

SPECIFICATION**A Method for Preparing Acrylonitrile****Technical Field**

5 The present invention relates to a method for preparing acrylonitrile by ammoxydation, more particularly to a method for improving the yield and production efficiency of acrylonitrile by sufficiently removing ammonia in the quenching process of a reacted gas.

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Background Art

 The method for preparing acrylonitrile by ammoxydation has been improved on various points since its success in industrialization and it is said that the method
15 has technically matured. Still, there have been made efforts to improve utility unit consumption and to reduce production cost by achieving production efficiency.

 It is well known that the reacted gas obtained from various raw materials contains by-products such as hydrocyanic
20 acid, acetonitrile or aldehyde in addition to acrylonitrile when acrylonitrile is prepared by ammoxydation. The by-products react with acrylonitrile in the presence of unreacted ammonia or react one another; as a result, high boiling point compounds are produced. The high boiling point compounds
25 cause not only reduction in the yield of acrylonitrile, the target product, but also clogging at various parts in the

quenching tower in the further processes. Consequently, the by-products must be immediately separated from the reacted gas. Especially, the unreacted ammonia facilitating undesired reactions has to be completely removed as soon as possible.

5 Conventionally, there has been adopted the method for separating unreacted ammonia in the form of a salt and simultaneously removing other impurities and the by-products mentioned above by preliminarily cooling the reacted gas produced in a reactor, immediately sending the cooled reacted
10 gas to a quenching tower, and washing and quenching the reacted gas with water containing acids such as sulfuric acid. As one of the conventional methods excellent in production efficiency, there is proposed a method employing a multi-story quenching tower to divide a quenching process into two steps
15 or more, which comprises separating most of unreacted ammonia in the form of a salt by contacting the water containing a sufficient amount of sulfuric acid to neutralize the unreacted ammonia and simultaneously condensing not all but a part of vapor contained in the reacted gas in the first chamber, and
20 condensing most of the residual vapor in the second chamber (U.S. Pat. No. 3,649,179).

 This method succeeded in reducing the cost of recovering and treating the salt because of its higher collectability of ammonia and higher concentration of the
25 resultant ammonium salt than the conventional methods. Yet, the collectability of ammonia achieved by this method is not

sufficient. Accordingly, problems such as an acrylonitrile loss and a trouble caused by clogging due to high boiling point compounds (heavy) have still remained.

5 Disclosure of the Invention

The present inventors have made extensive and intensive research to solve the above problems and found that the collectability of ammonia is remarkably enhanced by adjusting a two-dimensional density of spray nozzles and the
10 amount of supplying water in certain ranges. As a result, they achieved the present invention.

This invention is to provide a method for preparing acrylonitrile by reacting propylene and/or propane, ammonia and molecular oxygen in the presence of a catalyst and
15 purifying the reacted gas in a multi-story quenching tower. The method comprises the steps of supplying the reacted gas into the first quenching chamber of the multi-story quenching tower, said first quenching chamber has a water supply pipe equipped with spray nozzles at a two-dimensional density of 2
20 nozzles/m² or more of the sectional area of the multi-story quenching tower, and contacting the reacted gas with 5T or more of water per 1T of the reacted gas fed from the nozzles. The unit "T" is a measurement of weight and corresponds to 1000 kg.

25 The quenching tower used in the present invention includes a multi-story quenching tower divided into two or

more levels and each level consists of one chamber as shown in Fig. 1.

The gas obtained by reacting propylene and/or propane, ammonia and molecular oxygen is first introduced into the first quenching chamber (2) to cool and wash it by contacting with water containing an acid such as sulfuric acid. At this time, unreacted ammonia, high boiling point compounds, polymers, scattered catalysts and the like are removed. After that, the remaining by-products are removed in the chambers on stories above the first.

In the multi-story quenching tower, spray nozzles (17) are arranged on the end part of a water supply pipe (9). On the upper stories, chambers are filled with porcelain Raschig rings as a packed bed (11).

As the spray nozzles (17), nozzles having the differential pressure of 1 to 5 kg/cm²G and the spraying angle of 90 to 100° can be used, for example, hollow cone type and full cone type nozzles. The two-dimensional density of the spray nozzles (17) in the first chamber (2) must be 2 nozzles/m² or more of the sectional area of the multi-story quenching tower. As a result, the areas where each spray nozzle (17) can spray water are preferably overlapped with one another so that excellent collectability of ammonia be obtained. When the two-dimensional density is less than 2 nozzles/m², water is not spread all over the sectional area of the quenching tower because the sprayed areas are not

preferably overlapped. Therefore, the scattered water and the reacted gas do not sufficiently contact and the preferable collectability of ammonia is not achieved. The two-dimensional density of the nozzles is preferably 2 to 8
5 nozzles/m², more preferably 2 to 5 nozzles/m².

