

DESCRIPTION

METHOD FOR PRODUCTION OF DIALKYLAMINOALKYL (METH)ACRYLATE
QUATERNARY SALT

5 TECHNICAL FIELD

The present invention relates to a method of producing a dialkylaminoalkyl (meth)acrylate quaternary salt.

BACKGROUND ART

10 Dialkylaminoalkyl (meth)acrylate quaternary salts (hereinafter referred to as "DAM") are raw materials, etc., for cationic polymers, and are used for various purposes as raw materials for the production of polymer coagulants, paper strength-reinforcing agents, antistatic agents, soil conditioners,
15 and the like.

DAM is generally produced in the form of an aqueous solution of DAM. During production, using water as a solvent, raw materials, i.e., dialkylaminoalkyl (meth)acrylate (hereinafter referred to as "Da") and a quaternizing agent (e.g., methyl
20 chloride, etc.), are supplied in the presence of the solvent, and the mixture is reacted while removing the heat of the reaction. DAM is produced by a batch process in which raw materials are charged in one reactor, and products are removed after the reaction (for example, Patent Documents 1 and 2); or by a
25 continuous process in which raw materials are supplied to a reactor while the reaction mixture is continuously extracted from the reactor (Patent Document 3).

Moreover, the present inventors found that by supplying a quaternizing agent to the reactor by an ejector, the following
30 advantages are achieved: the reaction mixture does not flow backward into the quaternizing agent supply pipe; the quaternizing agent is sufficiently introduced in the reaction mixture; the solubility of the quaternizing agent in the reaction mixture is improved; and the reactivity becomes superior (Patent
35 Document 4).

Patent Document 1: Japanese Unexamined Patent Publication No.
1992-217649

Patent Document 2: Japanese Unexamined Patent Publication No.
1996-268985

5 Patent Document 3: Japanese Unexamined Patent Publication No.
2003-342244

Patent Document 4: Japanese Unexamined Patent Publication No.
2004-155669

10 DISCLOSURE OF THE INVENTION

Problems to be Solved by the Invention

Methyl chloride is generally used as a quaternizing agent, which is one of the above-mentioned raw materials for DAM. Methyl chloride forms an explosive gas mixture (a detonating gas;
15 explosive range of methyl chloride: 10.7 to 17.4 V/V%) upon mixing with air. Therefore, when using methyl chloride as a quaternizing agent, the control of the gas composition in the gas phase portion of the reactor is extremely important for the safe and stable production of DAM.

20 An overly low oxygen concentration in the gas phase portion causes polymerization of DAM in the reactor, producing only low-quality DAM. An overly low methyl chloride concentration slows quaternization reaction, promoting hydrolysis reaction. As a result, the hydrolyzed by-product, (meth)acrylic acid
25 (hereinafter referred to as "AA"), increases, and the product value of DAM is reduced.

However, there have been no reported examples to date of close examination of the gas composition in the gas phase portion of the reactor.

30 Therefore, the present invention aims to provide a method of producing high-purity DAM with high productivity while maintaining safety, by controlling the gas composition in the gas phase portion of the reactor within a predetermined range.

35 Means for Solving the Problem

The inventors conducted extensive research to solve the above-described problems, and found that high-purity DAM can be safely produced with high productivity as follows. Reaction is carried out while controlling the concentration of oxygen (hereinafter referred to as $[O_2]$), the concentration of methyl chloride (hereinafter referred to as $[MC]$), and the ratio of oxygen concentration to methyl chloride concentration ($[O_2]/[MC]$) in the gas phase portion of the reactor within a predetermined range. As a result of further research based on these findings, the present invention has been accomplished.

Specifically, the present invention provides a method of producing dialkylaminoalkyl (meth)acrylate quaternary salts, as described below.

Item 1. A method of producing a dialkylaminoalkyl (meth)acrylate quaternary salt by supplying a dialkylaminoalkyl (meth)acrylate, methyl chloride, and water to a reactor,

wherein a reaction is carried out while controlling the concentration of oxygen ($[O_2]$) to 3 to 12 vol.%, the concentration of methyl chloride ($[MC]$) to 22 to 80 vol.%, and the ratio of oxygen concentration to methyl chloride concentration ($[O_2]/[MC]$) to 0.05 to 0.5, in a gas phase portion of said reactor.

Item 2. A method according to Item 1, wherein a reaction is carried out while controlling the ratio of oxygen concentration to the sum of oxygen concentration and nitrogen concentration ($[O_2]/([O_2]+[N_2])$) in the gas phase portion of the reactor to 0.05 to 0.2.

