Title: PROCESS FOR ENHANCED ACETONE REMOVAL FROM CARBONYLATION PROCESSES

Abstract: A process for removing acetone from a methyl acetate, methyl iodide and acetone mixture includes the step of removing a side stream from the distillation zone rich in acetone. In a preferred embodiment the methyl acetate, methyl iodide and acetone mixture is contacted with water before or concurrently with fractional distillation. The process is particularly applicable to removing acetone by-product in a carbonylation process for the production of acetic anhydride.
PROCESS FOR ENHANCED ACETONE REMOVAL
FROM CARBONYLATION PROCESSES

BACKGROUND OF THE INVENTION

Field of the Invention

The present invention relates to an improved process of removal of acetone formed during the production of acetic anhydride by carbonylation. More particularly, the present invention relates to a separation process for removing acetone from a mixture which includes an alkyl halide, methyl acetate and acetone.

Background of the Invention

In recent years carbonylation processes, in particular rhodium catalyzed carbonylation processes, have become industrially important. One such important carbonylation process is for the production of acetic anhydride and the coproduction of acetic acid by contacting a mixture which includes methyl iodide and methyl acetate and/or dimethyl ether with carbon monoxide in the presence of a rhodium catalyst. Such rhodium catalysis processes are well known in the art and have been the subject of numerous patents.

An unwanted byproduct, acetone, is formed in the above-described carbonylation process, and typically accumulates in the acetic anhydride production system to a maximum level of about 5 weight percent, based on the total weight of the contents of the carbonylation reactor. Since the quantity of the acetone produced is relatively small, its value is not sufficient to justify the cost of its separating and purifying it to a sales grade acetone product. However, the acetone by-product in the process methyl acetate and methyl iodide recycle streams can lead to further undesirable by-products and/or reduction of the overall process efficiency. Among other things, acetone has been reported to inhibit the process catalyst. Acetone also forms "reducing substances" which cause quality problems in the acetic anhydride product. It has also been discovered that acetone reacts to form tar that must be removed from the process. An impediment to efficiently removing the acetone by-product from the carbonylation recycle streams is that
acetone is difficult to separate from methyl acetate and methyl iodide because of the
formation of binary azeotropes.

A number of acetone removal processes have been described in the patent literature,
for example U.S. Patent No. 4,252,748 describes a complex procedure in which all of the
methyl iodide, all of the acetone, and some methyl acetate is removed from a low-boiling,
recycle stream. The stream is then fractionated to obtain a methyl acetate acetone rich
stream which is then subjected to an azeotropic distillation with pentane, yielding methyl
acetate and an acetone-containing pentane stream. The acetone is extracted with water
and the pentane is recycled. This complex scheme requires the processing of large
volumes of effluent and entails a total of 5 operational steps (4 distillations and an
extraction). Furthermore, one would expect that a portion of the methyl iodide, the most
valuable process material in this stream, would accumulate in the pentane.

U.S. Patent No. 4,444,624 describes a system similar to the process of the '748
patent wherein a portion of the low boiler effluent is distilled with a countercurrent of
acetic acid to give a fraction rich in methyl iodide and methyl acetate and a second
fraction rich in methyl acetate and acetone, both of which contain very large quantities of
acetic acid. The acetone-containing fraction is further distilled to give a fraction
containing primarily methyl acetate and most of the acetone. The ratio of methyl acetate
to acetone is very high, generally about 50:1. The acetone is removed from the larger
amount of methyl acetate by azeotropic distillation with pentane and subsequent aqueous
extraction to remove the acetone. This process employs very large volumes of acetic
acid, generally around 1 part of acetic acid for every 2 parts of low boiler fraction to be
extractively distilled, and entails 4 operational steps (3 distillations and extraction.). This
process requires one less step than the process of the '748 patent only because it does not
attempt to purify the acetone from the acetone-water mixture. The processes of both the
'748 and '624 patents require the use of pentane which introduces the risk of product
contamination since pentane is not otherwise used in the acetic anhydride production
system.

U.S. Patent No. 4,717,454, discloses a process where acetone may be removed by
converting it to condensation products which may be removed from the production
system in the distillation as part of the ethylidene diacetate. This patent allows the acetone concentration to reach an equilibrium in the reaction system.

