

Appeal decision

Appeal No. 2018-13965

Tokyo, Japan

Appellant Mitsubishi Chemical Corporation

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The case of appeal against the examiner's decision of refusal of Japanese Patent Application No. 2014-219226, entitled "Method for Distilling Easily Polymerizable Compound under Reduced Pressure, and Method for Producing Acrylic Acid" [the application published on June 18, 2015, Japanese Unexamined Patent Application Publication No. 2015-110551] has resulted in the following appeal decision.

Conclusion

The appeal of the case was groundless.

Reasons

No. 1 History of the procedures

The Application was filed on October 28, 2014 (priority claim October 29, 2013), a written amendment was submitted on June 1, 2017, a notification of reasons for refusal was issued on December 5, 2017, a written opinion and a written amendment were submitted on April 10, 2018, a decision of refusal was made on July 20, 2018, and a demand for an appeal against the examiner's decision of refusal was filed on October 22, 2018.

With respect to the Application, information offer forms dated July 7, 2017, October 2, 2017, and April 23, 2018 have been submitted.

No. 2 The Invention

The invention disclosed in Claim 1 of the Application is as disclosed in Claim 1 of Scope of Claims amended by amendment of April 10, 2018.

"[Claim 1]

A method for manufacturing an acrylic acid, which comprises a step of executing vacuum distillation, using a steam ejector, on an acrylic acid resulting from gas-phase catalytic oxidation using propane, propylene, or acrolein as a material, wherein the vacuum distillation step includes a step of heating an outer surface of the steam ejector and the pressure of the drive vapor of the steam ejector is 0.5 to 2 MPaG" (hereinafter, referred to as the "Invention").

No. 3 Reasons for refusal stated in the examiner's decision

Reasons for refusal stated in the examiner's decision are recognized as follows:

The Invention according to Claim 1 of the Application could have been easily conceived by a person ordinarily skilled in the art based on the inventions disclosed in Publications 1 to 5 below distributed in Japan or a foreign country prior to the priority date, thus, the appellant should not be granted a patent for the Invention in accordance with the provisions of Article 29(2) of the Patent Act.

1 Japanese Unexamined Patent Application Publication No. 2001-13116

2 Japanese Unexamined Patent Application Publication No. 2000-344688

3 Japanese Unexamined Patent Application Publication No. 2012-7207

4 Japanese Unexamined Patent Application Publication No. 2003-73327

5 Japanese Unexamined Patent Application Publication No. 10-204030

Publications 2 to 5 indicate the common general technical knowledge as of the priority date of the case.

No. 4 Judgment by the body

The body judges that, as indicated in the reasons for refusal stated in the examiner's decision, the Invention could have been easily conceived by a person skilled in the art who knew the common general technical knowledge as of the priority date of the Application based on to the inventions disclosed in Publications 1 to 11 below distributed in Japan or a foreign country prior to the priority date, and thus, the appellant should not be granted a patent for the Invention in accordance with the provisions of Article 29(2) of the Patent Act.

The reasons are as follows:

- 1 Japanese Unexamined Patent Application Publication No. 2001-13116
- 2 Japanese Unexamined Patent Application Publication No. 2000-344688
- 3 Japanese Unexamined Patent Application Publication No. 2012-7207
- 4 Japanese Unexamined Patent Application Publication No. 2003-73327
- 5 Japanese Unexamined Patent Application Publication No. 10-204030
- 6 Japanese Unexamined Patent Application Publication No. 2009-249314
- 7 Japanese Unexamined Patent Application Publication No. 2004-261683
- 8 Japanese Unexamined Patent Application Publication No. 2003-103105
- 9 Japanese Unexamined Patent Application Publication No. 2009-149587
- 10 Japanese Unexamined Patent Application Publication No. 2003-103102
- 11 Japanese Unexamined Patent Application Publication No. 48-32513

Publications 2 to 11 indicate the common general technical knowledge as of the priority date of the case.

1. Descriptions in the cited publications

(1) Publication 1: Japanese Unexamined Patent Application Publication No. 2001-13116

Publication 1 cited in the examiner's decision that is a publication distributed before the priority date of the Application includes the following statements.

(1a) "[Claim 1] A method for distilling a liquid comprising an easily polymerizable compound in which the liquid comprising an easily polymerizable compound is distilled using a distillation column equipped with a condenser on the side of the outlet for the liquid, wherein at least one condenser is further provided in series on the side of the outlet of vapor of the condenser, and the easily polymerizable compound contained in the vapor from a condenser on the upstream side is condensed by a condenser on the downstream side.

...

[Claim 14] The method for distilling a liquid comprising an easily polymerizable compound disclosed any one of Claims 1 to 13, wherein the easily polymerizable compound is (meth)acrylic acid and/or (meth)acrylic acid ester" ([Scope of Claims]).

(1b) "[0005] In the conventional methods for distilling represented by those methods, however, since vapor coming out from the condenser contains easily polymerizable compounds such as (meth)acrylic acid and/or (meth)acrylic acid ester, no stable operation can be carried out because of frequently repeated unavoidable shut-downs of the device

due to generation of compounds in the line from the condenser to the ejector or the vacuum pump, in the line up to the pump, in the line from the condenser to the atmosphere or the industrial pretreatment facility, at the outlet section of the ejector, and in the vacuum pump.

[0006]

[Problem to be solved by the Invention] Accordingly, the problem to be solved by the invention is to provide a method that can prevent polymerization inside the distillation device in distilling liquids comprising easily polymerizable compounds such as (meth)acrylic acid and (meth)acrylic acid ester."

(1c) "[0007]

[Means for solving the problem] The inventors carried out intensive studies for solving the above problem to be solved by the invention. As a result, the inventors found out that the above problem to be solved by the invention can be solved by providing multiple condensers in series at the outlet of the distillation column and supplying vapor at the outlet of the condenser on the upstream side to the condenser on the downstream side. Thus, the Invention has been completed as described above.

[0008] Namely, the method for distilling a liquid comprising an easily polymerizable compound of the Invention is characterized in that, in a method for distilling a liquid comprising an easily polymerizable compound using a distillation column equipped with a condenser on the outlet side, at least one condenser is further provided in series on the side of the outlet of vapor of the condenser, and the easily polymerizable compound contained in the vapor from a condenser on the upstream side is condensed by a condenser on the downstream side."

(1d) "[0036] In the vapor line at the inlet of the condenser, a method for preventing condensation of vapor that causes polymerization by heating with a tracing or jacket piping, a method for preventing polymerization by maintaining the liquid film of the condensate through sufficient cooling by the trading or jacket piping, or a method to prevent polymerization by showering a liquid comprising stabilizer may be used for preventing polymerization. In this case, the liquid comprising stabilizer is recovered together with the condensate from the condenser."

(1e) "[0039]

[Examples] The Invention is concretely explained below, but the Invention is not limited to those examples.

