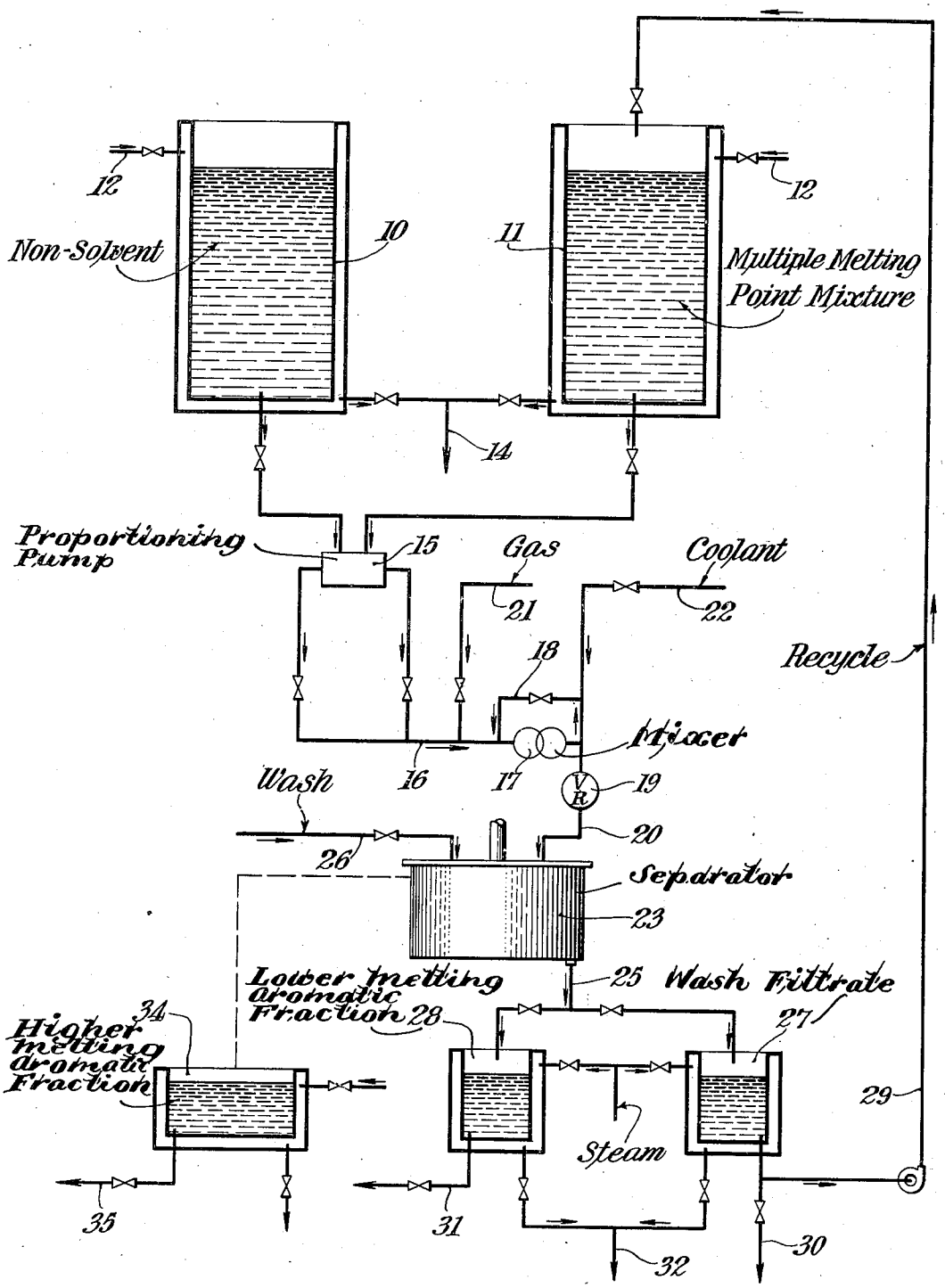
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SEPARATION OF DIFFERENT MELTING POINT MATERIALS

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## SEPARATION OF DIFFERENT MELTING POINT MATERIALS

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9 Claims. (Cl. 252—303)

This invention relates to a method for the partial separation of various melting point materials from mixtures thereof and more particularly to the concentration of higher melting point materials from lower melting point materials in a mixture thereof. It is, in a sense, a fractional solidification and separation by the aid of an emulsion whereby separation is facilitated. It is a continuation-in-part of my copending application, S. N. 274,412 filed May 18, 1939, and entitled "Separation of different melting point materials."