Item 3. A method according to Item 1 or 2, wherein the reactor is previously charged with a dialkylaminoalkyl (meth)acrylate quaternary salt aqueous solution before a dialkylaminoalkyl (meth)acrylate, methyl chloride, and water are supplied.

Item 4. A method according to Item 1, 2, or 3, wherein the sum of dialkylaminoalkyl (meth)acrylate quaternary salt concentration ($[DAM]$), dialkylaminoalkyl (meth)acrylate concentration ($[Da]$), and methyl chloride concentration ($[MC]$) in the reaction mixture is maintained at 75 to 84 wt.% during reaction.

Item 5. A method according to any one of Items 1 to 4, wherein the temperature of the reaction mixture is maintained at 30 to 80°C during reaction.

Item 6. A method according to any one of Items 1 to 5, wherein
5 the reactor is a sealed reactor, and wherein the internal pressure of said reactor is maintained at 0.10 to 1 MPaG during reaction.

Item 7. A method according to any one of Items 1 to 6, further comprising continuously supplying a dialkylaminoalkyl
10 (meth)acrylate, methyl chloride, and water to the reactor, while continuously extracting a reaction mixture from said reactor.

Item 8. A method according to any one of Items 1 to 7, wherein the dialkylaminoalkyl (meth)acrylate is dimethylaminoethyl (meth)acrylate.

15 Item 9. A method according to any one of Items 1 to 8, wherein the methyl chloride is supplied using an ejector.

Item 10. A method according to Item 9, further comprising:
extracting a part of the reaction mixture from the reactor;

20 cooling said reaction mixture by passing through a heat exchanger; and

supplying said cooled reaction mixture, together with methyl chloride, to said reactor using an ejector.

Item 11. A method according to any one of Items 1 to 10, further
25 comprising:

supplying the reaction mixture continuously extracted from the reactor to one or more sealed tanks; and then

gradually adjusting the pressure of the reaction mixture in said tank to atmospheric pressure.

30

Effect of the Invention

According to the present invention, high-purity DAM can be produced with high productivity while maintaining safety, by controlling the gas composition in the gas phase portion of the
35 reactor within a predetermined range during reaction.

Moreover, this method becomes superior for DAM production by controlling the temperature, pressure, etc., in the reactor and the concentration of DAM and Da in the reaction mixture within a predetermined range.

5 Therefore, the production method of the present invention is also quite advantageously suitable for industrial scale DAM production.

Brief Description of the Drawings

10 Fig. 1 schematically illustrates an example of a reactor.

Fig. 2 schematically illustrates an example of a reactor equipped with an ejector.

15 Fig. 3 schematically illustrates an example of a reaction apparatus comprising a reaction tank A, an ageing tank B, and a finishing tank C.

Description of Symbols

1: Water supply pipe
20 2: Da supply pipe
3: Methyl chloride supply pipe
4: Reaction-mixture extraction port
4': Reaction-mixture transfer pipe
5: Agitating blade
25 6: Reactor
7: Circulating pump
8: Ejector
9: Suction part
10: Jacket-type heat exchanger
30 11: Nozzle part
12: Reaction-mixture transfer pipe

BEST MODE FOR CARRYING OUT THE INVENTION

Embodiments of the present invention are described in
35 detail below. The present invention is not limited to these

embodiments, and can be suitably modified as long as the gist of the invention is unchanged. Note that the term (meth)acrylate in the specification means acrylate or methacrylate.

5 The production method of the present invention is described in detail below with reference to Figs. 1 to 3.

The present invention is a method of producing DAM by supplying Da, methyl chloride, and water to a reactor, wherein a reaction is carried out while adjusting the concentration of oxygen (hereinafter referred to as $[O_2]$) to 3 to 12 vol.%, the
10 concentration of methyl chloride (hereinafter referred to as $[MC]$) to 22 to 80 vol.%, and the ratio of oxygen concentration to methyl chloride concentration (hereinafter referred to as $[O_2]/[MC]$) to 0.05 to 0.5, in the gas phase portion of the reactor.

Reactor

15 The reactor used in the present invention is not limited in size, material, and structure as long as raw materials can be reacted to produce DAM.

As the reactor, those whose inner surface is coated and/or lined with glass, Teflon[®], or the like are preferable, so
20 that elution of metal components into the obtained DAM aqueous solution can be avoided and a high-purity DAM aqueous solution can be obtained. Moreover, sealable or pressurizable apparatuses are preferably used as a reactor. Examples of such a reactor include autoclaves, etc.