[Example 1] As shown in Fig. 3, using a distillation device consisting of a distillation column, a reboiler, a first upright multitubular condenser, a second upright multitubular condenser, and a vapor ejector, refinement was continuously carried out by supplying a liquid comprising acrylic acid to the distillation column, inputting oxygen-containing gas for preventing polymerization to the bottom part of the distillation column, and operating the equipment with the column top pressure of 47 hPa, and acrylic acid was distilled from the column top. The temperature during the operation was 95°C at the bottom of the distillation column. A polymerization inhibitor was added to the condensed liquid and the condensed liquid to which a polymerization inhibitor was added was showered to the upper part of the side of pipes of the first condenser and the second condenser and circulated. The heat transmission area of the second condenser was 45% that of the first condenser, and vapor was caused to flow to the second condenser as an upward flow. The inlet temperature of cooling water for the second condenser was 23°C, and the outlet temperature was 25°C. The liquid at the outlet for cooling water of the second condenser and cooling water were mixed with each other and supplied to the inlet for cooling water of the first condenser. The inlet temperature of cooling water for the first condenser was 24°C, and the outlet temperature was 36°C. As a result, the equipment could be operated without problem for 60 days. No adhesion of polymers was detected in the piping of the first condenser and the second condenser by inspection after stopping the equipment, and only slight adhesion of polymers was detected in the piping that connects the second condenser and the ejector."

(2) Publication 2: Japanese Unexamined Patent Application Publication No. 2000-344688

Publication 2 cited in the examiner's decision that is a publication distributed before the priority date of the Application includes the following statements.

(2a) "[Scope of Claims]

[Claim 1] A method for refining easily polymerizable compounds characterized in that, in distilling a liquid comprising an easily polymerizable compound, a refrigeration condenser for tower top vapor from a distillation column is installed so that its condensation plane is kept in a vertical position, and a polymerization inhibitor exists on the condensation plane.

[Claim 2] The method for refining easily polymerizable compounds of Claim 1, wherein a liquid comprising a polymerization inhibitor is sprayed to the upper part of the condensation plane of the refrigeration condenser for vapor.

[Claim 3] The method for refining easily polymerizable compounds of either one of Claims 1 or 2, characterized in that the refrigeration condenser for vapor is installed proximally to the top of the distillation column.

[Claim 4] The method for refining easily polymerizable compounds of Claim 3, characterized in that the temperature of the piping from the top of the distillation column to the inlet of the refrigeration condenser for vapor is maintained higher than the condensing temperature of the vapor.

[Claim 5] The method for refining easily polymerizable compounds of any one of Claims 1 to 4, wherein the easily polymerizable compound is at least a compound selected from a group consisting of acrylic acid, methacrylic acid, acrylic acid ester, and methacrylic acid ester.

[Claim 6] The method for refining easily polymerizable compounds of any one of Claims 1 to 5 wherein the polymerization inhibitor is at least a compound selected from a group consisting of p-methoxyphenol, hydroquinone, phenothiazine, and copper compounds.

[Detailed Description of the Invention]

[0001]

[Field of the Invention] The invention relates to a method for refining easily polymerizable compounds that have a polymerizable double bond in a molecule such as acrylic acid and methacrylic acid (hereinafter, collectively referred to as '(meth)acrylic acid'), (meth)acrylic acid ester and styrene, to be more exact, a method for refining for efficiently and safely carrying out distillation of easily polymerizable compounds."

(2b) "[0002]

[Conventional Art] Generally, compounds that have a double bond, particularly a carbon-carbon double bond in a molecule, are highly reactive and tend to polymerize. If polymerization occurs in a process for handling such compounds, particularly in a step for refining, it may cause operational troubles in equipment such as reduced yield, adherence of polymers, and clogging of piping, etc. caused by adherence of polymers. Accordingly, in refining such easily polymerizable compounds by distillation, a method of distillation by adding various metals or their compounds such as phenothiazine, p-methoxyphenol (commonly called, 'methoxyhydroquinone,' or 'methoquinone'), hydroquinone, and copper compounds, or oxygen or oxygen-containing gas, etc. polymerization inhibitor for preventing polymerization, and, in addition, a method to distill at a low processing temperature under high vacuum to suppress polymerization have widely been used.

[0003] However, generally, since such polymerization inhibitors have a low vapor pressure and hardly exist in vapor in the top of the distillation column, sometimes continuous operation becomes difficult because of polymerization of easily polymerizable compounds in a heat exchanger that carries out cooling and condensation of vapor distilled from the column top or piping from the top of the distillation column to the heat exchanger.

[0004]

[Problem to be solved by the Invention] The problem to be solved by the invention is to provide a method for effectively and safely carrying out refining by preventing polymerization in this cooling and condensing section in refining by distillation of an easily polymerizable compound."

(2c) "[0024] An aqueous solution of acrylic acid obtained by condensing a gas generated by gas phase catalytic oxidation of propylene dehydrated using toluene was supplied to a distillation column as crude acrylic acid at the rate of 8.0 kg per hour. The crude acrylic acid contained water 0.3% by weight water, 2.3% by weight of acetic acid, and 7.0% by weight of toluene. Through distillation, a liquid comprising 0.1% by weight of acetic acid, 98.2% by weight of acrylic acid, and a heavy component was obtained from the bottom of the column at 4.9 kg/hour, and acrylic acid comprising 0.7% by weight of water, 5.4% by weight of acetic acid, and 17.0% by weight of toluene was obtained from the top of the column at 3.3 kg/hour. After continuous operation of this distillation process for one week, the operation was stopped, and the heat exchanger and the piping connected to it were inspected and no polymers were detected in the piping and tubes for heat exchange."

(3) Publication 3: Japanese Unexamined Patent Application Publication No. 2012-7207

Publication 3 cited in the examiner's decision that is a publication distributed before the priority date of the Application includes the following statements.

(3a) "[Claim 4]

The device for treating impurities and exhaust-gas for vacuum degassing in secondary smelting for steel manufacture of any one of Claims 1 to 3, wherein the ejector on the fore stage side is provided with a heating means for drying its inner surface for almost the entirety of its outer circumference.

[Claim 5]

The device for treating impurities and exhaust-gas for vacuum degassing in secondary smelting for steel manufacture of Claim 4, wherein the heating means has a jacket that encircles the ejector on the fore stage side, and the heating means is constituted by injecting vapor into the space between the ejector on the fore stage side and the jacket."

(3b) "[Background Art]

[0002]

Vacuum degassing furnaces are used in steel manufacturing devices as secondary smelting devices, and gaseous impurities in liquid steel are removed using vacuum degassing furnaces, and decarburization for making steel low-carbon is also often carried out using vacuum degassing furnaces. Generally, exhaust gas from vacuum degassing furnaces is exhausted by a processing device provided with an ejector on the fore stage side that functions as a booster and a condenser (condensing appliance) provided in the downstream of the ejector, but, there has been a problem that a large amount of powder dusts is generated, particularly in a decarburizing step, and the powder dusts adhere to and stagnate on the inner surface of the ejector and the condenser on the aft stage and enormous time and labor are required to remove them."

(3c) "[0013]

The device of the invention of Claim 4 is a device of any one of Claims 1 to 3 in which the ejector on the fore stage side is provided with a heating means for drying its inner surface for almost the entirety of its outer circumference. In addition, the invention of Claim 5 is the invention of Claim 4 further embodied, and in this invention, the heating means has a jacket that encircles the ejector on the fore stage side, and the heating means is constituted by injecting vapor into the space between the ejector on the fore stage side and the jacket."

(3d) "[0024]

Since wetting of the inner surface with water can be prevented or remarkably suppressed if the heating means is provided in the ejector on the fore stage side as in Claim 4, adhesion of powder dusts to the inner surface of the ejector on the fore stage side can be prevented or remarkably suppressed. Therefore, according to the invention of Claim 4, powder dusts generated in the degassing step can be effectively collected with the first condenser without allowing them to adhere in the ejector on the fore stage side, and, as a result, adherence of powder dusts caused by degassing or by decarburization can be prevented or remarkably suppressed.

