

DESCRIPTION

PROCESS FOR PRODUCING METHACRYLIC POLYMER COMPOSITION

[0001]

5 This application claims priority to and the benefit of
Japanese Patent Application No. 2012-151147, filed July 5,
2012, the entire contents of which are incorporated herein
by reference.

10 Technical Field

[0002]

The present invention relates to a process for producing methacrylic polymer composition and a molded article obtainable by the process.

15

Background Art

[0003]

Polymers such as methacrylic ester based polymers are produced by continuous polymerization in which a raw material monomer, a polymerization initiator and so on are continuously supplied to a reactor to be polymerized. As such continuous polymerization processes, there are known a continuous solution polymerization process using a solvent (or a dispersion medium, which also applies hereinafter) to conduct continuous polymerization, and a continuous bulk

polymerization process using no solvent to conduct continuous polymerization.

[0004]

As a process for producing methacrylic polymer composition, for example, Patent Literature 1 and 2 disclose a process in which a raw material monomer mixed liquid and a polymerization initiator mixed liquid are supplied into a reactor of a complete mixing type so as to fully fill the reactor with liquid to exclude a gas phase part therefrom, and continuous bulk polymerization under an adiabatic condition is conducted with no heat transfer to or from the outside.

[0005]

In recent years, applications of resin compositions such as methacrylic ester based polymers has been expanded furthermore, a demand is increasing for more efficiently producing a polymer composition with high quality (for example, a polymer composition having superior properties such as heat resistance and thermal stability, and less immixed with impurities). However, it has been proved that the conventional processes for producing are not always sufficient to meet the demand.

Citation List

25 [0006]

Patent Literature

Patent Literature 1: JP H07-126308 A

Patent Literature 2: JP 2006-104282 A

5 Summary of Invention

[0007]

The purpose of the present invention is to provide a process for producing a polymer composition wherein the process can be conducted to more efficiently produce the
10 polymer composition suitable for producing a high thermally-stable and heat-resistant resin composition with high quality.

[0008]

The inventors have considered earnestly to achieve the
15 aforementioned purpose, and finally completed the present invention.

[0009]

The present invention provides the followings.

[1] A process for producing a methacrylic polymer
20 composition, which comprises

supplying a raw material mixture comprising a raw material monomer containing 50% by weight or more of methyl methacrylate and a polymerization initiator into a reactor of a complete mixing type through a supply port thereof;

25 subjecting to continuous polymerization in the

reactor; and

taking an obtained polymer composition from an effluent port thereof,

wherein a temperature of the raw material mixture supplied
5 into the reactor is -50°C to -10°C .

[2] The process for producing a methacrylic polymer composition according to the above [1], wherein the effluent port of the reactor is located at a top of the reactor.

10 [3] The process for producing a methacrylic polymer composition according to the above [1] or [2], wherein the continuous polymerization is conducted under an adiabatic condition.

[4] The process for producing a methacrylic polymer
15 composition according to any one of the above [1] to [3], wherein a polymerization temperature in the continuous polymerization is 120°C to 150°C .

[5] The process for producing a methacrylic polymer composition according to any one of the above [1] to [4],
20 wherein the continuous polymerization is continuous bulk polymerization.

[6] A molded article which is prepared by the process according to any one of the above [1]-[5].

[7] The molded article according to the above [6],
25 which is a light guide plate.

[0010]

According to the present polymerization process, a methacrylic polymer composition suitable for producing a high thermally-stable and heat-resistant methacrylic resin composition with high quality can be produced at high productivity.

Brief Description of Drawings

[0011]

Fig. 1 shows a schematic view of an apparatus for producing a polymer composition in one embodiment of the present invention.

[0012]

Following reference signs denote the following elements:

- 1 raw material monomer tank
- 3 polymerization initiator tank
- 4 raw material monomer supply line
- 5 monomer supplying means
- 6 polymerization initiator supply line
- 7 initiator supplying means
- 9 raw material supply line
- 10 reactor
- 11a supply port

11b effluent port
13 jacket
14 stirrer
15 effluent line
5 21 preheater
23 devolatilizing extruder
25 discharge line
27 recovery tank

10 Description of Embodiments

[0013]

A process for producing a polymer composition of the present invention is conducted using a reactor of a complete mixing type, and continuous polymerization such as
15 any of continuous bulk polymerization and continuous solution polymerization is conducted in the reactor.

Hereinafter, one embodiment of the present invention will be described in detail with reference to Fig. 1. First, an apparatus used for conducting the process for
20 producing a polymer composition according to the embodiment of the present invention will be explained.