25 Fig. 1 shows an example of a reactor usable in the present invention. The reactor comprises a Da supply pipe (2), a water supply pipe (1), a methyl chloride supply pipe (3), an agitating blade (5), and a reaction-mixture extraction port (4). Moreover, the reactor is usually equipped with a jacket-type heat
30 exchanger (10).

In addition, for better dispersibility of methyl chloride in the reaction mixture, methyl chloride is preferably supplied by an ejector. Fig. 2 shows an example of a reactor equipped with an ejector (8). The ejector is also referred to as
35 an aspirator. As shown in Fig. 2, the reaction mixture is

extracted from the reactor by a circulating pump (7) and then supplied to a nozzle part (11) of the ejector (8). Then, the pressure in the ejector is reduced, and by the driving force generated here, methyl chloride is supplied from a suction part
5 (9) into the reaction mixture. Generally, the reaction-mixture extraction port (4) is mounted in the bottom of the reactor, and the ejector is mounted in the upper portion of the reactor. The reactor may have one or more ejectors, and the optimal number of ejectors can be determined depending on the size of the reactor
10 and the size of the reaction.

Further, a heat exchanger (not shown) for cooling the extracted reaction mixture may be provided in a flow path for supplying the reaction mixture extracted by the circulating pump (7) from the reactor (6) to the ejector (8). Such a heat
15 exchanger can efficiently remove heat from the reactor, and temperature control in the reactor is greatly simplified.

Moreover, in addition to the above reactor, a finishing tank and ageing tank for treating the reaction mixture may be provided.

20 Supply of Raw Materials

In the DAM-producing method of the present invention, a dialkylaminoalkyl (meth)acrylate, methyl chloride, and water are used as raw materials.

Specific examples of Da include dimethylaminoethyl
25 (meth)acrylate, diethylaminoethyl (meth)acrylate, dipropylaminoethyl (meth)acrylate, dimethylaminopropyl (meth)acrylate, etc. Among them, dimethylaminoethyl (meth)acrylate is preferable.

Methyl chloride is flammable, but burns weakly. It
30 forms an explosive gas mixture upon mixing with air. The explosive range is 10.7 to 17.4 V/V%, and the boiling point is -24.1°C.

As the above-mentioned water used in the present invention, the purest possible water free from metals, etc., is
35 preferably used. Specific examples include distilled water, ion

exchange water, etc.

In the present invention, each raw material is supplied in a constant amount to the reactor. Reaction may be conducted either by a batch process or a continuous process. In terms of
5 DAM productivity, the quality of DAM, and ease of reaction control, a continuous process is preferable. Specifically, in a continuous process, Da, methyl chloride, and water are continuously supplied to the reactor while the reaction mixture is continuously extracted from the reactor.

10 Methyl chloride can be supplied to the reactor in various ways. For example, methyl chloride can be introduced, in gaseous form, directly into the gas phase portion of the reactor or into the reaction mixture in the reactor; or supplied in liquefied form. Methyl chloride, when supplied using an ejector,
15 is supplied into the reactor through the suction part (9) of the ejector (8) (Fig. 2). Da and water can be supplied in accordance with a standard manner.

Da, methyl chloride, and water may be supplied into the reaction mixture or to the upper space of the reaction mixture in
20 the reactor. As described above, methyl chloride, when supplied by an ejector, is preferably supplied into the reaction mixture. Further, in order to avoid deposition of DAC crystals during the supply of Da, which causes obstruction of the introducing pipe, Da is preferably supplied from the tip of a Da-introducing pipe
25 located in the upper space above the reaction mixture.

The present invention preferably employs a DAM aqueous solution as a reaction solvent. When raw materials are reacted without solvent, crystalline DAM is obtained. Due to the hygroscopic nature, DAM in a crystal form attaches to the inner
30 wall of the reactor, and thus heat removal becomes difficult. Additionally, since DAM takes the form of crystals, an oxygen-free condition occurs, causing the polymerization of DAM. In order to solve this problem, some methods use water as a solvent. In this case, however, the Da of the raw materials is hydrolyzed,
35 leading to decreased product purity. In contrast, the present

invention preferably uses a DAM aqueous solution as a reaction solvent to efficiently produce high-purity DAM.