[0025]

As a heating means, it is possible, for example, to wind an electric heater or a vapor heater to the outer surface of the ejector on the fore stage side, but, if a method to inject vapor between the ejector on the fore stage side and the jacket as in Claim 5 is used, since the ejector as a whole can be reliably heated with a simple structure, it can achieve a high heating effect and is preferable. In addition, since there are sufficient vapors in ironworks, it is beneficial as well from a cost perspective."

(4) Publication 4: Japanese Unexamined Patent Application Publication No. 2003-73327

Publication 4 cited in the examiner's decision that is a publication distributed before the priority date of the Application includes the following statements.

(4a) "[0062] First, vapor (exhaust) after using superheated vapor in the vapor turbine 121 as a source of kinetic energy is supplied from the steam line 179 through the steam line 181 to the piping 164 as vapor for gas-phase catalytic oxidation reaction and, after being mixed with raw material gas, is supplied to the first stage reactor 101 that is a vapor consuming device and used. This is because vapor is necessary for the reaction per se, and, for example, in a method for manufacturing acrylic acid, raw material gases such as propylene are oxidized by contact oxidation in gaseous phase in the presence of oxygen-containing gas and vapor under high temperature, and transformed into acrolein in the first stage oxidation reaction and, then, acrolein is transformed to acrylic acid in the second stage oxidation reaction."

(4b) "[0068] Furthermore, in distilling and refining organic acids and organic acid esters, heaters are provided for heating raw materials for a distillation column and stripping tower such as a solvent separating tower, light-boiling separator, and heavy-boiling separator, and vapor can be used as a heat medium for the heaters. Heaters used here may include heat exchangers such as temperature raising devices and evaporators."

(4c) "[0072] Furthermore, if it is used for driving a vacuum generating device, the pressure in the steam line is normally controlled to 0.3 to 2 MPa gauge, preferably to 0.5 to 1 MPa gauge."

(4d) "[0113] In addition, the manufacturing method of the invention is applicable to all organic acids that can be manufactured by gas-phase catalytic oxidation. For example, (meth)acrylic acid, phthalic acid, and maleic acid can be pointed out as such organic acids,

but preferred organic acid is (meth)acrylic acid. In addition, the manufacturing methods of the invention may include a step to manufacturing organic acid ester by further esterifying organic acids. For example, (meth)acrylic acid esters such as methyl (meth) acrylate, ethyl (meth)acrylate, n-butyl (meth)acrylate, hydroxypropyl (meth)acrylate, and 2 ethylhexyl (meth)acrylate can be pointed out as such organic acid esters."

(4e) "[0117] Example 2

Acrylic acid was manufactured with the configuration in Fig. 2. Raw materials, propylene, and oxygen-containing gas were fed to the reactor 201, and a mixture of potassium nitrate and sodium nitrite at 50% by weight each was used as a heat medium for the reactor. The reactor 201 was divided with closure plates and each heat medium was mixed and fed to the heat medium boiler 213 and, after generating high pressure vapor, circulated to the reactor. ... Acrylic acid was obtained by distilling and refining the collected liquid at the bottom of the collection tower in an acid purifying process. In the acid purifying process, vapor was used in a natural circulation reboiler, a heater, and a vacuum generating device (steam ejector). Pressures of used vapor were 0.6 MPa gauge (medium pressure vapor) and 0.2 MPa gauge (low pressure vapor), and the vapor consumption was 8.6 T/h."

(5) Publication 5: Japanese Unexamined Patent Application Publication No. 10-204030

Publication 5 cited in the examiner's decision that is a publication distributed before the priority date of the Application includes the following statements.

(5a) "[Scope of Claims]

[Claim 1] A method for refining (meth)acrylic acid ester characterized in that pressure is reduced using a steam ejector or a water ejector in distilling a mixture comprising (meth)acrylic acid ester under a reduce pressures."

(5b) "[0010] The "steam ejector" is a device that jets a high pressure drive vapor from the nozzle and, by ejecting secondary vapor by suction, and conveying force of the high pressure jet flow, pressures of objects (distillation still, etc.) connected to it are reduced."

(5c) "[0024] The vacuum generating device 5 is a steam ejector of 2-stage booster, 4-stage steam type. Namely, this device 5 is provided with 4 ejectors (E₁ to E₄), and two ejectors E₁ and E₂ in the front row are booster ejectors. Intermediate condensers C₁ and

C₂ are arranged respectively between the second ejector E₂ from the front and the third ejector E₃, and between the third ejector E₃ and the fourth ejector E₄. Those ejectors E₁ to E₄ have constructions as shown in Fig. 2, and suction the drive vapor S₁ (normally, vapor with the pressure of 1.5 to 15 kg/cm²) from the drive vapor inlet 81 and jets from the nozzle 82."

(6) Publication 6: Japanese Unexamined Patent Application Publication No. 2009-249314

Publication 6 that is a publication distributed before the priority date of the Application includes the following statements.

(6a) "[0003]

Normally, decompression in a reactor and introduction of gas in the reactor to the absorption tower are carried out by the ejector.

The ejector is provided with a suction chamber, a nozzle inserted into the suction chamber, and a diffuser in which a fluid passageway connects with the suction chamber coaxially with the direction of axis of the nozzle and runs through in the direction of axis. In the ejector, as the drive fluid (vapor, etc.) jetted from the nozzle passes through the fluid passageway of the diffuser, the inside of the diffuser has negative pressure and gas is suctioned by the negative pressure from the suction port to the inside of the suction chamber and joins together with the drive fluid in the diffuser and sent out from the diffuser.

[0004]

For the purpose of easily carrying out maintenance work, the ejector is installed to have the axis line in a horizontal position. In a horizontally installed ejector, however, raw materials, (meth)acrylic acid and ethylene oxide, and the target product hydroxyalkyl (meth)acrylate ester contained in the gas sucked into the suction chamber from the suction port tend to stagnate together with the condensed drive fluid (water, etc.) in the suction chamber and/or the diffuser. In addition, since the suction raw materials, (meth)acrylic acid as well as ethylene oxide and the target product, (meth)acrylic acid hydroxyalkyl ester stagnated in the suction chamber, etc. tend to polymerize, polymers tend to adhere to the inside of the suction chamber. Therefore, it is necessary to frequently stop manufacture of (meth)acrylic acid hydroxyalkyl ester and carry out maintenance (cleaning, etc.) of the ejector."

(6b) "[0010]

As shown in Fig. 2, the ejector 20 is provided with a suction chamber 62 that has a gas suction port 60, a nozzle 64 inserted into the suction chamber 62, a diffuser 68 in which a fluid passageway 66 connects with the suction chamber 62 coaxially with the direction of axis of the nozzle 64 and runs through in the direction of axis, and a heating means 70 provided in the circumference of the suction chamber 62. The ejector 20 is installed so that the axis line of the nozzle 64 and the axis line of the diffuser 68 are placed perpendicular to the horizontal plane, and the nozzle 64 is placed in a position higher than the diffuser 68.

The fluid passageway 66 has a small diameter part 72 that has the smallest inner diameter, a reduced diameter part 74 formed on the upstream side to the small diameter part 72 whose diameter decreases toward the downstream, and an expanded part 76 formed on the downstream side to the small diameter part 72 whose diameter increases toward the downstream.