The water supply pipe (9) is arranged above the gas leading port (19) so that the spray nozzles equipped on the pipe may face the bottom of the chamber. The distance between the port (19) and the spray nozzles is preferably 0.5 m or
10 more, more preferably 1.0 m or more though it depends on a diameter of the quenching tower.

Hereinafter, the production method in the present invention will be described referring to Fig. 1, an example of the multi-story quenching tower. The quenching tower has a
15 diameter of 2.5 to 3.0 m and a height of 9 to 10 m. In general, the reacted gas obtained by reacting propylene and/or propane, ammonia and molecular oxygen in the production of acrylonitrile has a composition as follows:

20	< ingredient >	< vol. % >
	acrylonitrile	6.0 - 7.0
	ammonia	0.0 - 1.0
	propylene and/or propane	0.2 - 0.6
	acetonitrile	0.1 - 0.5
25	hydrocyanic acid	0.8 - 1.6
	non-condensable gas	60.0 - 67.0

water vapor	25.0 - 30.0
other materials (acrolein, high boiling point compound, etc.)	0.0 - 0.2

This gas is supplied to the first quenching chamber
5 (2) through a gas leading pipe (1) at about 15 to 25 T/Hr at
260° to 280° C.

In the first chamber (2), water is supplied to the
chamber from the spray nozzles (17) in the direction against
the flow of the gas at 150 to 170 T/Hr. The water contacted
10 with the gas may be supplied through a discharging pipe (4)
and the water supply pipe (9) to the first quenching chamber
again. If necessary, a refrigerator (6) may be used.

The amount of the water supplied to the first chamber
(2) must be 5T or more per 1T of the reacted gas. If it is
15 less than 5T per 1T of the reacted gas, ammonia and high
boiling point compounds are not sufficiently collected. As a
result, the reacted gas flows up into the above stories and
the high boiling point compound clogs the packed bed (11) and
pipes. Further, the water is preferably fed in an amount of
20 20T or less per 1T of the reacted gas to obtain desirable
collectability of ammonia and utility unit consumption. The
more preferable amount of the water is 7T to 10T per 1T of the
reacted gas.

An acid necessary for neutralization of ammonia,
25 preferably sulfuric acid, is added to the water in the first
chamber (2). The acid is preferably added so as to adjust the

pH value of the water preferably to 5 to 6, more preferably to 5.3 to 5.8. Controlling the pH value within this range, loss of acrylonitrile can be minimized. When the pH value is too high or low, loss of acrylonitrile increases, and moreover,
5 the tower, packed beds, pipes and trays are soiled or clogged in a quenching step and an adsorption step following the quenching step.

Preferably, the reacted gas is supplied to the first chamber (2) at a linear velocity of 0.10 to 0.90 m/sec. When
10 the linear velocity of the reacted gas is more than 0.90 m/sec., the collectability of ammonia is likely to be reduced because the reacted gas tends to cause entrainment of water into the later steps. To the contrary, when the linear velocity of the reacted gas is less than 0.10 m/sec., the
15 effects of the present invention are not efficiently attained and the low linear velocity is not preferable in the aspects of initial investment in plant and equipment. The more preferable linear velocity is 0.50 to 0.80 m/sec.

The reacted gas treated in the first chamber (2) is
20 transferred to the upper stories than the first (Fig. 1 only shows a two-story quenching tower), and the removal of ammonia in the reacted gas is continued. On the upper stories, the reacted gas can be treated according to the conventional methods.

25 In the case of the quenching tower shown in Fig. 1, the water in the second chamber (3) is cooled down to about

36° to 38° C with a refrigerator (13) and supplied into the second chamber through the water supply pipe (15) at 170 to 190 T/Hr. The vapor initially contained in the reacted gas is only partially condensed, and most of the vapor is liquefied in this second chamber (3). The water used in the quenching chambers on stories above the first can be recycled in each quenching chamber on stories above the first. If necessary, the water containing an acid can be contacted with the reacted gas in the chambers on the upper stories.

10 The temperature in the first quenching chamber (2) at the time when the inside of the multi-story quenching tower reaches its equilibrium state is 80 to 100° C though it changes in some degree depending on the temperature and composition of the reacted gas transferred in the chamber.

15 When the inside of the quenching tower reaches its equilibrium state, the water in the first quenching chamber (2) becomes a solution containing an extremely small amount of acrylonitrile, ammonium sulfate and viscous high boiling point compounds. The water is discharged from the first quenching chamber (2)

20 out of the tower at about 0.5 to 1.0 T/Hr. The pressure at the bottom of the tower is about 0.4 to 0.6 kg/cm²G.

 The reacted gas, from which ammonia is removed in the multi-story quenching tower is transferred to an absorption tower. The temperature of the gas at the transferring pipe

25 (16) is about 37° to 39° C.

According to the present invention, collectability of ammonia of 90 % or more is achieved and other by-products can be effectively removed while maintaining the original effects of the multi-story quenching tower. Consequently, loss of acrylonitrile and clogging of pipes or packed beds due to the high boiling point compounds can be prevented.

