ordinary room temperature with from 20 to 70 per cent with reference to said olefine of a polynuclear aromatic substance in which none of the nuclei is saturated with hydrogen to condensation at a temperature between 70° and 250° C. and in the presence of at least 20% with reference to said olefine of a condensing agent of the Friedel-Crafts type for a reaction time such as to produce fluorescence producing substances, soluble in hydrocarbon oils, and having a mean molecular weight of between 600 and 800.

2. A process for the production of a fluorescence producing substance which is soluble in hydrocarbon oils which comprises subjecting a mixture of an olefine of purely aliphatic constitution and liquid at ordinary room temperature with from 20 to 70 per cent with reference to said olefine of a polynuclear aromatic substance in which none of the nuclei is saturated with hydrogen to condensation at a temperature between 125° and 200° C. and in the presence of at least 20% with reference to said olefine of a condensing agent of the Friedel-Crafts type for a reaction time such as to produce fluorescence producing substances, soluble in hydrocarbon oils, and having a mean molecular weight of between 600 and 800.

3. A process for the production of a fluorescence producing substance which is soluble in hydrocarbon oils which comprises subjecting a mixture of a liquid at least mainly consisting of olefines liquid at ordinary temperature, which is obtained by cracking paraffin wax with from 20 to 70 per cent, with reference to the olefines contained in said liquid of naphthalene to condensation at a temperature between 70° and 250° C. and in the presence of at least 20% with reference to said olefine of aluminium chloride

for a reaction time such as to produce fluorescence producing substances, soluble in hydrocarbon oils, and having a mean molecular weight of between 600 and 800.

4. A process for the production of a fluorescence producing substance which is soluble in hydrocarbon oils which comprises subjecting a mixture of a liquid consisting at least mainly of olefines of purely aliphatic constitution and liquid at ordinary room temperature with from 20 to 70 per cent with reference to the olefines contained in said liquid of a polynuclear aromatic substance in which none of the nuclei is saturated with hydrogen and which is devoid of substituents to condensation at a temperature between 70° and 250° C. and in the presence of from 20 to 60 per cent by weight of said mixture of aliminium chloride for a reaction time such as to produce fluorescence producing substances, soluble in hydrocarbon oils, and having a mean molecular weight of between 600 and 800.

5. A process for the production of a fluorescence producing substance which is soluble in hydrocarbon oils, which comprises subjecting a mixture of a liquid olefine of purely aliphatic constitution with more than 20 per cent with reference to said olefine of a polynuclear aromatic substance in which none of the nuclei is saturated with hydrogen, to condensation at a condensing temperature and in the presence of at least 20% with reference to said olefine of a condensing agent of the Friedel-Crafts type.

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