The heating means 70 may be a steam line, an electric heater, hot-water piping, heating medium piping, etc."

(6c) "[0023]

In the device for manufacturing hydroxyalkyl ester 10 described above, since the ejector 20 is provided with a heating means for heating the suction chamber 62, the drive fluid (vapor, etc.) hardly stagnates in the suction chamber 62. Therefore, raw materials, (meth)acrylic acid and ethylene oxide, and the target product, hydroxyalkyl (meth)acrylate ester contained in the gas suctioned from the suction port 60 into the suction chamber 62 together with stagnated drive fluid (water, etc.) hardly stagnate in the suction chamber 62 and/or the diffuser 68. Accordingly, the suction chamber polymers hardly adhere to the inside of the suction chamber 62, etc."

(7) Publication 7: Japanese Unexamined Patent Application Publication No. 2004-261683

Publication 7 that is a publication distributed before the priority date of the Application includes the following statements.

(7a) "[Scope of Claims]

[Claim 1]

A passing device for easily polymerizable compounds that is provided with a cylinder that constitutes a part of the pathway in which vapor phase comprising an easily polymerizable compound passes, wherein the cylinder is provided with a jacket that

encircles the outer circumference of the cylinder and reduced water vapor is passed through the jacket.

[Claim 2]

The passing device for easily polymerizable compounds of Claim 1, wherein the cylinder is installed in a portion in the pathway in which the temperature distribution of the vapor phase is within the range of 2°C.

[Claim 3]

The passing device for easily polymerizable compounds of either one of Claim 1 or 2, wherein the temperature of the reduced-pressure water vapor is set to a temperature 0.1 to 15°C higher than the temperature of the vapor phase passing through the cylinder.

[Claim 4]

The passing device for easily polymerizable compounds of any one of Claims 1 or 3, wherein a drainage pipe is provided for draining vapor drain generated in the jacket, and the drainage pipe extends vertically downward from the bottom of the jacket, turns around in a U-shape to rises, and connects to a vent that is normally kept open.

[Claim 5]

A method for distilling easily polymerizable compounds using a vapor delivering line and a condenser, wherein a cylinder constituting at least a part of the section from the void tower portion of the distillation column to the vapor delivering line consists of the cylinder of the passing device for easily polymerizable compounds of any one of Claims 1 to 4.

[Claim 6]

A method for distilling easily polymerizable compounds of Claim 5 that is a step for distilling in a method for manufacturing hydroxyalkyl(meth) acrylate provided with a step to generate and a step to distillate hydroxyalkyl(meth)acrylate, wherein the step to distillate is carried out using a distillation column, a vapor advancing line, and a condenser."

(7b) "[0011]

[Embodiments of the Invention]

Hereinafter, the passing device for easily polymerizable compounds of the invention and its use are explained in detail, but the scope of the invention is not restricted by the explanation, and the invention may be worked outside the following examples by appropriately modifying within the scope not deviating from the import of the invention. Easily polymerizable compounds tend to polymerize if they are evaporated and condensed. Namely, high-temperature vapor of an easily polymerizable compound

tends to polymerize if it is cooled and condensed by heat release, etc. Therefore, it is necessary to make vapor of an easily polymerizable compound below 100°C pass the pathway not heated to a high temperature but not caused to condensate by heating to a low temperature and to keep it warm as much as possible.

[0012]

Since the whole pathway can be evenly heated and kept at a low temperature by using the passing device for easily polymerizable compounds of the invention, vapor phase comprising an easily polymerizable compound can be passed without condensing or polymerizing in the pathway. Since the temperature of reduced pressure water vapor can be easily adjusted to a temperature below 100°C, vapor of the easily polymerizable compound with a comparatively low temperature of, for example, around 70 to around 90°C can be passed through the pathway without condensing and polymerizing."

(7c) "[0024]

The passing device for easily polymerizable compounds of the invention can be preferably applied to a method for distilling easily polymerizable compounds by using a distillation column, a vapor delivering line, and a condenser. Namely, the cylinder constituting at least a part of the section from the void tower portion of the distillation column to the vapor delivering line is constituted with the passing device for easily polymerizable compounds of the invention.

As easily polymerizable compounds to which such method for distilling can be preferably applied, unsaturated carboxylic acids such as (meth)acrylic acid, unsaturated carboxylic acid esters such as methyl acrylate, ethyl acrylate, methyl methacrylate, butyl acrylate, octyl acrylate, 2-hydroxyethyl (meth)acrylate, 2-hydroxypropyl (meth)acrylate, and butyl methacrylate, acrylic compounds such as acrylonitrile and acrylamide, and polymerizable vinyl compounds such as styrene are pointed out without limiting to them.

[0025]

The distillation method of the invention can be effectively applied preferably to distillation and refining of liquids comprising at least one type of compound selected from (meth)acrylic acids and (meth)acrylic acid esters, and more preferably to the method for distilling hydroxyalkyl(meth) acrylate.

Due to such reasons, the distillation method of the invention can be preferably applied to, for example, a method for manufacturing hydroxyalkyl (meth) acrylate. Namely, this manufacturing method includes a step to generate and a step to distill hydroxyalkyl (meth)acrylate, and the step of distilling is carried out using a distillation column, a vapor delivering line and condenser, and, in the step of distilling, the cylinder

constituting at least a part of the portion from the void tower portion of the distillation column to the vapor delivering line is constituted with the passing device for easily polymerizable compounds.

[0026]

Describing an example of the method for manufacturing hydroxyalkyl (meth)acrylate in more detail, the method consists of a step to generate hydroxyalkyl (meth)acrylate by esterification of (meth)acrylic acid and alkylene oxide in the presence of a catalyzer, and a step for distilling hydroxyalkyl (meth) acrylate from the reaction solution after the reaction in which unreacted alkylene oxide and/or unreacted (meth) acrylic acid remain.

Using the passing device for easily polymerizable compounds, since vapor phase containing vapor of hydroxyalkyl (meth)acrylate is not heated to any high temperature and is maintained at a low temperature to prevent condensation as much as possible, occurrence of polymerization in the inlet section of the condenser can be suppressed. Therefore, the method for distilling hydroxyalkyl (meth)acrylate of the invention, it becomes easier to use an efficient device such as an upright multitubular heat exchanger. In this case, it is recommendable to prevent polymerization by circulating distillate and showering it in the vapor as in the case of a barometric condenser."

(8) Publication 8: Japanese Unexamined Patent Application Publication No. 2003-103105

Publication 8 that is a publication distributed before the priority date of the Application includes the following statements.

(8a) "[Scope of Claims]

[Claim 1] A method for distilling an easily polymerizable compound in which the easily polymerizable compound is distilled in a distillation column, wherein at least a part of the column top and/or the pipe are heated when discharging vapor containing easily polymerizable compound from the column top through the pipe.

[Claim 2] The method for distilling an easily polymerizable compound of Claim 1, wherein the column top and/or the pipe is heated to a temperature higher than the temperature inside the column.

[Claim 3] The method for distilling an easily polymerizable compound of Claim 1, wherein the column top and/or the pipe is heated to a temperature higher than the temperature inside the column but not exceeding the temperature inside the column for over 100°C.

[Claim 4] The method for distilling an easily polymerizable compound of any one of Claims 1 to 3, wherein the easily polymerizable compound is a vinyl compound.