U.S. Patent No. 5,057,192 discloses a process for the removal of acetone from the acetic anhydride production system in which the low boiling stream having methyl acetate, methyl iodide, acetic acid and acetone is distilled. The process includes (1) obtaining from the production system a low-boiling stream comprising methyl acetate, methyl iodide, acetic acid and acetone; (2) distilling the stream of step (1) to obtain: (a) an overhead stream comprising methyl acetate, methyl iodide and acetone; and (b) an underflow stream comprising methyl acetate, methyl iodide, acetone and essentially all of the acetic acid; (3) extracting the step(2)(a) stream with water to obtain: (a) a methyl iodide phase containing methyl acetate; and (b) an aqueous phase containing methyl acetate, methyl iodide and acetone; and (4) distilling the aqueous phase to obtain: (a) a vapor phase comprising methyl acetate, methyl iodide and minor amounts of acetone and water; and (b) an aqueous stream containing methyl acetate and acetone.

U.S. Patent No. 5,244,545 discloses a process for the removal of acetone from the acetic anhydride production system. The process includes the steps of: (1) obtaining from the production system a low-boiling stream comprising methyl acetate, methyl iodide, acetic acid and acetone; (2) distilling the stream of Step (1) to obtain: (a) an overhead stream comprising methyl acetate, methyl iodide and acetone; and (b) an underflow stream comprising methyl acetate, methyl iodide, acetone and essentially all of the acetic acid; (3) (a) feeding the Step (2)(a) stream to the middle section of a distillation column; and (b) feeding water to the upper section of the distillation column; or, instead of steps (3)(a) and (3)(b), (c) feeding water to the Step (2)(a) stream and feeding the resulting mixture to the middle section of a distillation column; and (4) removing from the distillation column: (a) an overhead vapor stream comprising methyl iodide and methyl acetate; (b) an underflow liquid stream comprising water; and (c) a vapor stream comprising acetone and water from the lower section of the distillation column; or, instead of streams (4)(b) and (4)(c), (d) an underflow liquid stream comprising water and acetone.

U.S. Patent No. 5,262,014 discloses a process for removing acetone from a mixture comprising acetone, methyl acetate and methyl iodide, the process includes the steps: (a)
introducing a mixture comprising acetone, methyl acetate and methyl iodide into a distillation zone; (b) introducing water into the distillation zone at one or more points above the point of introduction into the distillation zone of the acetone/methyl acetate/methyl iodide mixture; (c) introducing acetic acid at one or more points at or above the point of introduction into the distillation zone of the acetone/methyl acetate/methyl iodide mixture; (d) removing from the distillation zone a heads product stream comprising methyl acetate and methyl iodide; and (e) removing from the distillation zone water, acetic acid and acetone at one or more points below the introduction point of the acetone/methyl acetate/methyl iodide mixture into the distillation zone.

Accordingly, there remains a need for improved processes that efficiently and effectively remove acetone from a mixture and desirably a mixture from a carbonylation process where acetic anhydride and acetic acid is produced by contacting carbon monoxide with a mixture comprising methyl iodide and methyl acetate and/or dimethyl ether in the presence of a catalyst system which includes a methyl halide promoter wherein the mixture includes methyl acetate, methyl iodide and acetone.

SUMMARY OF THE INVENTION

Advantageously, and quite surprisingly, the process of the present invention provides a more efficient means for the removal of acetone using a minimum of steps and processing equipment without the loss of any significant amounts of materials in the acetone disposal stream.

Broadly, the present invention is a process for the removal of acetone from a mixture which includes a methyl halide promoter, preferably methyl iodide, wherein the mixture includes methyl acetate, methyl iodide and acetone. In a preferred embodiment the mixture is from a carbonylation process where acetic anhydride and acetic acid is produced by contacting carbon monoxide with a mixture comprising methyl iodide and methyl acetate and/or dimethyl ether in the presence of a catalyst system, such as rhodium, which includes a methyl halide promoter. The process of the present invention includes the steps: (1) introducing a mixture which includes acetone, methyl acetate and methyl iodide into a first distillation zone; (2) withdrawing a side stream having acetone,
methyl acetate and methyl iodide from the first distillation zone; (3) introducing the side stream into a second distillation zone; (4) introducing water into the second distillation zone at substantially the same feed point as the side stream feed or at one or more points above the side stream feed; and (5) removing an overhead product comprising methyl acetate and substantially all of the methyl iodide fed to the second distillation zone and at one or more points below the side stream feed point an underflow product comprising acetone, methyl acetate and water.

