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[54] **METHOD FOR INCREASING CONVERSION EFFICIENCY FOR OXIDATION OF AN ALKYL AROMATIC COMPOUND TO AN AROMATIC CARBOXYLIC ACID**

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[52] **U.S. Cl.** **562/414**

[58] **Field of Search** 562/414

[56] **References Cited**

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[57] **ABSTRACT**

A method and system for increasing conversion efficiency of aromatic alkyl reactant to aromatic carboxylic acid product and for improving the quality of the product, are disclosed. The method and system provide for the continuous production of an aromatic carboxylic acid by the liquid phase, exothermic oxidation of an aromatic alkyl in a vaporizable solvent in an oxidation reactor. The reactor makes use of a vented, overhead condenser system and a separator system for condensation of vaporized reactor material, separation of the condensed solvent therefrom, and reflux of separated solvent back into the reactor. The improvement comprises combining the reactor liquid feedstream with the refluxed solvent upstream from the oxidation reactor.

7 Claims, 2 Drawing Sheets

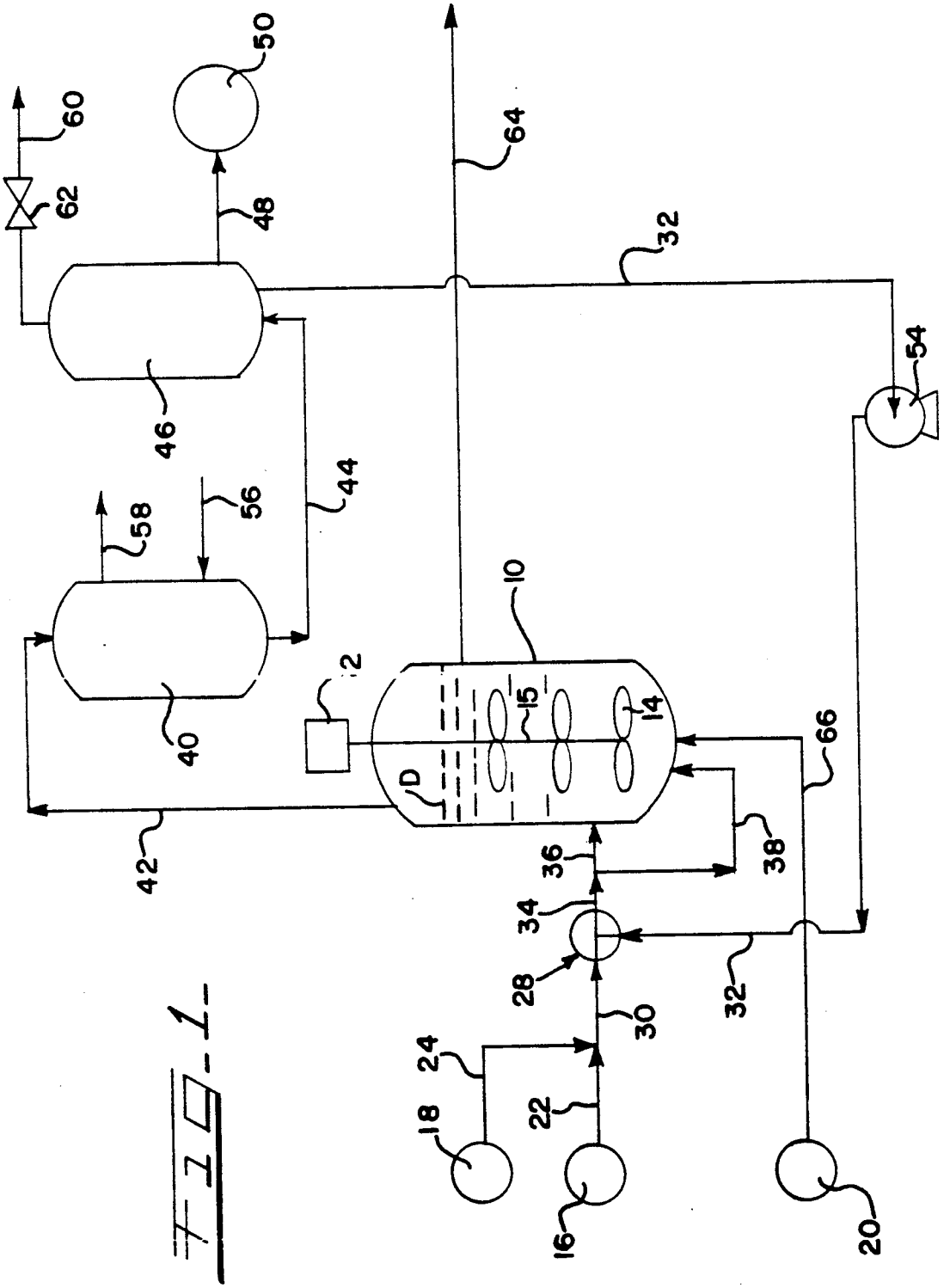

FIG. 1

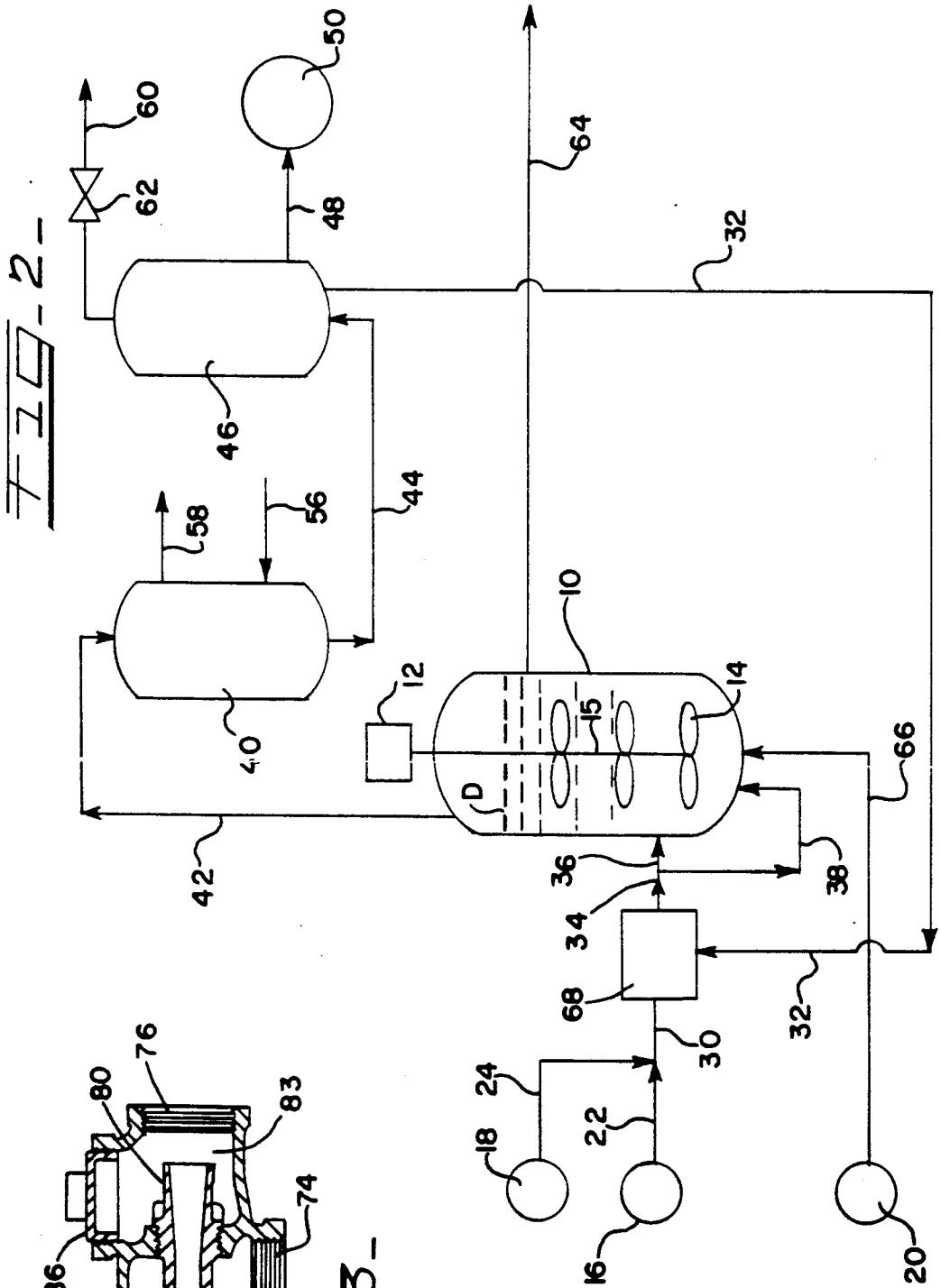

FIG. 2

FIG. 3

METHOD FOR INCREASING CONVERSION EFFICIENCY FOR OXIDATION OF AN ALKYL AROMATIC COMPOUND TO AN AROMATIC CARBOXYLIC ACID

FIELD OF THE INVENTION

This invention relates generally to the continuous, liquid phase oxidation of an aromatic alkyl to an aromatic carboxylic acid. More particularly, the present invention concerns a method and system for increasing reactor conversion efficiency and for improving the aromatic carboxylic acid product quality as well.