[Claim 5] A distillation device used for the method for distilling an easily polymerizable compound of any one of Claims 1 to 4, wherein the column top is provided with a pipe for discharging the easily polymerizable compound, and a heating means is provided at least at a part of the column top and/or the pipe.

[Claim 6] The method for distilling an easily polymerizable compound of Claim 5, wherein the heating means is a heating jacket for passing a heat medium provided on the surface of the column top and/or the pipe.

[Claim 7] The method for distilling an easily polymerizable compound of Claim 5, wherein the heating means is a tracing piping for passing a heat medium provided to contact the surface of the column top and/or the pipe."

(8b) "[0007]

[Problem to be solved by the Invention] The main purpose of the invention is to provide a method for distilling that reduces generation of compounds and enables continuous operation in distilling easily polymerizable compounds represented by vinyl compounds. Another objective of the Invention is to provide a distillation device that reduces generation of polymers and enables continuous operation.

[0008]

[Means for solving the problem] The inventors carried out intensive studies for finding a solution for the above problem to be solved by the invention, and, as a result, found out that the positions in the distillation column where the easily polymerizable compound tends to polymerize is a site in which vapor comprising the easily polymerizable compound gets condensed and tends to form liquid accumulation in a state in which no polymerization inhibitor exists. In addition, the inventors also found out that formation of areas in which easily polymerizable compounds tend to polymerize in the distillation column can be prevented by making vapor comprising the easily polymerizable compound not easily get condensed at sites in the column top and/or the pipe in which the vapor hardly contacts a liquid comprising polymerization inhibitor through heating at least a part of the column top and/or the pipe, and thereby completed the invention."

(8c) "[0031]

[Advantage of the Invention] According to the invention, since the column top and the pipe can be maintained at a temperature higher than the temperature inside the column by providing heating means such as heating jackets and tracing piping in the column top and

the pipe, condensation of the easily polymerizable compound in such sites can be prevented, and, thanks to this, the invention has such advantageous effects that generation of polymers is reduced and continuous operation of the distillation column is possible."

(9) Publication 9: Japanese Unexamined Patent Application Publication No. 2009-149587

Publication 9 that is a publication distributed before the priority date of the Application includes the following statements.

(9a) "[Scope of Claims]

[Claim 1]

An easily polymerizable compound handling device provided with a main unit, a nozzle projecting from the main unit, a cover sealing the opening of the nozzle, a metallic thin film provided to block between the base end and the tip of the nozzle and formed with a vent, and a gas supply means for supplying displacing gas between the cover and the metallic thin film.

[Detailed Description of the Invention]

[Technical field]

[0001]

The invention relates to a device for handling easily polymerizable compounds.

[Background Art]

[0002]

In devices (reactors, distillation columns, etc.) for handling easily polymerizable compounds ((meth)acrolein, (meth)acrylic acid, etc.), polymerization of easily polymerizable compounds tends to occur. Particularly, easily polymerizable compounds tend to stagnate on and polymers tend to adhere to the inner wall of the nozzle protruding from the main body of the device and the surface of the cover that closes the opening of the nozzle contacting the easily polymerizable compound. Therefore, it is necessary to frequently stop operation of the device and carry out maintenance (cleaning, etc.) of the device.

[0003]

The following device is proposed as a device in which stagnation of the easily polymerizable compound in the nozzle and clogging of the nozzle with polymers are suppressed.

An oxidation reactor provided with a main body of the reactor and a nozzle protruding from the reactor, wherein the reactor is further provided with a means to feed inert gas to the nozzle and a means to heat the nozzle (Patent Document 1).

[0004]

In the oxidation reactor, however, since replacement of atmosphere in the nozzle by inert gas is not sufficient, there is a problem that the easily polymerizable compound remains in the nozzle and the easily polymerizable compound tends to stagnate and polymers tend to adhere to the inner wall of the nozzle protruding from the main body of the device and the surface of the cover that closes the opening of the nozzle contacting the easily polymerizable compound. Therefore, it is necessary to frequently stop operation of the oxidation reactor and carry out maintenance (cleaning, etc.) of the oxidation reactor."

(9b) "[0017]

Hereinafter, as an example of the device for handling easily polymerizable compounds of the invention, a multitubular reactor is described.

Fig. 1 is a section that shows a portion around a nozzle for mounting a rupture disc in a multitubular reactor. The multitubular reactor 10 (device for handling easily polymerizable compounds) is provided with a main body of the reactor 12 (main body of the device), a nozzle 14 protruding from the upper channel 62 of the main body of the reactor 12, a rupture disc section 16 that closes the opening of the nozzle 14, a discharge pipe 22 connected to the nozzle 14 through the rupture disc section 16 using a bolt 18 and a nut 20, and a nozzle heating means 24 provided in the circumference of the nozzle 14.

[0018]

The nozzle 14 has a double pipe structure consisting of an outer pipe 26 (main body of the nozzle) whose base end communicates with the inside of the main body of the reactor 12, and an inner pipe 28 inserted into the outer pipe 26.

The inner pipe 28 is arranged so that a flange 30 formed on the tip (on the rupture disc section 16 side) is engaged with the tip of the outer pipe 26 and the base end (on the main body of the reactor 12 side) is arranged to locate in the midway of the outer pipe 26 and near the base end of the outer pipe 26.

A void 32 is formed between the outer pipe 26 and the inner pipe 28, and the void 32 is sealed with a sealing member 34 at the base ends of the outer pipe 26 and the inner pipe 28.

[0019]

An L-shaped first through-hole 36 whose one end is connected to the compressor (not illustrated) (gas source) provided outside the nozzle 14 and whose other end communicates with the void 32 is formed on a flange 30 of the inner pipe 28.

Also, a second through-hole 38 whose one end communicates with the inside of the nozzle 14 (inner pipe 28) and whose other end communicates with the void 32 is formed on a peripheral wall of the inner pipe 28.

The first through-hole 36, the void 32 and the second through-hole 38 form a gas supply passage, and a combination of the gas supply passage and the compressor (gas source) constitutes a gas supply means.

[0020]

An aluminum foil 40 is mounted on the base end of the inner pipe 28 in a way to close the opening of the base end. By this, the aluminum foil 40 is provided to block the midway of the nozzle 14.

A ventilation hole 42 is formed in the middle of the aluminum foil 40.

[0021]

The rupture disc section 16 is provided with a rupture disc 46 (lid) that has a dome section 44 that swells toward the nozzle 14, and two holders 48 and 50 that clamp the rim of the rupture disc 46.

In the rupture disc section 16, if the pressure inside the main body of the reactor 12 abnormally increases, the dome section 44 of the rupture disc 46 buckles and reverses, and the dome section 44 collides a cross-shaped knife 52 provided inside the holder 50 on the side opposite to the nozzle side and the rupture disc 46 bursts and opens. This releases the excessive pressure inside the main body of the reactor 12, and the released pressure is discharged to the outside through the discharge pipe 22.

[0022]

Sealings 54 (O-rings) are clamped between the tip of the outer pipe 26 and the flange 30 of the inner pipe 28, between the flange 30 and the rupture disc section 16, and between the rupture disc section 16 and the end of the discharge pipe 22.

[0023]

A steam line, an electric heater, etc. can be pointed out as nozzle heating means 24. By providing the nozzle heating means 24, the nozzle 14 is heated and easily polymerizable compounds adhered to the inner wall of the nozzle 14 vaporize and stagnation of easily polymerizable compounds inside the nozzle 14 is suppressed."