Surprisingly, the process of the present invention allows a greater quantity of acetone to be removed from the smaller quantity of low boiling material from an acetic anhydride production system. Advantageously, smaller equipment can be used to remove acetone with reduced capital and operating expenses.

It is an object of the present invention to provide a process for the removal of acetone. More specifically, it is an object of the present invention to provide a process for the removal of acetone from a low-boiling stream having methyl acetate, methyl iodide and acetone.

It is another object of the present invention to provide a method for recovering essentially all of the methyl iodide from a low-boiling stream having methyl acetate, methyl iodide and acetone.

These and other objects and advantages will become more apparent to those skilled in the art in view of the following description and the accompanying drawings wherein like reference numerals designate similar parts throughout the various views and embodiments.

BRIEF DESCRIPTION OF THE DRAWINGS

The following figures are provided to illustrate the preferred configurations of the present invention in which:

FIG. 1 illustrates a schematic illustration of one embodiment of the process in which a mixture containing acetone, methyl acetate, acetic acid and methyl iodide is introduced to a first distillation column; a side stream is withdrawn having acetone, methyl acetate and methyl iodide from the middle zone of the first distillation column; the
side stream is introduced into a second distillation column; water is introduced into the
second distillation column at substantially the same feed point as the side stream feed or
at one or more points above the side stream feed; and overhead product is removed from
the second distillation column comprising methyl acetate and substantially all of the
methyl iodide fed to the second distillation column and at one or more points below the
side stream feed point an underflow product comprising acetone, methyl acetate and
water is removed.

FIG. 2 is an illustration of another embodiment of the process of the present
invention where a side stream comprising methyl acetate, water and essentially all of the
acetone fed to the second distillation column is withdrawn and where a portion of a liquid
bottom stream comprising essentially water is recycled to the water feed point for the
second distillation column.

FIG. 3 is an illustration of another embodiment of the process of the present
invention where a portion of the overhead stream from the first distillation column is
contacted with water in an aqueous extraction column to produce a product stream having
methyl acetate, methyl iodide, acetone and water.

FIG. 4 is an illustration of another embodiment of the process of the present
invention where in addition to the side stream and liquid bottom stream being withdrawn
from the second distillation column, a portion of the liquid bottom stream is used in an
aqueous extraction column to produce a product stream having methyl acetate, methyl
iodide, acetone and water and/or is recycled to the water feed point for the second
distillation column.

DETAILED DESCRIPTION OF THE INVENTION

The present invention will now be described in detail in conjunction with the
accompanying drawings. Referring to FIG. 1, a low-boiling mixture comprising methyl
acetate, methyl iodide, acetic acid and acetone is fed by conduit 10 to a first distillation
column 12. The low boiling mixture may be obtained from the acetic anhydride
production system described in Example 1 of U.S. Pat. No. 4,374,070, as well as other
known acetic anhydride manufacturing processes. The low boiling mixture is the portion
of the reactor effluent remaining after removal of the catalyst components, a substantial
amount of the low boiling components and essentially all of the acetic anhydride and higher boiling by-products. Typically, the low boiling mixture consists of about 75 to 45 weight percent methyl acetate, 30 to 15 weight percent methyl iodide and 20 to 5 weight percent acetic acid with the concentration of the acetone varying from about 8 to 4 weight percent, depending on the length of time the acetone removal process has been operated. The mixture also may contain a trace, e.g., about 0.1 to 0.5 weight percent, of acetic anhydride. While the mixture may contain significant amounts, e.g., 30 to 40 weight percent of acetic anhydride, the preferred process normally is carried out using a mixture from which substantially all of the acetic anhydride and other high boilers such as ethylidene diacetate have been removed elsewhere in the production system. The boiling points of the volatile materials in the process are given in Table 1 below:

<table>
<thead>
<tr>
<th>Material</th>
<th>Boiling Point (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MeI</td>
<td>42.5</td>
</tr>
<tr>
<td>Acetone</td>
<td>56.2</td>
</tr>
<tr>
<td>MeOAc</td>
<td>57.0</td>
</tr>
<tr>
<td>MeI / Acetone</td>
<td>42.4</td>
</tr>
<tr>
<td>MeI / MeOAc</td>
<td>42.1</td>
</tr>
<tr>
<td>MeOAc / Acetone</td>
<td>55.8</td>
</tr>
</tbody>
</table>

wherein MeI is methyl iodide, MeOAc is methyl acetate, and MeI/Acetone, MeI/MeOAc and MeOAc/Acetone are constant boiling mixtures (binary azeotropes) consisting of, by weight, 95% methyl iodide and 5% acetone, 97.3% methyl iodide and 2.7% methyl acetate, and 50% methyl acetate and 50% acetone, respectively.