BACKGROUND OF THE INVENTION

Liquid phase oxidation of an aromatic alkyl to an aromatic carboxylic acid is a highly exothermic chemical reaction. Volatilizable aqueous acidic solvents are used to contain the reaction mixture and to dissipate the heat of reaction. Conventionally, the oxidation of aromatic alkyls in the liquid phase to form aromatic carboxylic acids is generally performed in a vented, well-mixed oxidation reactor, with a substantial portion of the heat generated by the exothermic oxidation reaction being removed by evaporating directly from the reaction mixture a portion of the aqueous solvent and aromatic alkyl contained within the reactor.

The materials vaporized as a result of the heat generated in the exothermic reaction, together with unreacted oxygen and other aqueous components that may be present, pass upwardly through the reactor and are withdrawn from the reactor at a point above the reaction mixture liquid level for the reactor. The vapors are passed upwardly and out of the reactor to an overhead reflux condenser system where the vaporized solvent, water and aromatic alkyl are condensed. The resultant condensate is thereafter separated, e.g., in a reflux splitter, into a portion having a relatively higher water concentration and a portion having a relatively lower water concentration. The separated portion having a relatively lower water concentration, now at a temperature less than the reactor contents' temperature, is refluxed back into the reactor by gravity. Conventionally, the refluxed portion of the condensate is returned directly to the reactor through a process line external to the reactor. The non-condensable gases, carried along with the vaporized reactor material, are vented.