(10) Publication 10: Japanese Unexamined Patent Application Publication No. 2003-103102

Publication 10 that is a publication distributed before the priority date of the Application includes the following statements.

(10a) "[Scope of Claims]

[Claim 1] A method for distilling easily polymerizable compounds, wherein, in distilling easily polymerizable compound by refluxing, reflux liquid is heated.

[Claim 2] The method for distilling easily polymerizable compounds of Claim 1, wherein the reflux liquid is heated to a temperature within the range of $\pm 10^{\circ}\text{C}$.

[Claim 3] The method for distilling easily polymerizable compounds of Claim 1 or Claim 2, wherein the easily polymerizable compound is a vinyl compound.

[Claim 4] A distillation device for using in the method for distilling of any one of Claims 1 to 3, wherein a heating means to heat the reflux liquid is provided at least in a part of the reflux path.

[Claim 5] The distillation device of Claim 4, wherein the heating means is a heat exchanger."

(10b) "[0007]

[Problem to be solved by the Invention] The main purpose of the invention is to provide a distillation method in which generation of polymers is reduced and continuous operation is made possible in distilling easily polymerizable compounds represented by vinyl compounds. Another purpose of the invention is to provide a distillation device that reduces generation of polymers and enables continuous operation."

(10c) "[0024]

As described above, reflux liquid is heated by heat exchangers such as a shell and tube exchanger, tracing piping 3, and a heating jacket 4, and the temperature of reflux liquid is controlled to the above-mentioned range in the invention. By doing so, by virtue of reducing the possibility of formation of liquid agglutination by condensation of vapor comprising easily polymerizable compound in locations such as the neighborhood of the flux pipe and the liquid distribution device (especially, on lower side) in which contact with reflux liquid comprising polymerization inhibitor rarely occurs, polymerization can be suppressed."

(11) Publication 11: Japanese Patent Publication S48-32513

Publication 11 that is a publication distributed before the priority date of the Application includes the following statements.

(11a) "<Detailed Description of the Invention>

The invention relates to a method for distilling polymerizable compounds. In more detail, the Invention relates to a method for distilling polymerizable vinyl compounds, such as vinyl acetate, acrylonitrile, methacrylonitrile, acrylic acid, methacrylic acid, acrylic acid ester, methacrylic acid ester, acrolein, methacrolein, and vinyl pyridine. ... polymerization reaction occurs on the surface of the distillation column where the material has rough surfaces or where the liquid stagnates for a long time, and, if such situation appears once, it spreads at an accelerated pace, and then a part of the distillation column is clogged by such polymers and distillation operation frequently becomes impossible to carry out" (column 1, line 20 to column 2, line 13).

(11b) "In an actual distillation column, if only a column wall is further provided with lagging material for heat retention, there is heat leakage from column liquid to the outside as far as distillation is carried out at a temperature equal to or higher than room temperature. This means that condensation of vapor occurs on the column wall during distillation operation, and such phenomenon is not preferable from the purpose of the invention. Namely, the invention succeeded in totally avoiding polymerization during operation by providing the tower wall with appropriate heating devices and using a perforated plate tower without a weir with which no heat leakage to the outside during operation occurs.

In general, in distilling a substance in a distillation column, since each part of the distillation column has a different temperature from the other parts, it is practically impossible to make heat transfer between parts of the outer wall of the column completely zero. Therefore, from the purpose of the invention, practically, the invention is worked by carrying out operation in a state that the temperature of the outer wall of the column is slightly higher than that of the substance in the column to be distilled. In this case, unless the temperature of the outer wall is made remarkably higher than the temperature inside the distillation column, rectifying effect is hardly sacrificed. On this occasion, it is desirable that the difference in temperatures between the outer wall and the inside of the distillation column is not larger than 30°C.

As a source of heat for the outer wall, an appropriate heat source such as steam, electric heat, warm wind, warm water, and/or heat medium may be used. On the other hand, the column may be heated by segments, or together as a whole." (column 4, lines 5 to 29).

2. Regarding the Invention described in Publication 1

According to the description (1a), among methods for distilling liquids comprising easily polymerizable compounds using a distillation column provided with a condenser on the outlet side, Publication 1 describes a distillation method for distilling liquids comprising easily polymerizable compounds in which at least one condenser is further provided in series on the vapor outlet side of the condenser, and the easily polymerizable compound is (meth)acrylic acid and/or (meth)acrylic acid ester, and the description (1e) states, as a description of a concrete example that corresponds to the description in the scope of claims, that, using a distillation device consisting of a distillation column, a reboiler, a first upright multitubular condenser, a second upright multitubular condenser, and a vapor ejector, acrylic acid-containing liquid was supplied to the distillation column, and the device was operated with the column top pressure of 47 hPa, acrylic acid is distilled from the column top and refinement was carried out continuously, polymerization inhibitor is added to the condensed liquid, and the condensed liquid was showered to the upper part of the sides of the first condenser and the second condenser and circulated.

Then, it can be deemed that Publication 1 describes as an invention according to a concrete example "in methods for distilling liquids comprising easily polymerizable compounds using a distillation column provided with a condenser on the outlet side, a distillation method for distilling liquids comprising easily polymerizable compounds in which at least one condenser is further provided in series on the vapor outlet side of the condenser, and, as a method for distilling acrylic acid-containing liquid in which easily polymerizable compound contained in vapor coming out from the condenser on the upstream side is condensed by the condenser on the downstream side, acrylic acid-containing liquid is supplied to the distillation column using a distillation device consisting of a distillation column, a reboiler, a first upright multitubular condenser, a second upright multitubular condenser, and a vapor ejector, the device is operated with a column top pressure of 47 hPa, acrylic acid is distilled from the column top and refinement is carried out continuously, polymerization inhibitor is added to the condensed liquid, and the condensed liquid is showered to the upper part of the sides of the first condenser and the second condenser and circulated" (hereinafter, the "invention of Publication 1").

3. Comparison/Judgment

(1) Comparison between the Invention and invention of Publication 1

"Acrylic acid is supplied to the distillation column, ... the device is operated with a column top pressure of 47 hPa, acrylic acid is distilled from the column top, and

refinement is carried out continuously," in the invention of Publication 1 corresponds to "a step of executing vacuum distillation" of "acrylic acid" in the Invention, and "vapor ejector" in the invention of Publication 1 corresponds to "steam ejector" in the Invention, and, since the "vapor ejector" of the invention of Publication 1 functions as a vacuum device, "using a distillation device consisting of a distillation column, a reboiler, a first upright multitubular condenser, a second upright multitubular condenser, and a steam ejector, acrylic acid-containing liquid is supplied to the distillation column, and the device is operated with a column top pressure of 47 hPa, acrylic acid is distilled from the column top and refining is carried out continuously" in the invention of Publication 1 corresponds to "method for manufacturing an acrylic acid, which comprises a step of executing vacuum distillation, using a steam ejector," in the Invention.

Then, the Invention and the invention of Publication 1 are identical in that they relate to:

"a method for manufacturing acrylic acid comprising a step of executing vacuum distillation of acrylic acid,"

and different in the following points

Different Feature 1: With respect to the raw material for acrylic acid and synthesis reaction, while the invention specifies them as "resulting from gas-phase catalytic oxidation using propane, propylene, or acrolein as a material," the invention of Publication 1 does not specify such matter.