Distillation column 12 is operated at ambient pressure, a base temperature of about 60° to 70°C maintained by a heat source such as reboiler 14, and a top temperature of about 40° to 50°C. to fractionate the low boiling mixture into (1) an overhead stream comprising methyl acetate, methyl iodide and acetone; (2) a side stream comprising methyl acetate, methyl iodide and acetone and (3) a bottom stream, i.e., a base product stream, comprising methyl acetate, methyl iodide, acetone and essentially all of the acetic acid, acetic anhydride and higher boilers fed to the column. The bottom stream is
removed from distillation column 12 via conduit 16 and may be recycled to the
carbonylation reactor along with additional methyl acetate, methyl iodide and catalyst
components. The primary function of column 12 is to remove all, or essentially all, of the
acetic acid from the low boiling mixture. To accomplish this objective, a substantial
portion of the methyl iodide and acetone fed to column 12 is removed with the acetic
acid.

The vaporized overhead stream is removed from distillation column 12 by means of
conduit 18 and passed through condenser 20 wherein substantially all of the stream is
converted to a liquid. A portion of the condensate from condenser 20 may be returned via
lines 22 and 24 as reflux to distillation column 12. The remainder of the condensate may
be recycled to the carbonylation system via line 26.

A side stream comprising methyl acetate, methyl iodide and acetone is removed
from distillation column 12 via line 28 and is passed through condenser 30 to convert
substantially all of the constituents of the stream to a liquid phase. The side stream 28
desirably has a ratio of acetone / methyl iodide of from 1:20 and preferably from 1:10 and
more preferably from 1:8.5 which advantageously allows for greater acetone removal
from a smaller quantity of low-boiling material. The side stream, exiting the condenser
30, is fed via line 36 to a second distillation column or fractional distillation column 38.
Desirably, the side stream 36 has a feed point at or near the middle of the second
distillation column 38. Optionally, a portion of the condensate from the overhead of
distillation column 12 not returned for reflux purposes, i.e., the material in line 26, can be
combined with the side stream 36 before being introduced into the second distillation
column 38, or the condensate in line 26 can be introduced at or substantially near the feed
point of stream 36.

Water is introduced into the second distillation column 38 via line 39 at
substantially the same feed point as the side stream feed 36 or at one or more points above
the side stream feed 36. The amount of water added to second distillation column 38 will
depend upon such factors as the composition of the acetone / methyl acetate / methyl
iodide mixture, the required compositions of the product streams and the number of
separation stages in the distillation zone. Typically, the ratio of water to mixture may
suitably be from about 1:1 to about 10:1.
The second distillation column 38 typically contains sufficient packing or other internals to provide a mass transfer adequate to remove essentially all the methyl iodide overhead, typically 8 to 20 ideal stages. The second distillation column 38 operates at approximately ambient pressure, at a base temperature of about 98° to 105° C maintained by a reboiler 40 and a top temperature of about 45° to 55°C. A vapor stream comprising methyl acetate, minor amounts of acetone and water, and essentially all of the methyl iodide, is removed from the top of distillation column 38 through line 42, condensed in condenser 44, and where at least a portion of the resulting liquid may be returned to distillation column 38 as reflux via line 46 or recycled to the carbonylation process by conduit 48.

An aqueous stream comprised of water, methyl acetate and most of the acetone, e.g., at least 75 weight percent of the acetone fed to distillation column 38, is removed as a liquid through line 50 and transported to a conventional waste water treatment plant. The acetone removal system may be operated in a manner that produces a process effluent via line 50 which contains a methyl acetate:acetone weight ratio of not more than about 3:1, and preferably not more than about 1:1. Furthermore, operation of the system provides for recovery of at least 95 weight percent, normally at least 99 weight percent, of the methyl iodide, the most valuable of the chemicals fed to the system. Thus, the amount of iodine present in the process effluent removed from the system via line 50 desirably does not exceed 200 parts per million, and preferably is less than 100 ppm.