In operation, the reactor is fed by a liquid feed stream containing the aromatic alkyl, aqueous acidic solvent and an oxidation catalyst. An oxygen-containing gas is separately introduced into the reactor for oxidizing the aromatic alkyl to the aromatic carboxylic acid in the presence of the catalyst.

The reaction mixture contained in the reactor typically comprises a suspension of crystalline aromatic carboxylic acid in liquid, volatilizable, aqueous acidic solvent as mother liquor. The mother liquor contains, in addition to dissolved catalyst, some dissolved aromatic carboxylic acid product and lesser amounts of partially-converted species of such product. The mother liquor can also include a minor amount of unreacted, aromatic alkyl.

Aromatic carboxylic acid product quality is measured by optical density. At present, optical density of the obtained product limits the oxidation reactor operating temperature and pressure, as well as the reactor throughput and mother liquor recycle rate into the reactor. Because of the commercial importance of the oxidation of aromatic alkyls, however, it is highly desirable to improve the reactor conversion efficiency and quality of aromatic carboxylic acids produced by the oxidation of aromatic alkyls.

The invention disclosed herein tends to diminish so-called reactor "entrance" effects, thought to be caused by an oxygen deficiency at the point where the reactor feedstream feeds the reactor. The invention disclosed herein also tends to minimize color-body generation, known to limit aromatic carboxylic acid plant operating flexibility and capacity.