In the present invention, the above-mentioned DAM aqueous solution (reaction solvent) and each raw material can be supplied to the reactor in any order as long as the reaction of the raw materials proceeds. Specifically, in one method, for example, the DAM aqueous solution is previously charged in the reactor before each raw material is supplied; in another method, the DAM aqueous solution is supplied to the reactor simultaneously with or during the supply of the raw materials. The former method is preferable because supplying each raw material after previously charging a DAM aqueous solution as a reaction solvent in the reactor can reduce problems such as the crystallization of DAM and following polymerization of DAM, and the hydrolysis of the raw material Da.

DAM contained in the previously-charged DAM aqueous solution is generally the same type of DAM as that obtained by reaction. The concentration of the DAM aqueous solution can be selected from a wide range. Usually, the concentration of the DAM aqueous solution is 50 to 84 wt.%, preferably 75 to 84 wt.%, and more preferably 78 to 83 wt.%.

In the DAM-producing method of the present invention, Da, methyl chloride, and water may be supplied to the reactor so that the following reaction conditions are satisfied inside the reactor. Moreover, since the amounts of nitrogen and oxygen contained in Da and water vary with temperature, a small amount of gas in the gas phase portion can be extracted, as required; or, nitrogen and air can be supplied, as required. In addition, nitrogen and air can be supplied by blowing nitrogen and air into Da and water.

[O₂] in the gas phase portion of the reactor is controlled to 3 to 12 vol.%, preferably 4 to 10 vol.%, and more preferably 5 to 9 vol.%. [MC] is controlled to 22 to 80 vol.%, preferably 35 to 65 vol.%, and more preferably 40 to 55 vol.%.

Moreover, [O₂]/[MC] in the gas phase portion of the

reactor is controlled to 0.05 to 0.5, preferably 0.07 to 0.35, and particularly preferably 0.10 to 0.2.

5 This range ensures safe reaction without forming a detonating gas, as well as the production of high-purity DAM with high productivity.

Moreover, it is preferable to control the ratio of oxygen concentration to the sum of oxygen concentration and nitrogen concentration ($[\text{O}_2]/([\text{O}_2] + [\text{N}_2])$) in the gas phase portion of the reactor to 0.05 to 0.2, further 0.07 to 0.18, and 10 particularly 0.09 to 0.15. This range ensures highly safe and stable production.

The concentrations of these gases can be obtained by extracting the gas in the gas phase portion of the reactor, and measuring by gas chromatography.

15 Moreover, using a sealed reactor as a reactor, the pressure inside the reactor during reaction is preferably maintained at 0.1 to 1 MPaG, preferably 0.1 to 0.8 MPaG, and more preferably 0.1 to 0.6 MPaG. This can increase the amount of methyl chloride dissolved in the reaction mixture.

20 Furthermore, after the supply of raw materials and the start of reaction, Da, water, and methyl chloride are preferably supplied so that the sum of the DAM concentration ($[\text{DAM}]$), the Da concentration ($[\text{Da}]$), and the methyl chloride concentration $[\text{MC}]$ in the reaction mixture is 75 to 84 wt.%, more preferably 77 to 84 wt.%, and particularly preferably 79 to 84 wt.%. This is 25 primarily for the suppression of the hydrolysis of Da and DAM. Moreover, when MC is supplied using an ejector, an overly high concentration increases the viscosity of the reaction mixture, reducing flowability of the reaction mixture supplied from the 30 pump. Thus, the capacity of the ejector may decline.

In order to arrange conditions in the reactor during reaction within the above range, the rate of supplying each raw material may be suitably determined depending on the size of the reactor, the production volume per unit time, etc.

35 For example, Da is preferably supplied at a rate of 3

to 70 mass%/hour, and more preferably 5 to 35 mass%/hour, with respect to the mass of the reaction mixture in the reactor.

Methyl chloride is preferably supplied at a rate of 1 to 30 mass%/hour, and more preferably 2 to 15 mass%/hour, with respect to the mass of the reaction mixture in the reactor.

Water is preferably supplied at a rate of 1 to 30 mass%/hour, and more preferably 2 to 15 mass%/hour, with respect to the mass of the reaction mixture in the reactor.

It is preferable in the present invention to keep the inside of the reactor at a constant temperature in a step to continuously supply each raw material to the reactor and in a reaction step. The reaction temperature is preferably 30 to 80°C, and more preferably 40 to 70°C.