Referring to FIG. 2, a modification of the process depicted in FIG. 1 is illustrated. In this embodiment, the lower section of fractional distillation column 38 is a liquid phase consisting substantially of water with the possibility of containing minute amounts of acetone and methyl acetate that collects at the bottom of the column. Preferably, the lower section of extractive distillation column 38 has less than about 5 total weight % of methyl acetate and acetone. The liquid phase is drained from the base of column 38 through conduit 50 and a portion is fed to heat source 40 which is then recycled back to the base of the column to maintain a base temperature of about 90° to 95° C. The remainder of the liquid phase is recycled, by means of line 51, to the upper portion of extractive distillation column 38 and desirably to a point at or above the feed for side
stream 36 and preferably is combined with the water feed line 39 prior to introducing the water into the second distillation column 38.

A second side stream is removed from the second distillation column 38 via line 52. The second side stream 52 includes from about 75-95 weight % of the acetone fed to the second distillation column 38 via line 36 and more preferably from 80-95 weight % of acetone fed to the second distillation column 38 via line 36 with the remaining weight % comprising water and methyl acetate. The second side stream 52 is condensed in condenser 54 and transported via line 56 to a suitable industrial waste treatment plant.

Optionally, if the acetic acid content in the low-boiler feed stream 10 is sufficiently low to be of little economic value, desirably, less than 1 weight % of the feed stream 10, the first distillation column can be omitted and the low-boiling stream 10 can be fed directly to the fractionation distillation column 38. In such a case, the process of the present invention would include the steps of: (1) introducing a mixture which includes acetone, methyl acetate and methyl iodide into a distillation zone; (2) introducing water into the distillation zone at substantially the same feed point as the mixture feed stream or at one or more points above the mixture feed stream; and (3) removing from the distillation column (a) an overhead vapor stream comprising methyl acetate and essentially all of the methyl iodide fed to the distillation column; (b) a side stream having acetone, methyl acetate and water; and (c) an underflow or bottoms product consisting substantially of water.

Referring to FIG. 3, another embodiment of the present invention is illustrated and represents a process similar to that depicted in FIG. 1, except with the following modifications. The portion of the condensate from condenser 20 not returned via lines 22 and 24 as reflux to distillation column 12 is fed by via line 26 to the upper portion of a counter-current, water extraction column 31. Typical ratios for the amount of reflux to extractor feed, i.e., the conduit 24 reflux : conduit 26 extractor feed, are from about 2:1 to 10:1. Water is fed to extraction column 31 via line 32 located near the bottom of extraction column 31. The water extraction column 31 is typically equipped with four beds packed having 0.625 inch pall ring packing material. The upwardly-flowing water extracts essentially all, e.g., at least 98 weight percent, of the acetone and a substantial portion, e.g., about 92 to 99 weight percent, of the methyl acetate. A small amount, e.g.,
about 5 weight percent, of the methyl iodide fed to the water extractor column 31 overflows with the aqueous phase. The remainder of the methyl acetate is removed from the base of water extraction column 31 via conduit 33 and may be recycled to the carbonylation reactor or used as the methyl iodide source in the tar removal process described in U.S. Pat. No. 4,388,217. The methyl iodide stream obtained from water extractor 31 typically has a purity of at least 95 weight percent and preferably at least 98 weight percent. The aqueous phase overflows extraction column 31 at or near the top and is transported through conduit 34, to the mid-section of fractional distillation column 38. The aqueous phase overflow from the water extraction column 31 is fed to the fractional distillation column 38 at substantially the same feed point as the side stream feed 36, or may be combined with the side stream 36 prior to being introduced into the second distillation column 38, or may be introduced into the second distillation column 38 at one or more points above the side stream feed 36.