Different Feature 2: With respect to the pressure of the drive vapor of the steam ejector, while the invention specifies it as "0.5 to 2 MPaG," the invention of Publication 1 does not specify such matter.

Different Feature 3: While the step of executing vacuum distillation is specified by the Invention as "includes a step of heating an outer surface of the steam ejector," the invention of Publication 1 does not specify such matter.

(2) Examination of the different features

The above-mentioned different features are examined below.

A Regarding Different Feature 1

First, Different Feature 1 is examined.

With respect to the raw material and the synthesis reaction for acrylic acid, as stated in the statement in the description (2c) in Publication 2 "An aqueous solution of acrylic acid obtained by condensing a gas generated by gas phase catalytic oxidation of propylene dehydrated using toluene was supplied to a distillation column as crude acrylic acid at the rate of 8.0 kg per hour", and the statement in the description (4a) in Publication 4, "for example, in a method for manufacturing acrylic acids, raw material gases such as propylene are oxidized by contact oxidation in gaseous phase in the presence of oxygen-containing gas and vapor under high temperature, and transformed into acrolein in the first stage oxidation reaction and, then, acrolein is transformed to acrylic acid in the second stage oxidation reaction," it is a well-known technical matter that acrylic acid is obtained by gas-phase catalytic oxidation using propylene or acrolein as a material, with respect to the material for acrylic acid and the synthesis reaction of the invention of Publication 1, it is a technical matter that a person skilled in the art could have easily carried out to specify at least "resulting from gas-phase catalytic oxidation using propane, propylene, or acrolein as a material."

B Regarding Different Feature 2

Next, Different Feature 2 is examined below.

Statement concerning the driving pressure of the steam ejector in the invention of Publication 1 does not exist in Publication 1, but it is an obvious matter that a certain driving pressure is required for causing an ejector function as a vacuum device.

As stated in the statement in description (4c) in Publication 4, "if it is used for driving a vacuum generating device, the pressure in the steam line is normally controlled to 0.3 to 2 MPa gauge, preferably to 0.5 to 1 MPa gauge" and the statement in description (4e), "Acrylic acid was manufactured Raw materials, propylene and oxygen-containing gas are fed to the reactor 201, and a mixture of potassium nitrate and sodium nitrite at 50% by weight each was used as a heat medium for the reactor. The reactor 201 was divided with closure plates and each heat medium was mixed and fed to the heat medium boiler 213 and, after generating high pressure vapor, circulated to the reactor. ... Acrylic acid was obtained by distilling and refining the collected liquid at the bottom of the collection tower in acid purifying process. In the acid purifying process, vapor was used in a natural circulation reboiler, a heater, and a vacuum generating device (steam ejector). Pressures of used vapor were 0.6 MPa gauge (medium pressure vapor) and 0.2 MPa gauge (low pressure vapor), and the vapor consumption was 8.6 T/h." and statements in descriptions (5a) to (5c) in Publication 5, the range "0.5 to 2 MPaG" specified by the

invention is a normal range for the driving pressure for steam ejectors in manufacture of acrylic acid.

Accordingly, it is a technical matter that a person skilled in the art could have easily carried out to specify the pressure range of the drive vapor in the invention of Publication 1.

C Regarding Different Feature 3

Furthermore, Different Feature 3 is examined below.

As stated in the statement in the description (1b) in Publication 1, "[0005] In the conventional methods for distilling represented by those methods, however, since vapor coming out from the condenser contains easily polymerizable compounds such as (meth)acrylic acid and/or (meth)acrylic acid ester, no stable operation can be carried out because of frequently repeated unavoidable shut-downs of the device due to generation of compounds in the line from the condenser to the ejector or the vacuum pump, in the line up to the pump, in the line from the condenser to the atmosphere or the industrial pretreatment facility, at the outlet section of the ejector, and the vacuum pump.

[0006]

[Problem to be solved by the Invention] Accordingly, the problem to be solved by the invention is to provide a method that can prevent polymerization inside the distillation device in distilling liquids comprising easily polymerizable compounds such as (meth)acrylic acid and (meth)acrylic acid ester." (underlines added by the body; the same applies hereinafter)," the invention of Publication 1 was made under recognition of the problem that, as vapor coming out from the condenser contains easily polymerizable compounds such as acrylic acid, polymers are generated in various sections such as the line from the condenser to the ejector or the vacuum pump, the line from the condenser to the atmosphere or the industrial pretreatment facility, the outlet area of the ejector and the vacuum pump, and polymers, and device shutdown is required frequently and no safe operation can be carried out, and, as stated in the statement in the description (1d), "In the vapor line at the inlet of the condenser, a method for preventing condensation of vapor that causes polymerization by heating with a tracing or jacket piping" is introduced as an addition means to prevent condensation, and there is a description of the means similar to that used for ejector in and there is a description of a means similar to that used for ejector in the invention.

In addition, as also described in descriptions (3a) to (3d) in Publication 3, and descriptions (6a) to (6c) in Publication 6, it is a well-known technical matter as a means

to heat the outer surface for solving the problem of adhering substances from the condenser on the inner surface of the ejector, and as is clear particularly from the statement in the description (6c), "since the ejector 20 is provided with a heating means for heating the suction chamber 62, the drive fluid (vapor, etc.) hardly stagnates in the suction chamber 62. Therefore, raw materials, (meth)acrylic acid and ethylene oxide, and the target product, hydroxyalkyl (meth)acrylate ester contained in the gas suctioned from the suction port 60 into the suction chamber 62 together with stagnated drive fluid (water, etc.) hardly stagnate in the suction chamber 62 and/or the diffuser 68. Accordingly, the suction chamber polymers hardly adhere to the inside of the suction chamber 62, etc.," it is a technical matter recognized already as of the priority date of the application that adherence of polymers as adhering substances to the ejector section can be suppressed with a heating means.

In addition, judging from the fact that, under recognition stated in the statement in the description (7b) in Publication 7, "[0012]

Since the whole pathway can be evenly heated and kept at a low temperature by using the passing device for easily polymerizable compounds of the invention, vapor phase comprising an easily polymerizable compound can be passed without condensing or polymerizing in the pathway. Since the temperature of reduced pressure water vapor can be easily adjusted to a temperature below 100°C, vapor of the easily polymerizable compound with a comparatively low temperature of, for example, around 70 to around 90°C can be passed through the pathway without condensing and polymerizing" and the statement in the description (7c) in Publication 7, "The passing device for easily polymerizable compounds of the invention can be preferably applied to a method for distilling easily polymerizable compounds by using a distillation column, a vapor delivering line, and a condenser. Namely, the cylinder constituting at least a part of the section from the void tower portion of the distillation column to the vapor delivering line is constituted with the passing device for easily polymerizable compounds of the invention.

As easily polymerizable compounds to which such method for distilling can be preferably applied, unsaturated carboxylic acids such as (meth)acrylic acid, ...," the statement in the description (8b) in Publication 8, "... to provide a method for distilling that reduces generation of compounds and makes continuous operation possible in distilling easily polymerizable compounds represented by vinyl compounds. Another objective of the Invention is to provide a distillation device that reduces generation of polymers and makes continuous operation possible.