As described above, in the present invention, when reaction is conducted by a batch process, the reaction mixture is extracted through the reaction-mixture extraction port after the reaction. When reaction is conducted by a continuous process, the reaction mixture is extracted from the reactor while supplying each raw material during the reaction. The reaction mixture can be subsequently supplied to a finishing tank or an ageing tank.

When employing a continuous process, the rate of extraction of the reaction mixture may be suitably determined according to the amount of supplied raw materials. The reaction mixture is preferably extracted at a rate of 5 to 130 mass%/hour, and more preferably 9 to 65 mass%/hour, with respect to the mass of the reaction mixture in the reactor. Under such conditions, reactions progress with a suitable raw material proportion, ensuring the production of high-purity DAM with high productivity.

Finishing Tank

In the present invention, it is preferable that the reaction mixture extracted from the reactor be supplied to one or more sealed finishing tanks, and that the pressure of the reaction mixture in the finishing tank be subsequently adjusted to atmospheric pressure. Here, the term finishing tank means a tank used to remove methyl chloride from the reaction mixture by

reducing the pressure of the reaction mixture to atmospheric pressure, and further aerating the reaction mixture as required. The reaction mixture extracted from the reactor may be directly treated in the finishing tank (for example, see Fig. 3), or
5 treated in the finishing tank after being treated in an ageing tank, described later.

Unreacted methyl chloride dissolved in the reaction mixture in the reactor is hydrolyzed to hydrogen chloride, which is unfavorable for the quality of final products. Therefore, it
10 is necessary to remove unreacted methyl chloride. However, if the pressure of the reaction mixture is immediately returned to atmospheric pressure, methyl chloride is vaporized at a stretch, and simultaneously oxygen dissolved in the reaction mixture is also vaporized. As a result, the reaction mixture enters a low-
15 oxygen condition, thereby causing the polymerization of DAM in the reaction mixture.

The pressure inside the finishing tank is not limited. Preferably, the pressure is not more than that in the reactor, more preferably atmospheric pressure to 0.7 MPaG, and further
20 preferably atmospheric pressure. The polymerization of DAM in the reaction mixture can be preferably prevented in this pressure range.

The above-mentioned problem can be resolved by using a finishing tank to gradually reduce the pressure of the extracted
25 reaction mixture. The reaction mixture may be supplied either continuously or intermittently to the finishing tank; however, continuous supply is preferred for the stable production of DAM.

In the present invention, a gas containing at least oxygen can be blown into the reaction mixture transferred to the
30 finishing tank. This is preferable because oxygen is supplied to the reaction mixture in the finishing tank to prevent the polymerization of DAM, and unreacted components after reaction, particularly methyl chloride, can be removed. The type of such a gas is not limited as long as at least oxygen is contained.
35 Generally, air is used.

The number of finishing tanks may be at least one, but a larger number of tanks are preferable in terms of removing methyl chloride. The number of finishing tanks is generally one or more, preferably two or more, and more preferably three or more. The size, material, and structure of the finishing tank are suitably selected according to used raw materials, the production volume of DAM, the purposes, etc. Specifically, those exemplified as the above-mentioned reactor can be used.

As described above, in order to suppress the hydrolysis of Da in the reactor, it is preferable to produce DAM at a high concentration. However, to reduce viscosity for easy handling and to prevent crystallization in winter, final products preferably contain 60 to 80 mass% of DAM. In this case, water is preferably added to the finishing tank to adjust the concentration suitable for final products.

Ageing Tank

In the present invention, it is preferable that the reaction mixture continuously extracted from the reactor is further supplied to one or more sealed ageing tanks. The term ageing tank means a tank used to increase the reaction rate for producing DAM (to age) more than that in the reaction mixture extracted from the reactor. The reactor, ageing tank, and finishing tank are usually connected in this order.

Specifically, a reaction can be aged by, for example, maintaining the pressure in the ageing tank below the pressure in the reactor, and also reducing the temperature in the ageing tank below the temperature in the reactor.

The use of an ageing tank is preferable because even when reaction does not sufficiently progress in the reactor due to reaction conditions, etc., the reaction can sufficiently progress in the ageing tank. In addition, residual unreacted raw materials in products can be avoided. Although the reaction mixture may be supplied either continuously or intermittently to the ageing tank, continuous supply is preferred for the stable production of DAM.

The pressure and temperature in the ageing tank are not limited. The pressure in the ageing tank is generally lower than the pressure in the reactor. When using an ageing tank, the pressure in the ageing tank can be adjusted to atmospheric
5 pressure. The pressure is preferably atmospheric pressure to 0.8 MPaG, and more preferably atmospheric pressure to 0.6 MPaG. The temperature in the ageing tank may be generally below the temperature in the reactor, and is preferably 30 to 60°C. The pressure in the ageing tank is preferably maintained constant.
10 Further, the temperature in the ageing tank is also preferably maintained constant.