SUMMARY OF THE INVENTION

The present invention is an improvement in a method and in a system for the continuous production of an aromatic carboxylic acid by liquid phase oxidation of an aromatic alkyl in an oxydation reactor. The improvement includes combining upstream from the reactor a liquid feed stream, containing an aromatic alkyl, with condensed acidic solvent medium that is refluxed back into the reactor, thereby providing a reflux-containing feed mixture, and then introducing the reflux-containing feed mixture into the oxidation reactor. A system embodying the present invention includes a liquid-liquid mixing means for effecting the "combining" step.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. **1** is a schematic process flow diagram of one embodiment of the present invention;

FIG. **2** is a schematic process flow diagram of another embodiment of the present invention; and

FIG. **3** is a detail on an enlarged scale showing a preferred liquid-liquid mixing means.

The drawings of FIGS. **1** and **2**, being process flow diagrams, are mere schematic illustrations. Accordingly, details which are not necessary for an understanding of the present invention have been omitted.

DETAILED DESCRIPTION OF PREFERRED EMBODIMENTS

Aromatic carboxylic acid is produced in an oxidation reactor at an elevated temperature and pressure by liquid phase, exothermic oxidation of an aromatic alkyl by an oxygen-containing gas in a vaporizable, aqueous acidic solvent medium. Oxidation of the aromatic alkyl to the aromatic carboxylic acid takes place in the aqueous acidic solvent medium in the presence of an oxidation catalyst. The conversion of aromatic alkyl to aromatic carboxylic acid is exothermic. Heat generated in the oxidation reaction is at least partially dissipated by vaporization of a portion of the solvent, water, aromatic alkyl and other vaporizable constituents of the reaction mixture present in the oxidation reactor. Vaporized reaction mixture constituents are withdrawn from the oxidation reactor, condensed in an overhead condenser system, and separated in a reflux splitter or a similar device into condensate portions having different water concentrations. Condensate portion having a relatively lower water concentration, and thus a relatively higher acidic solvent concentration, is fed back into the oxidation reactor.

A liquid feedstream for the oxidation reactor contains the aromatic alkyl, the acidic solvent medium, and an effective amount of an oxidation catalyst for effecting in the reactor a liquid phase oxidation of the aromatic alkyl, in the presence of oxygen, to the aromatic carboxylic acid. The improvement of the present invention

comprises combining the refluxed condensate portion with the oxidation reactor liquid feed stream upstream from the oxidation reactor to produce a reflux-containing liquid feed mixture which is at a temperature below the reactor contents' temperature. The reflux-containing feed mixture is then introduced into the oxidation reactor.

Referring to FIG. **1**, an elongated, vertically-disposed, continuous stirred-tank oxidation reactor **10** for oxidizing an aromatic alkyl to an aromatic carboxylic acid is shown. The oxidation reaction is continuous and proceeds in the liquid phase. The reactor **10** includes an agitator **12** which drives impeller blades **14**, fixed to an agitator shaft **15**. The reactor **10** further includes internal baffles (not shown). Each impeller blade **14** is rotated by the shaft **15** in a generally horizontal plane at a pre-selected rotational speed so that the contents of the reactor **10** are well mixed.

The contents of the reactor **10** are subjected to an elevated pressure and temperature sufficient to maintain the contained volatilizable solvent and aromatic alkyl substantially in the liquid state.

An aromatic alkyl, such as para-xylene, from a source **16**, and a volatilizable aqueous acidic solvent medium, such as a catalyst-containing aqueous acetic acid solution, from a source **18**, are combined to form a mixture. A liquid, reactor reflux stream from reflux splitter **48**, contained in transfer pipe **32** and having a relatively higher acetic acid concentration than the non-refluxed condensate portion exiting via discharge pipe **48**, is further combined with the formed mixture and is introduced into the reactor **10**, via side inlet **36**, as will be described in greater detail below. An oxygen-containing gas from a source **20** is introduced into the bottom of the reactor **10** via a gas inlet line **66**. The oxygen-containing gas serves to oxidize the aromatic alkyl to an aromatic carboxylic acid in the presence of the catalyst.

Localized pockets of relatively low oxygen concentration or relatively high aromatic alkyl or catalyst concentration, such as are in the vicinity of the reactor inlet or the reactor baffles, are thought to reduce conversion efficiency of aromatic alkyl to aromatic carboxylic acid. To counteract these so-called "entrance" and "other" effects, it has been discovered that, when the reactor feed stream containing the aromatic alkyl and the volatilizable aqueous acidic solvent medium (the solvent medium containing the oxidation catalyst) is combined with the liquid reactor reflux stream to produce a reflux-containing mixture and the reflux-containing mixture is then introduced into the reactor **10**, the overall conversion efficiency of aromatic alkyl to aromatic carboxylic acid is increased and the product quality is improved as well.