Referring to FIG. 4, another embodiment of the present invention is illustrated and represents a process similar to that depicted in FIG. 3, except with the following modifications. The bottoms of fractionating distillation column 38 is a liquid phase consisting substantially of water, and desirably comprises greater than about 95 weight % water and less than about 5 weight % total of methyl acetate and acetone. Preferably, the liquid bottoms of fractionating distillation column 38 comprises greater than about 98 weight % water and less than about 2 weight % total of methyl acetate and acetone. The liquid phase is drained from the base of column 38 through conduit 50 and a portion is fed to heat source 40 which is then recycled back to the base of the column to maintain a base temperature of about 90° to 95° C. The remainder of the liquid phase can be recycled, by means of line 51, to either (a) a feed point above the location of the side stream 36 feed point and/or above the aqueous stream 34 feed point; and/or (b) is used as the water source for the water extraction column 31 along with fresh water provided by a fresh water make-up line, not shown. Desirably, up to about 60 weight percent of the liquid phase transported by conduit 51 may be recycled to water extraction column 31 for use as the water source.

A second side stream is removed from the second distillation column 38 via line 52. The second side stream 52 includes from about 75-95 weight % of the acetone fed to the
second distillation column 38 via line 36 and more preferably from about 80-95 weight % of acetone fed to the second distillation column 38 via line 36 with the remaining weight % comprising water and methyl acetate. The second side stream 52 is condensed in condenser 54 and transported via line 56 to a suitable industrial waste treatment plant.

The process of the present invention may be employed continuously or semi-continuously as necessary to lower the concentration of the acetone in the carbonylation reactor within a predetermined range. Operation of the carbonylation process in the presence of lower levels of acetone, e.g., acetone concentrations of about 2.0 to 2.5 weight percent based on the total weight of the reactor contents, results in the production of acetic anhydride containing lower levels of “reducing substances”. One of the purity specifications for acetic anhydride which is difficult to achieve is the level of “reducing substances”, a specification which is particularly important to manufacturers of cellulose acetate. A typical specification requires a permanganate reducing substances test value of at least 30 minutes according to a modification of the Substances Reducing Permanganate Test, American Chemical Society Specifications published in Reagent Chemicals, 6th Ed., American Chemical Society, Washington, D.C., pp. 66 and 68. The use of acetic anhydride containing lower levels of reducing substances decreases the amount of bleaching agents required in cellulose acetate manufacturing processes, thereby lowering the manufacturing costs and further enhancing the value of the acetic anhydride. The process provided by the present invention provides a means for producing, by the carbonylation processes described above, acetic anhydride which will more consistently pass the reducing substances test.

We also have found that lower acetone concentrations result in an increased production rate, an improvement in the color of the acetic anhydride product, a lowered tar formation rate and a decrease in the tendency of the tar formed to bind rhodium. At least a portion of the production rate increase is due simply to the reactor volume made available for more reactants by the lower volume of acetone present. For example, lowering the acetone level to about 1.4 weight % as described herein results in about 2 % increase in production rate due to increased useful reactor volume.

A reduction in the amount of tar produced by the carbonylation process requires the processing of lower amounts of catalyst-tar mixtures which in turn reduces significantly
the risk of rhodium losses in such processes. Due to the dramatic rise in the cost of rhodium, any process improvements which reduce the risk of its loss in the overall acetic anhydride production system have become increasingly important.

The present invention is illustrated in greater detail by the specific example presented below. It is to be understood that the example is an illustrative embodiment and is not intended to be limiting of the invention, but rather the invention is to be construed broadly within the scope and content of the appended claims.

The following example illustrates the operation of the process of the present invention in conjunction with an acetic anhydride production system as described in U.S. Pat. No. 4,374,070 wherein a mixture of methyl iodide and methyl acetate is contacted with carbon monoxide in the presence of a catalyst system comprising rhodium and a lithium salt at a temperature of about 160° to 220°C and at a pressure of about 21.7 to 83.7 bar absolute (about 300 to 1200 psig). In the carboxylation process, a feed mixture containing methyl acetate is continuously fed to a carboxylation reactor and a reaction product mixture containing acetic anhydride is continuously removed. The feed to the reactor is such so as to maintain within the reaction mixture about 500 to 1000 ppm rhodium, about 1500 to 3700 ppm lithium, about 7 to 35 weight percent methyl iodide and about 5 to 40 weight percent acetic acid.

The effluent from the liquid phase carboxylation reactor is processed to remove unreacted carbon monoxide and other non-condensible gases and catalyst components. Any dimethyl ether fed to the carboxylation reactor which is not converted to methyl acetate is also removed as a component of the non-condensible gases. The remainder of the effluent is fed to a distillation column from which a crude acetic anhydride/acetic acid mixture is obtained. The vaporized low-boiler stream removed at or near the top of the distillation column comprises methyl acetate, methyl iodide, acetic acid and acetone. The low-boiler stream is condensed and all or a portion, typically about 5 to 25 weight percent, of it is subjected to the acetone removal process of the present invention.