It is preferable that the reaction mixture, after being supplied to the ageing tank for ageing, is further supplied to the finishing tank. In this case, it is preferable to gradually
15 reduce the pressures in the reactor, ageing tank, and finishing tank. This can prevent the polymerization of DAM in the low-oxygen conditioned reaction mixture, which occurs because the methyl chloride and oxygen dissolved in the reaction mixture are vaporized at a stretch. When the ageing tank is adjusted to
20 atmospheric pressure, only aeration and the addition of water may be performed in the finishing tank.

The number of ageing tanks in the present invention may be at least one; a larger number of tanks is preferable, however, in terms of productivity improvement. The number of ageing tanks
25 is generally one or more, preferably two or more, and more preferably three or more. When two or more ageing tanks are connected, it is preferable to gradually reduce the pressure. Specifically, when two ageing tanks are connected, the pressure is preferably 0.05 to 0.8 MPaG in the first ageing tank, and
30 atmospheric pressure to 0.5 MPaG in the second ageing tank. Moreover, the size, material, and structure of the ageing tank may be suitably selected according to the raw materials used, the production volume of DAM, the purposes, etc. Specifically, those exemplified as the reactor can be used.

35 The residence time ([the mass of reaction mixture])/[the

rate of extraction of reaction mixture (mass/hour)]) in the present invention can be determined to cover various ranges, depending on reaction conditions, the number of finishing tanks or ageing tanks, and other conditions. Usually, for a two-tank
5 continuous process using a finishing tank, the above residence time is generally 4 hours or more, preferably 4.5 hours or more, and more preferably 5 hours or more. For a three-tank continuous process using an ageing tank and a finishing tank, the residence
10 time is generally 1 hour or more, preferably 1.5 hours or more, more preferably 2 hours or more, and further more preferably 2 to 15 hours. For the residence time in this range, the amount of residual acrylic acid derived from the above-mentioned raw materials is preferably decreased. The amount of residual acrylic acid derived from the raw materials in DAM obtained in the
15 present invention is usually 1,500 ppm or less, preferably 1,200 ppm or less, and more preferably 1,000 ppm or less.

Examples

The present invention is described in detail below with
20 reference to examples. Note that the term "%" means "mass%".

Example 1

Using a reaction apparatus comprising a reaction tank A, an ageing tank B, and a finishing tank C, as shown in Fig. 3, a methyl chloride adduct (hereinafter referred to as "DAC") of
25 dimethylaminoethyl acrylate was produced.

As each of the reaction tank A, ageing tank B, and finishing tank C, a 10 m³ tank reactor comprising an inner agitator, an outer jacket-type heat exchanger (not shown), and a glass-lined inner wall was used.

30 After 8,000 kg of 81% DAC aqueous solution was charged in the reaction tank A, the tank was sealed and heated to 50°C.

Subsequently, while stirring the DAC aqueous solution with the agitator of the reaction tank A, methyl chloride (hereinafter abbreviated as "MC") was supplied from a supply pipe
35 (3) to the sealed reaction tank (6) via a suction part (9) of an

ejector (11). MC was continuously supplied at a rate of 410 kg/hour, and, in order to maintain the pressure inside the reaction tank A at 0.28 MPaG, only the amount of MC consumed by reaction was supplied.

5 Then, dimethylaminoethyl acrylate (hereinafter referred to as "DA") was continuously supplied at a rate of 1,040 kg/hour to the reaction tank A via a supply pipe (2). At the same time, pure water was continuously supplied at a rate of 300 kg/hour to the reaction tank A via a supply pipe (1).

10 Simultaneously with the start of supply of each raw material, the reaction mixture was continuously extracted from the reaction tank A to the ageing tank B through a transfer pipe (4'), and the pressure was reduced to 0.12 MPaG. Further, the reaction mixture was continuously extracted from the ageing tank
15 B to the finishing tank C through a transfer pipe (12). At this time, the pressure in the finishing tank C was maintained at atmospheric pressure, while air was blown into the reaction mixture in the finishing tank C to remove superfluous MC dissolved in the reaction mixture. In order to obtain a product
20 concentration of 79%, pure water was continuously supplied at 95 kg/hour to the finishing tank C.