[0008]

[Means for solving the problem] The inventors carried out intensive studies for finding a solution for the above problem to be solved by the invention, and, as a result, found out that the positions in the distillation column where the easily polymerizable compound tends to polymerize is a site in which vapor comprising the easily polymerizable compound gets condensed and tends to form liquid accumulation in a state in which no polymerization inhibitor exists. In addition, the inventors also found out that formation of areas in which easily polymerizable compounds tend to polymerize in the distillation column can be prevented by making vapor comprising the easily polymerizable compound not easily get condensed at sites in the column top and/or the pipe in which the vapor hardly contacts a liquid comprising polymerization inhibitor through heating at least a part of the column top and/or the pipe, and thereby completed the invention," and the statement in the description (9a) in Publication 9, "In devices (reactors, distillation columns, etc.) for handling easily polymerizable compounds ((meth)acrolein, (meth)acrylic acid, etc.), polymerization of easily polymerizable compounds tends to occur. Particularly, easily polymerizable compounds tend to stagnate on and polymers tend to adhere to the inner wall of the nozzle protruding from the main body of the device and the surface of the cover that closes the opening of the nozzle contacting the easily polymerizable compound. Therefore, it is necessary to frequently stop operation of the device and carry out maintenance (cleaning, etc.) of the device.

[0003]

The following device is proposed as a device in which stagnation of the easily polymerizable compound in the nozzle and clogging of the nozzle with polymers are suppressed.

An oxidation reactor provided with a main body of the reactor and a nozzle protruding from the reactor, wherein the reactor is further provided with a means to feed inert gas to the nozzle and a means to heat the nozzle (patent document 1).

[0004]

In the oxidation reactor, however, since replacement of atmosphere in the nozzle by inert gas is not sufficient, there is a problem that the easily polymerizable compound remains in the nozzle and the easily polymerizable compound tends to stagnate and polymers tend to adhere to the inner wall of the nozzle protruding from the main body of the device and the surface of the cover that closes the opening of the nozzle contacting the easily polymerizable compound. Therefore, it is necessary to frequently stop operation of the oxidation reactor and carry out maintenance (cleaning, etc.) of the oxidation reactor" (underlines added by the body; the same applies hereinafter), there is

a description of a means for solving in the description (9b), "... is provided with ... a nozzle heating means 24 provided in the circumference of the nozzle 14.

...

The nozzle 14 has a double pipe structure consisting of an outer pipe 26 (main body of the nozzle) whose base end communicates with the inside of the main body of the reactor 12 , and an inner pipe 28 inserted into the outer pipe 26," as well as statements in the descriptions (10a) to (10c) in Publication 10 and statements in descriptions (11a) and (11b) in Publication 11, it can be deemed that the problem of adhering substances by polymerization of polymerizable substances is a matter of common general technical knowledge in a device manufacturing as a whole including a distillation device for polymerizable substances such as acrylic acid, and it can be deemed to be a well-known conventional art to prevent condensation and/or polymerization of polymerizable substances by providing heating means from the outside in various places such as the vapor delivering line, the column top, the pipe, the nozzle, and areas around the reflux pipe, and the distillation column including ejectors in which vapor comprising polymerizable substances exists and can condensate, as described in Publications 6 to 11.

Accordingly, in the invention of Publication 1, it can be deemed to be a technical matter that a person skilled in the art could have easily carried out, in manufacturing polymerizable substances including acrylic acid, to apply a well-known conventional art of heating from the outside to a vapor (steam) ejector section for suppressing condensation and/or polymerization of polymerizable substances in sections including ejector section that had been recognized as a problem to be solved as a means to solve the problem.

C Advantageous effect of the Invention

The advantageous effect of the Invention is examined below.

From the statements in [0017] and [0057] in the specification of the Application, it can be deemed that the advantageous effect of the Invention is that, in a method for manufacturing acrylic acid that is an easily polymerizable compound, clogging of the steam ejector due to polymerization of easily polymerizable compounds can be prevented in the case a steam ejector is used as a pressure reducing device in the step of executing vacuum distillation, and it can be deemed advantageous in continuing operation for a long time.

As examined in above B, since the problem of adhering substances due to polymerization of polymerizable substances in the whole device for manufacturing

polymerizable substances such as acrylic acid including distillation devices, and it can be deemed to be a well-known conventional art to prevent condensation and/or polymerization of polymerizable substances by providing heating means from the outside in various sections including ejectors in which vapor comprising polymerizable substances can exist and can condensate and/or polymerize, the advantageous effects of the Invention as mentioned above are predictable to a person skilled in the art.

D Appellant's allegation

(A) Appellant alleges on page 3 of the written opinion dated April 10, 2018 that, since Cited Document 1 (Publication 1) does not have any description or suggestion regarding the pressure of the steam ejector, there is no motivation to include it in the scope of the Invention.

As examined with respect to Different Feature 2 in the above B, however, since it is obvious that a certain driving pressure to cause the effector function as a vacuum device, and it is known that the range specified by the Invention is known as a normal range for the driving pressure for a steam ejector in manufacture of acrylic acid, it is a technical matter that a person skilled in the art could have easily carried out to specify it to a range of around 0.5 to around 2 MPaG, and the above allegation by Appellant cannot be accepted.

(B) In addition, Appellant alleged in the written demand for trial dated October 22, 2018 that, based on reasons that, since polymerization of easily polymerizable compounds such as (meth)acrylic acid and (meth)acrylic acid ester occurs in a site in which the temperature decreases, it is effective for preventing polymerization to heat sites in which temperature decreases; namely, outlet areas of lines and ejectors, and vacuum pumps, sections heated in the Invention include the main body of the steam ejector (outer surface) and, right from the beginning, the temperature of such sections is high, a person skilled in the art does not consider to increase the temperature by further heating the main body of the steam ejector (outer surface), and, in addition, such advantageous effect that generation of polymers can be suppressed by heating the main body of the steam ejector (outer surface) cannot be predicted.

As examined with respect to Different Feature 3 in above B, however, as described in Publications 6 to 11, it is a well-known conventional art to prevent condensation and/or polymerization of polymerizable substances by providing heating means from the outside in various sections including ejectors in which vapor comprising polymerizable substances can exist and can condensate and/or polymerize, such as the

vapor delivering line, the column top, the pipe, the nozzle, areas around the reflux pipe, and each part of the distillation column, and, as described in [0055] in the specification of the Application, it is obvious that, if not heated, the ejector section has a temperature of the outer surface of 40 to 46°C under which polymerizable substances may get condensed depending on the position. In addition, as shown in [0012] in Publication 7 and [0026] to [0029] in Publication 8, there is no significant difference between suppressing condensation and/or polymerization of vapor of easily polymerizable compound by maintaining at around 70 to around 90°C and heating carried out in [0045] of the Invention.

In addition, it is not specified in Scope of Claim to limit the heating position of the steam ejector, and the allegation to discriminate the outlet section and main body from each other is not based on the Scope of Claim.

Accordingly, the above allegation by Appellant cannot be accepted.

(C) As described above, none of the appellant's allegation can be accepted.

No. 5 Closing

As described above, since the Invention could have easily been invented before the priority date of the Application by a person skilled in the art based on the invention disclosed in Publication 1 and the common general technical knowledge, and thus it cannot be granted a patent in accordance with the provisions of Article 29(2) of the Patent Act, without examining the inventions according to the other claims.

Therefore, the appeal decision shall be made as described in the conclusion.

June 25, 2019

Chief administrative judge: SASAKI, Hidetsugu
Administrative judge: SERA, Satoki
Administrative judge: SAITO, Mayumi