At the commencement of the operation of the acetone removal process, the concentration of the acetone in the reactor was 4.0 to 4.5 weight percent. All parts given are by weight and flow rates are parts per minute.
EXAMPLE

In accordance with the flow diagram in Figure 3, the low-boiler stream from carbonylation was fed through conduit 10 at a rate of 1000 parts to the lower midsection of distillation column 12. Distillation column 12 was maintained by a heat source 14 at a base temperature of 60° to 65°C to give an acetic acid underflow or bottoms stream removed by line 16. A vapor side stream was removed at a rate of 50 parts through conduit 28 and condensed in condenser 30. This material was then fed to the middle of distillation column 38 by means of conduit 36.

The vapor removed from the top of column 12 was condensed and a portion was returned to the column, via line 24, as reflux. The balance of the condensate was transferred through conduit 26 to the upper part of water extraction column 31 at a rate of 200 parts. Water was fed near the bottom of extractor 31 at a rate of 520 parts by conduit 32. Methyl iodide having a purity near 99% was removed from the water extraction column by conduit 33.

The aqueous phase from the top of water extractor 31 was removed at a rate of 590 parts and was fed via line 34 to the middle of fractionation distillation column 38. The side stream feed from distillation column 12 (line 36) feeds the fractionation distillation column 38 on the same tray. The fractionation distillation column 38 was maintained at a base temperature of 95 to 100°C by heat source 40. A vapor stream of methyl acetate, methyl iodide, and minor amounts of acetone and water was removed from distillation column 38 and condensed. A portion of the condensate was returned through conduit 46 to the top of column 38 as reflux. The balance of the condensate was returned via conduit 48 to the carbonylation system at a rate of 120 parts. Water containing methyl acetate and acetone was removed from the bottom of the fractionation column 38 by line 50 at a rate of 520 parts. The ratio of methyl acetate:acetone removed from the base of the column is in the range of 2:1 to 1:2.

The compositions of streams transported by conduits 10, 26, 28, 34, 48, and 50 are given in Table 2 as weight percentages based on the total weight of the stream.
Table 2

<table>
<thead>
<tr>
<th>Component</th>
<th>Stream Number</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>10</td>
</tr>
<tr>
<td>Acetone</td>
<td>6.1</td>
</tr>
<tr>
<td>Water</td>
<td>0</td>
</tr>
<tr>
<td>Acetic Acid</td>
<td>10</td>
</tr>
<tr>
<td>MeI</td>
<td>26</td>
</tr>
<tr>
<td>MeOAc</td>
<td>58</td>
</tr>
</tbody>
</table>

Operation of the described acetone removal process resulted in a decrease in the amount of acetone present in the carbonylation reactor, e.g., acetone concentrations of about 2.0 to 3.0 weight percent based on the total weight of the reactor contents.

Although the present invention has been described in terms of the presently preferred embodiment, it is to be understood that such disclosure is not to be interpreted as limiting to the invention described herein. No doubt that after reading the disclosure, various alterations and modifications will become apparent to those skilled in the art to which the invention pertains. It is intended that the appended claims be interpreted as covering all such alterations and modifications as fall within the spirit and scope of the invention.
CLAIMS

I claim:

1. A process for the removal of acetone from a mixture that includes methyl acetate, methyl iodide and acetone, the process comprising the steps:

   (1) introducing a mixture which includes acetone, methyl acetate and methyl iodide into a distillation zone;
   (2) introducing water into said distillation zone at substantially the same point as, or at one or more points above, the location where the mixture stream is introduced into the distillation zone; and
   (3) removing from the distillation column:
       (a) an overhead vapor stream comprising methyl acetate and essentially all of the methyl iodide fed to the distillation column;
       (b) a side stream comprising acetone, methyl acetate and water; and
       (c) a bottoms product consisting substantially of water.