 At this time, the total concentration of DAC, DA, and MC in the reaction tank A was 81.0%, and the concentrations of MC and oxygen in the gas phase portion of the reaction tank A were
25 46.1% and 6.5%, respectively. The values of the total concentration were calculated from the charged amount, and the values of the concentration in the gas phase portion were analyzed by gas chromatography.

 As a result, a 79% DAC aqueous solution could be stably
30 and continuously produced at a rate of 1,875 kg/hour.

 The content of acrylic acid (hereinafter abbreviated as "AA") in the final product DAC aqueous solution was measured, and the condition of the final product was visually observed. Table 1 shows the results. The values of the AA concentration were
35 analyzed by liquid chromatography.

Examples 2 and 3

DAC was continuously produced in the same manner and amount as in Example 1, except that the amounts of pure water supplied to the reaction tank A were 395 kg/hour (Example 2) and 260 kg/hour (Example 3), and the amounts of pure water supplied to the finishing tank C were 0 kg/hour (Example 2) and 135 kg/hour (Example 3).

Table 1 shows the total concentration of DAC and DA in the reaction tank, and each concentration of MC and oxygen in the gas phase portion of the reaction tank.

The final product DAC aqueous solution was evaluated in the same manner as in Example 1. Table 1 shows the results.

Examples 4 and 5

DAC was produced in the same manner and amount as in Example 1, except that the amounts of MC supplied to the reaction tank A were 435 kg/hour (Example 4) and 390 kg/hour (Example 5).

Table 1 shows the total concentration of DAC and DA in the reaction tank, and each concentration of MC and oxygen in the gas phase portion of the reaction tank.

The final product DAC aqueous solution was evaluated in the same manner as in Example 1. Table 1 shows the results.

Example 6

DAC was produced in the same manner and amount as in Example 1, except that the amount of pure water supplied to the reaction tank A was 200 kg/hour, and the amount of pure water supplied to the finishing tank C was 195 kg/hour.

Table 1 shows the total concentration of DAC and DA in the reaction tank, and each concentration of MC and oxygen in the gas phase portion of the reaction tank.

The final product DAC aqueous solution was evaluated in the same manner as in Example 1. Table 1 shows the results.

Table 1

Example		1	2	3	4	5	6
Raw material supply (kg/hr)	DA supply	1,040	1,040	1,040	1,040	1,040	1,040
	Pure water supply to reaction tank	300	395	260	300	300	200
	MC supply	410	410	410	435	390	410
	Pure water supply to finishing tank	95	0	135	95	95	195
Concentration in water phase portion and gas phase portion (%)	Total concentration of DAC, DA, and MC in reaction tank	81.0	78.7	82.4	81.4	81.1	84.3
	[MC] in gas phase portion	46.1	45.5	44.6	54.3	38.3	44.6
	[O ₂] in gas phase portion	6.5	7.2	7.2	4.1	7.3	7.2
	[O ₂]/[MC]	0.14	0.16	0.16	0.08	0.19	0.16
	[O ₂]/([O ₂]+[N ₂])	0.12	0.13	0.13	0.09	0.12	0.13
Reactor condition		No problem	No problem	No problem	No problem	No problem	Load on circulating pump
Product quality	AA concentration (ppm)	624	932	569	553	434	434
	Product condition	No problem	No problem	No problem	No problem	No problem	No problem
	Product quality	No problem	No problem	No problem	No problem	No problem	No problem

In the Examples, DAC could be produced stably, and the final products had lower AA concentrations as well as excellent product quality. In Examples 1 to 5, the amount of supplied water was within a more preferable range, and no load was imposed on the circulating pump.

Comparative Examples 1 to 3

DAC was produced in the same manner and amount as in Example 1, except that the amounts of supplied MC were 460 kg/hour (Comparative Example 1), 500 kg/hour (Comparative Example 2), and 370 kg/hour (Comparative Example 3).

Table 2 shows the total concentration of DAC and DA in the reaction tank, and each concentration of MC and oxygen in the gas phase portion of the reaction tank.

The final product DAC aqueous solution was evaluated in the same manner as in Example 1. Table 2 shows the results.