The principal object of my invention is to provide an improved method of separation of mixtures of materials which have separate melting points and of which at least one will solidify when cooled. Such separation of many materials is normally very difficult because of the tendency of the initially solidifying material to obstruct the flow of the liquid medium or the tendency of the liquid component to adhere to the solid matter. Filter rates, or settling rates are frequently very low, and therefore a liquid-solid separation is often impracticable.

I have discovered that I can obtain a highly effective separation of mixtures of materials having different melting points by forming an emulsion of said mixture with a non-solvent material and thereafter cooling said emulsion to such a point that one of the materials to be separated becomes filterable or separable by liquid-solid separation from the other. In this case the non-solvent surrounds the crystals or solid particles and the liquid particles and produces a free settling, free draining and free filtering system. I then separate the liquids from the solids and thereafter separate out the non-solvent material.

I believe that my invention is of rather general application inasmuch as I have found it entirely successful on materials of different characteristics. As an example of the materials on which I have had considerable success, I have separated wax from lubricating oils to materially improve the cold test thereof; and I have also been able to separate crude scale wax or finished high melting point wax from slack wax while producing a relatively low melting point foots oil.

In my prior co-pending application, I have noted that I can separate certain mixtures of animal and vegetable oils, the constituents of which have various melting points, and particularly mixtures of fatty acids such as a mixture of stearic and oleic acids and a mixture of linoleic and oleic acids with water as the non-solvent.

My present invention relates more particularly to the separation of mixtures of aromatic compounds such as a mixture of naphthalene and anthracene as well as mixtures of other similar products obtained in the distillation of coal tars. The invention may also be carried out in the separation of mixtures of isomers such as a mixture of alpha and beta naphthols or their derivatives with water as the non-solvent.

In accordance with a preferred form of embodiment of my invention, as shown on the attached drawing, which represents a general or typical flow sheet, it will be noted that I conveniently provide a plurality of tanks 10 and 11, in one of which I provide a suitable source of non-solvent liquid which in many cases can be water, and in the other tank I provide the mixture of materials having different melting points. These tanks may be steam-jacketed if high melting point materials are treated, and in such case the steam enters at 12 and the condensate is removed at 14.

Predetermined quantities of the non-solvent and multiple melting point mixture are then intimately mixed as by proportioning pump 15, the streams being consolidated in the manifold 16 and introduced into mixer 17. This mixer is of any suitable type of emulsifier or homogenizer, but I find that in many cases a rotary pump is adequate if provided with a by-pass 18 and a relief valve 19. It will be appreciated that by adjusting the capacity of the mixer 17 it will be possible to recycle the material through the mixer as many times as may be desirable so that the resulting material discharged through line 20 is suitably emulsified. I may find it desirable to add a gas through line 21 to the mixture in manifold 16 to aerate or otherwise expand the emulsion, and in some cases, I can use the gas to the exclusion of a liquid non-solvent material. I prefer to directly cool the emulsion as by the direct introduction of a coolant at 22 into the by-pass line 18, where this is possible, otherwise indirect cooling may be applied satisfactorily. Both direct and indirect cooling may be used, if desired.

Having established a suitable emulsion with a large interfacial area between the crystallized material and the non-solvent, I am able to obtain a very high rate of separation in the liquid-solid separator 23. Although this is indicated as of the centrifugal type, it may be either a centrifugal filter, having a foraminous filter basket,

or it may be of a clarifier type, having a solid bowl. It is, of course, in the contemplation of my invention, that the separator, the primary function of which is to separate liquids from solids, may be of any desired type, including pressure or vacuum filters of the continuous or other type, and having filter discs or a drum as may be desired. In some cases gravity settling may be practicable.

In the preferred form of embodiment of my invention, and for some types of mixtures to be separated, I find a centrifugal basket filter, having a perforated, rotating basket lined on the interior with a foraminous filter medium, to be most effective. This is rotated at such a speed that a high separating force is obtained, which may be in the range of 500 to 1,000 times gravity or more.