The prior art teaches recycling the reflux stream to the bottom of the reactor **10** and introducing the feedstream into the side of the reactor **10**. The present invention, however, contemplates introducing the combined reflux-containing liquid feed mixture either at the bottom or the side of the reactor **10**, as desired.

Accordingly, in one embodiment of this invention, a liquid-liquid mixing means, such as the piping "T" connection **28** (FIG. **1**), is provided. The aromatic alkyl is supplied to the "T" connection **28** by an inlet pipe **30** which carries the aromatic alkyl feed stream from source **16** via pipe **22** and the aqueous acidic solvent (containing the oxidation catalyst) from source **18** via pipe **24**. The aromatic alkyl and aqueous acidic solvent mixture is further combined with the reactor reflux stream in "T" connection **28**, with the reflux stream being introduced into "T" connection **28** by transfer pipe **32**. The resultant reflux-containing reactor feed exiting the "T" connection **28** is transferred via discharge pipe **34** into the reactor **10** either at side inlet **36**, bottom inlet **38**, or both, as desired. The reactor side-inlet **36** is located below the reactor liquid level D. The temperature of the reflux-containing feed mixture is less than the reactor temperature.

The source of oxygen for the oxidation of this invention can vary. Air and oxygen-enriched gas such as oxygen-enriched air or gaseous oxygen can be used. The oxygen-containing gas, from whatever source, supplied to the reactor **10** provides sufficient oxygen to result in an exhaust gas-vapor mixture containing from about two to about eight volume percent oxygen (measured on a solvent-free basis) when the oxidation reactor is in operation. For example, when each alkyl substituent on the aromatic ring of the aromatic alkyl is a methyl group, a feed rate of the oxygen-containing gas sufficient to provide oxygen in the amount of from about 1.4 to about 2.8 moles per methyl group will provide such two to eight volume percent oxygen concentration in the gas-vapor mixture in the condenser **40**.

In operation, the minimum pressure at which the reactor **10** is maintained is that pressure which will maintain a substantial amount of the aromatic alkyl present in the liquid phase and at least about 70 percent of the volatilizable, aqueous acidic solvent in the liquid phase. When the aqueous acidic solvent is an acetic acid-water mixture, suitable gauge pressures in the reactor **10** can be up to about 35 kg/cm^2 and typically are in the range of about 10 kg/cm^2 to about 30 kg/cm^2.

The process temperature employed is, on the one hand, low enough that the oxidation occurs with relatively low heat losses but, on the other hand, is high enough so that an economically desirable degree of conversion of the aromatic alkyl to the corresponding aromatic carboxylic acid is obtained. Process temperatures suitable for use in practicing the method of this invention generally are in the range of about 120° C. to about 240° C., preferably about 150° C. to about 230° C. Various narrower ranges may be preferred for a particular aromatic alkyl being oxidized. For example, when the aromatic alkyl is para-xylene, the preferred overall temperature range within the reactor **10** is about 175° C. to about 225° C., and the preferred temperature of the reflux-containing liquid feed mixture is about 85° C.

The residence time of the reactor is defined as the quotient of the reactor liquid volume divided by the liquid feed-stream flow rate into the reactor **10**. Typically, in a commercial operation, the residence time in the reactor **10** is in the range of about 20 to about 90 minutes.

Suitable aromatic alkyls for use in the method of this invention include toluene, ortho-, meta-, and para-xylene, and the trimethylbenzenes. The respective aromatic carboxylic acid products are benzoic acid, orthophthalic acid, isophthalic acid, terephthalic acid, and the benzenetricarboxylic acids. Preferably, the method of this invention is used to produce terephthalic acid, isophthalic acid, and trimellitic acid (1, 2, 4-benzenetricarboxylic acid). More preferably, the method of this invention is used to produce terephthalic acid.

Suitable volatilizable, aqueous acidic solvents for use in the method of this invention can be aqueous solutions of any C_2–C_6 fatty acid such as acetic acid, propionic

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