Table 2

Comparative Example		1	2	3
Raw material supply (kg/hr)	DA supply	1,040	1,040	1,040
	Pure water supply to reaction tank	300	300	300
	MC supply	460	500	370
	Pure water supply to finishing tank	95	95	95
Concentration in water phase portion and gas phase portion (%)	Total concentration of DAC, DA, and MC in reaction tank	81.6	81.4	81.0
	[MC] in gas phase portion	64.6	80.1	20.6
	[O ₂] in gas phase portion	2.8	0.8	7.8
	[O ₂]/[MC]	0.04	0.01	0.38
	[O ₂]/([O ₂]+[N ₂])	0.08	0.04	0.10
Reactor condition		No problem	Polymerization occurred in reactor	No problem
Product quality	AA concentration (ppm)	480	-	1,540
	Product condition	Slightly sticky	-	No problem
	Product quality	Existence of polymer	-	Overly high AA concentration

In Comparative Example 1, the values of $[O_2]$ and $[O_2]/[MC]$ in the gas phase portion were slightly less than the lower limit of the present invention. Although the reaction proceeded without any problems, the final product was sticky and
5 contained polymer. Thus, the product quality was inadequate.

In Comparative Example 2, the values of $[O_2]$ and $[O_2]/[MC]$ in the gas phase portion were significantly less than the lower limit of the present invention, and the value of $[MC]$ slightly exceeded the upper limit of the present invention.
10 Polymerization of DAC occurred in the reactor.

In Comparative Example 3, the value of $[MC]$ in the gas phase portion was less than the lower limit of the present invention. Although the reaction proceeded without any problems, an overly low methyl chloride concentration slowed the
15 quaternization reaction, thereby promoting hydrolysis. As a result, the final product had a high AA concentration. Thus, the product quality was inadequate.

Industrial Applicability

20 According to the DAM-producing method of the present invention, high-purity DAM can be safely produced with high productivity. Therefore, the method is suitable for the industrial scale production of DAM.

Claims

1. A method of producing a dialkylaminoalkyl
(meth)acrylate quaternary salt by supplying a dialkylaminoalkyl
5 (meth)acrylate, methyl chloride, and water to a reactor,
wherein a reaction is carried out while controlling the
concentration of oxygen ($[O_2]$) to 3 to 12 vol.%, the concentration
of methyl chloride ($[MC]$) to 22 to 80 vol.%, and the ratio of
oxygen concentration to methyl chloride concentration ($[O_2]/[MC]$)
10 to 0.05 to 0.5, in a gas phase portion of said reactor.
2. A method according to claim 1, wherein a reaction is
carried out while controlling the ratio of oxygen concentration
to the sum of oxygen concentration and nitrogen concentration
15 ($[O_2]/([O_2]+[N_2])$) in the gas phase portion of the reactor to 0.05
to 0.2.
3. A method according to claim 1 or 2, wherein the
reactor is previously charged with a dialkylaminoalkyl
20 (meth)acrylate quaternary salt aqueous solution before a
dialkylaminoalkyl (meth)acrylate, methyl chloride, and water are
supplied.
4. A method according to claim 1, 2, or 3, wherein the
25 sum of dialkylaminoalkyl (meth)acrylate quaternary salt
concentration ($[DAM]$), dialkylaminoalkyl (meth)acrylate
concentration ($[Da]$), and methyl chloride concentration ($[MC]$) in
the reaction mixture is maintained at 75 to 84 wt.% during
reaction.
- 30 5. A method according to any one of claims 1 to 4,
wherein the temperature of the reaction mixture is maintained at
30 to 80°C during reaction.
- 35 6. A method according to any one of claims 1 to 5,

wherein the reactor is a sealed reactor, and wherein the internal pressure of said reactor is maintained at 0.10 to 1 MPaG during reaction.

5 7. A method according to any one of claims 1 to 6, further comprising continuously supplying a dialkylaminoalkyl (meth)acrylate, methyl chloride, and water to the reactor, while continuously extracting a reaction mixture from said reactor.

10 8. A method according to any one of claims 1 to 7, wherein the dialkylaminoalkyl (meth)acrylate is dimethylaminoethyl (meth)acrylate.

 9. A method according to any one of claims 1 to 8,
15 wherein the methyl chloride is supplied using an ejector.

 10. A method according to claim 9, further comprising:
extracting a part of the reaction mixture from the
reactor;

20 cooling said reaction mixture by passing through a heat exchanger; and

 supplying said cooled reaction mixture, together with methyl chloride, to said reactor using an ejector.

25 11. A method according to any one of claims 1 to 10, further comprising:

 supplying the reaction mixture continuously extracted from the reactor to one or more sealed tanks; and then

 gradually adjusting the pressure of the reaction
30 mixture in said tank to atmospheric pressure.