2. The process of claim 1 wherein said mixture contains less than about 1 weight % acetic acid.

3. A process for the removal of acetone from a mixture that includes methyl acetate, methyl iodide and acetone, the process comprising the steps:

   (1) introducing a mixture which includes acetone, methyl acetate and methyl iodide into a first distillation zone;
   (2) withdrawing from said first distillation zone a side stream having acetone, methyl acetate and methyl iodide;
   (3) introducing said first distillation side stream into a second distillation zone;
   (4) introducing water into said second distillation zone at substantially the same point as, or at one or more points above, the location where said first distillation side stream is introduced into the second distillation zone; and
   (5) removing from said second distillation zone:
       (a) an overhead vapor stream comprising methyl acetate and essentially all of the methyl iodide fed to the distillation column; and
(b) at one or more points below the location where said first distillation side stream is introduced into the second distillation zone an underflow or bottom product comprising acetone, methyl acetate and water.

4. A process for the removal of acetone from a mixture that includes methyl acetate, methyl iodide, acetic acid, and acetone, the process comprising the steps:

   (1) distilling said mixture in a first distillation zone to obtain:
   (a) an overhead stream comprising methyl acetate, methyl iodide and acetone;
   (b) a side stream comprising methyl acetate, methyl iodide and acetone; and
   (c) a bottom stream comprising methyl acetate, methyl iodide, acetone and essentially all of the acetic acid;

   (2) contacting said overhead stream with water to obtain:
   (a) a methyl iodide phase containing methyl acetate; and
   (b) an aqueous phase containing methyl acetate, methyl iodide and acetone;

   (3) feeding the streams from steps (1)(b) and (2)(b) to a second distillation zone;

   (4) removing from said second distillation zone:
   (a) an overhead vapor stream comprising methyl acetate and essentially all of the methyl iodide fed to said second distillation zone; and
   (b) a bottoms stream comprising water and acetone.

5. The process of claim 4 wherein said streams (1)(b) and (2)(b) are combined prior to feeding to said second distillation zone.

6. The process of claim 4 wherein said stream (2)(b) is fed to said second distillation zone at one or more points above the feed point of stream (1)(b) into said second distillation zone.

7. The process of claim 4 wherein said bottoms stream from said second distillation zone contains less than about 200 ppm iodine.

8. The process of claim 4 wherein said bottoms stream from said second distillation zone contains less than about 100 ppm iodine.
9. A process for the removal of acetone from a mixture that includes methyl acetate, methyl iodide, acetic acid, and acetone, the process comprising the steps:

(1) distilling said mixture in a first distillation zone to obtain:
   (a) an overhead stream comprising methyl acetate, methyl iodide and acetone;
   (b) a side stream comprising methyl acetate, methyl iodide and acetone; and
   (c) a bottom stream comprising methyl acetate, methyl iodide, acetone and essentially all of the acetic acid;

(2) contacting said overhead stream with water to obtain:
   (a) a methyl iodide phase containing methyl acetate; and
   (b) an aqueous phase containing methyl acetate, methyl iodide and acetone;

(3) feeding the streams from steps (1)(b) and (2)(b) to a second distillation zone;

and

(4) removing from said second distillation zone:
   (a) a second overhead vapor stream comprising methyl acetate and essentially all of the methyl iodide fed to said second distillation zone;
   (b) a second side stream from said second distillation zone, said second side stream comprising methyl acetate, water and from about 75-95 weight % of the acetone fed to the second distillation column; and
   (c) a second bottoms stream comprising water and acetone.

10. The process of claim 9 wherein said second side stream from said second distillation zone comprises from about 80-95 weight % of the acetone fed to the second distillation column.

11. The process of claim 9 wherein said second bottoms stream comprises greater than about 95 weight % water.

12. The process of claim 9 wherein said second bottoms stream comprises greater than about 98 weight % water.

13. The process of claim 9 further comprising recycling a portion of said second bottoms stream to said second distillation zone to a point above the feed of the step (2)(b) aqueous phase.
14. The process of claim 9 wherein up to about 60 weight percent of said recycled second bottoms stream is used for contacting said overhead stream of step (1)(a) with water.
**INTERNATIONAL SEARCH REPORT**

**A. CLASSIFICATION OF SUBJECT MATTER**

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According to international Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

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Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

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☐ Further documents are listed in the continuation of box C.  ☒ Patent family members are listed in annex.

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**Date of the actual completion of the international search**

18 April 2001

**Date of mailing of the international search report**

02/05/2001

**Name and mailing address of the ISA**

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Fax: (+31-70) 340-3016

**Authorized officer**

Bonnevalle, E
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