The cake formed is normally a fine-grained, free-filtering, porous type, and as the filtering force is gravitational by nature, the entire cake is submitted to the filtering force without an external crushing action. It is, of course, possible and usually desirable to wash such cake with a wash liquid from line 26, and such wash liquid will readily permeate the cake, and uniform drying and washing of the cake are thus possible.

The wash liquid may be either a diluent for the liquid material of the emulsion, or a liquid which is inert and non-reactive therewith. In the method of dewaxing an oil as disclosed in my patent, No. 2,168,140, issued August 1, 1939 on application S. N. 192,534, filed February 25, 1938, the temperature of the material is such that naphtha or other oil solvent is desirable. However, it may be preferable to use an inert and non-solvent wash liquid as set forth in my patent, No. 2,168,306, issued August 1, 1939 on application S. N. 232,263, filed September 29, 1938; and in such case the wash liquid does not dilute the liquid part of the emulsion and yet serves as a suitable displacing medium which is especially effective where the solid part of the emulsion is porous as in the instant case.

With the proper temperature control, the liquid that is removed from the filter cake is withdrawn through the line 25 either into the wash tank 27 or into the filtrate tank 28, each of which is suitably heated to assure continuity of flow. If the quality of the material in the wash tank 27 is found to have substantial value, it can be conveniently recycled through the line 29 to the multiple melting point mixture tank 11. The tanks 27 and 28 have discharge lines 30 and 31 and a steam condensate drain at 32. The filter cake, which is the high melting point material; may be removed from the separator and placed in the tank 34 and heated so that the higher melting material may be withdrawn as a liquid at 35.

My process is applicable to the separation of mixtures of aromatic-type organic compounds whether such mixtures are customarily separated by distillation, crystallization, or other fractionation procedure or are ordinarily difficultly separable. Such a mixture may be composed of homologous or isomeric aromatic compounds, or the components of the mixture may be structurally unrelated aromatic compounds which have relatively close melting points or boiling points, for example.

As an example of the application of my invention to the separation of a mixture of aromatic compounds of different melting points, I have separated a mixture of alpha naphthol and beta naphthol into fractions each respectively containing a greater proportion of one of the constituents. The particular mixture separated had a melting point of approximately 135° F. and was emulsified with water; the resulting emulsion was cooled to approximately 127° F. and charged to a centrifugal separator. The filtrate had a melting point of approximately 122° F., and the unwashed filter cake had a melting point of 185° F.

Naphthalene and anthracene may also be separated from a mixture thereof by emulsifying the mixture with a suitable non-solvent liquid and chilling the resulting emulsion to a temperature at which the anthracene solidifies (the melting point of anthracene is 421° F., and that of naphthalene is 176° F.). For some mixtures of anthracene and naphthalene water can be used as the non-solvent emulsifying liquid.

Separation of mixtures of other aromatic compounds such as mixtures of phenol (melting point 106° F.) and the cresols (melting point of ortho-cresol 88° F., melting point of meta-cresol 54° F., and melting point of para-cresol 95° F.) as well as mixtures of the cresols themselves can be accomplished by my process. Mixtures of the chlortoluenes (melting point of ortho-chlortoluene —34° F., melting point of meta-chlortoluene —54° F., and melting point of para-chlortoluene 46° F.), particularly mixtures of ortho-chlortoluene and para-chlortoluene, which are difficultly separable by distillation, can also be separated according to my invention.

In each case it is necessary only to select a suitable non-solvent, inert emulsifying fluid, whether liquid, gas, or both, whereby an at least temporarily stable emulsion is formed so that the desired separation can be effected in a centrifugal device or the like.

The principal advantage of the invention is that the materials are separated by their melting points rather than by their boiling points, and constant boiling mixtures or tarry decomposition products caused by distillation are avoided. This method of emulsification with a non-solvent fluid is applicable to virtually all separations where fractional distillation or the like is now used. The process is far quicker and more economical than the usual methods of crystallization since it makes possible a better separation with truly continuous operation and accurate temperature control. In addition, the use of the process is not limited to the separation of crystallizable materials; for, with amorphous materials, it is possible to use a freezing and crystallizing non-solvent liquid so that the resultant crystalline mass obtained upon cooling facilitates the desired separation.