Brief Description of Drawings

Fig. 1 is a schematic view of the multi-story quenching tower used in the present invention.

Description of Numerals

- 1: gas leading pipe
- 2: first quenching chamber
- 15 3: second quenching chamber
- 4: water discharging pipe
- 5: water discharging pipe out of the tower
- 6: refrigerator
- 7: acid controlling valve
- 20 8: acid supply pipe
- 9: water supply pipe
- 10: water discharging pipe
- 11: packed bed
- 12: water discharging pipe out of the tower
- 25 13: refrigerator
- 14: pH controlling meter

- 15: water supply pipe
- 16: gas transferring pipe
- 17: spray nozzle
- 18: gas transferring port
- 5 19: gas leading port
- 20: spray nozzle

Best Mode for Carrying Out the Invention

< Example 1 >

10 The multi-story quenching tower disclosed in Fig. 1
was used. The quenching tower has a diameter of 2.7 m and a
height of 9.7 m. In the first quenching chamber (2) of the
tower, spray nozzles (17) were arranged on the end part of a
pipe (9) at the two-dimensional density of 2.3 nozzles/m² for
15 the sectional area of the quenching tower. The distance from
the tip of the nozzle (17) to the gas leading port (19) was
0.5 m. The type of the nozzles was a hollow cone type.

Propylene, ammonia and molecular oxygen were reacted
in a reaction tower to obtain a reacted gas with the
20 composition as follows:

< ingredient >		< vol. % >
	acrylonitrile	6.5
	ammonia	0.5
25	propylene and propane	0.4
	acetonitrile	0.3

	hydrocyanic acid	1.2
	non-condensed gas	63.5
	water vapor	27.5
	other materials (acrolein,	
5	high boiling point compound, etc.)	0.1

The gas was introduced through the gas leading pipe (1) into the first quenching chamber (2) at 20 T/Hr. The linear velocity of the reacted gas was 0.66 m/sec. The ammonia contained in this reacted gas was 55 kg/Hr.

The water of 85° C, to which sulfuric acid was added so as to adjust the pH value of the water to 5.5, was sprayed from the spray nozzles (17) in an amount of 7.4T per 1T of the reacted gas at 148 T/Hr. The water was sprayed all over the section of the first quenching chamber (2).

93 % of ammonia contained in the reacted gas was neutralized and collected in the first quenching chamber (2). The water was discharged out of the tower at 0.8 T/Hr.

The reacted gas treated in the first quenching chamber (2) was transferred to the second quenching chamber (3) through the gas transferring port (18). The packed bed (11) of the second quenching chamber (3) was filled with porcelain Raschig rings and the spray nozzles (20) were equipped in the same manner as in the first chamber (2). The water of 37° C whose pH value was adjusted to 5.5 with sulfuric acid was supplied at 180 T/Hr from the spray nozzles

(20).

7 % of ammonia left in the reacted gas was completely collected in the second chamber (3); therefore, the gas exhausted through the transferring pipe (16) did not contain ammonia. When the system in the tower reached its equilibrium state, the pressure at the bottom of the tower was 0.5 kg/cm²G.

< Comparative Example 1 >

The reacted gas with the composition shown in Example 1 was purified in the same manner as in Example 1 except that the two-dimensional density of the nozzle was 3.7 nozzles/m² and the amount of water supplied to the chamber was 3.9 T per 1T of the reacted gas. The collectability of ammonia from the reacted gas was 77 %.

< Comparative Example 2 >

The reacted gas with the composition shown in Example 1 was purified in the same manner as in Example 1 except that the two-dimensional density of the nozzle was 1.9 nozzles/m². The collectability of ammonia from the reacted gas was 88 %.

20 Industrial Application

According to the method of the present invention, collectability of ammonia is improved so that a yield of acrylonitrile can be increased and the troubles such as clogging in the tower due to the high boiling point compounds are avoided.



What is claimed is:

1. A method for preparing acrylonitrile by reacting propylene and/or propane, ammonia and molecular oxygen in the presence of a catalyst and purifying the reacted gas in a multi-story quenching tower, said method comprising the steps of supplying the reacted gas into the first quenching chamber of the multi-story quenching tower, said first quenching chamber has a water supply pipe equipped with nozzles at a two-dimensional density of 2 nozzles/m² or more of the sectional area of the multi-story quenching tower and contacting the reacted gas with 5T or more of water per 1T of the reacted gas fed from the nozzles.

2. A method for preparing acrylonitrile according to claim 1, wherein the reacted gas is supplied at a linear velocity of 0.1 to 0.9 m/sec. in the first chamber.

3. A method for preparing acrylonitrile according to claim 1, wherein the spray nozzle is selected from the group consisting of a full cone type nozzle and a hollow cone type nozzle.