Where the freezing point curve of such a mixture has a eutectic or minimum point, it will be appreciated that the composition of the filter cake, whether composed principally of one substance or the other in solidified form, will depend on which side of the eutectic point the composition of the starting mixture falls. If a particular mixture is being separated, one of the components may be added to the mixture as its composition approaches the eutectic point so that the resulting mixture can be further concentrated for the recovery of that component.

While I have shown and described a preferred form of embodiment of my invention, I am aware that modifications may be made thereto; and I, therefore, desire a broad interpretation of my invention within the scope and spirit of the disclosure herein and of the claims appended hereinafter.

I claim:

1. The method of separating a mixture of at least two aromatic compounds having different melting points, which comprises maintaining said mixture in liquid condition, forming an emulsion thereof with a non-solvent, non-reactive fluid, cooling said emulsion to a temperature such that at least one of the components of the initial mixture is partially solidified, and thereafter effecting a liquid-solid separation of the chilled emulsion to obtain a fraction containing at least one of said components in greater concentration than in the initial mixture.

2. The method of separating a mixture of at least two aromatic compounds having different melting points, which comprises maintaining said mixture in liquid condition, forming an emulsion thereof with a non-solvent, non-reactive liquid, cooling said emulsion to a temperature such that at least one of the components of the initial mixture is partially solidified, and thereafter effecting a liquid-solid separation of the chilled emulsion to obtain a fraction containing at least one of said components in greater concentration than in the initial mixture.

3. The method of separating a mixture of at least two aromatic compounds having different melting points, which comprises maintaining said mixture in liquid condition, forming an emulsion thereof with a non-solvent, non-reactive liquid and an inert, non-solvent gas, cooling said emulsion to a temperature such that at least one of the components of the initial mixture is partially solidified, and thereafter effecting a liquid-solid separation of the chilled emulsion to obtain a fraction containing at least one of said components in greater concentration than in the initial mixture.

4. The method of separating a mixture of at least two aromatic compounds having different melting points, which comprises maintaining said mixture in liquid condition, forming an emulsion thereof with a non-solvent, non-reactive liquid and an inert, non-solvent gas, cooling said emulsion to a temperature such that the material desired to be separated is solidified, and thereafter effecting a liquid-solid separation of the chilled emulsion to separate said solidified material from the remaining liquid.

5. The method of separating a mixture of alpha and beta naphthols, which comprises maintaining said mixture in liquid condition, forming an emulsion thereof with a non-solvent, non-reactive liquid and an inert, non-solvent gas, cooling said emulsion to a temperature such that one of the components of the initial mixture is partially solidified, and thereafter effecting a liquid-solid separation of the chilled emulsion to obtain fractions respectively containing alpha naphthol and beta naphthol in greater proportion than in the initial mixture.

6. The method of separating a mixture of alpha and beta naphthols, which comprises maintaining said mixture in liquid condition, forming an emulsion thereof with water and an inert, non-solvent gas, cooling said emulsion to a temperature such that one of the components of the initial mixture is partially solidified, and thereafter effecting a liquid-solid separation of the chilled emulsion to obtain fractions respectively containing alpha naphthol and beta naphthol in greater proportion than in the initial mixture.

7. The method of separating a mixture of anthracene and naphthalene, which comprises maintaining said mixture in liquid condition, forming an emulsion thereof with a non-solvent, non-reactive liquid and an inert, non-solvent gas, cooling said emulsion to a temperature such that one of the components of the initial mixture is partially solidified, and thereafter effecting a liquid-solid separation of the chilled emulsion to obtain fractions respectively containing anthracene and naphthalene in greater proportion than in the initial mixture.

8. The method of separating a mixture of phenolic compounds, which comprises maintaining said mixture in liquid condition, forming an emulsion thereof with a non-solvent, non-reactive liquid and an inert, non-solvent gas, cooling said emulsion to a temperature such that at least one of the components of the initial mixture is partially solidified, and thereafter effecting a liquid-solid separation of the chilled emulsion to obtain a fraction containing at least one of said components in greater concentration than in the initial mixture.

9. The method of separating a mixture of phenol and cresols, which comprises maintaining said mixture in liquid condition, forming an emulsion thereof with water and an inert, non-solvent gas, cooling said emulsion to a temperature such that one of the components of the initial mixture is partially solidified, and thereafter effecting a liquid-solid separation of the chilled emulsion to obtain fractions respectively containing phenol and cresols in greater proportion than in the initial mixture.

AUGUST HENRY SCHUTTE.