[0014]

The process for producing a polymer composition according to the embodiment of the present invention is
25 conducted using a reactor of a complete mixing type 10.

The reactor 10 is used to conduct continuous bulk polymerization as continuous polymerization in this embodiment.

More specifically, the reactor 10 is provided with a supply port 11a and an effluent port 11b, and preferably further provided with a jacket 13 as a temperature regulating means for regulating a temperature of an outer surface of the reactor and a stirrer 14 for stirring contents therein. The effluent port 11b is located at a top of the reactor in this embodiment, but not limited thereto. On the other hand, the supply port 11a may be generally located at an appropriate position of a lower part of the reactor, although this embodiment is not limited thereto. This reactor 10 may be provided with a temperature sensor T as a temperature detecting means for detecting a temperature in the reactor.

[0015]

The stirrer 14 is a member for substantially attaining a complete mixing condition in the reactors. This stirrer may have any appropriate stirring blade(s), for example, may have blades of MIG impeller, MAXBLEND impeller (registered trademark, manufactured by Sumitomo Heavy Industries, Ltd.), paddle impeller, double helical ribbon impeller, FULLZONE impeller (registered trademark, manufactured by Kobelco Eco-Solutions Co., Ltd.) and so on.

In order to increase stirring effect in the reactor, it is preferable to provide the reactor with a baffle(s). However, this embodiment is not limited thereto, but may have any appropriate configuration in place of the stirrer
5 14 as long as a complete mixing condition can be substantially attained in the reactor.

[0016]

In general, the reactor 10 is more preferable when it has a higher stirring efficiency. However, in view of
10 avoiding the reactor from being added with an unnecessary amount of heat by the stirring operation, it is preferable that a power of stirring is not more than necessary. The power of stirring is not particularly limited, but preferably 0.5 to 30 kW/m³, and more preferably 1 to 15
15 kW/m³. As a viscosity of the reaction system becomes higher (or a content ratio of a polymer in the reaction system becomes higher), it is preferable to set the power of stirring at a larger level.

[0017]

20 As shown in the drawings, the supply port 11a of the reactor 10 is connected through a raw material monomer supply line (pipe) 4 and a raw material supply line 9 to a raw material monomer tank (a supply source of a raw material monomer) 1 and through a polymerization initiator
25 supply line (pipe) 6 and a raw material supply line 9 to a

polymerization initiator tank (a supply source of a polymerization initiator and, if necessary, of a raw material monomer) 3. The raw material monomer supply line 4 and the polymerization initiator supply line 6 are provided with a monomer supply means (pump 5) and a polymerization initiator supply means (pump 7), respectively. In this embodiment, the supply sources of the raw material monomer and the polymerization initiator to the reactor 10 are the raw material monomer tank 1 and the polymerization initiator tank 3, respectively. However, the number of the supply sources of the raw material monomer and the polymerization initiator, the forms of the raw material monomer and the polymerization initiator (in a case of a mixture, for example, a composition thereof) and so on are not particularly limited as long as the raw material monomer and the polymerization initiator can be supplied to the reactor 10, appropriately. The effluent port 11b of the reactor 10 is linked up to an effluent line 15.

[0018]

As the monomer supply means and the polymerization initiator supply means, for example, a pump 5 introducing a raw material monomer to the reactor 10 through the supply port 11a and a pump 7 introducing a polymerization initiator to the reactor 10 through the supply port 11a,

respectively. The pumps 5 and 7 are not particularly limited, but preferably pumps being able to set flow rates from the raw material monomer tanks 1 and the polymerization initiator tank 3 at constant values. More specifically, multiple reciprocating pumps are preferred, and more preferred are pulsation-free controlled-volume pumps such as a duplicate pulsation-free controlled-volume pump and a triplex pulsation-free controlled-volume pump. By using them, it is possible to control a supply amount (or a supply flow rate, which also applies hereinafter) of the raw material monomer and the polymerization initiator to the reactor 10.

[0019]

At least one selected from the group consisting of the raw material monomer tank 1, the polymerization initiator tank 3, the raw material monomer supply line 4, the polymerization initiator supply line 6 and the raw material supply line 9 (hereinafter referred to as a tank and/or line) is provided with a temperature regulating means. The raw material monomer tank 1 and/or the polymerization initiator tank 3 may be provided with, for example, a jacket covering at least partially an outer surface of a tank as the temperature regulating means, and such temperature regulating means can regulate a temperature of the raw material monomer in the raw material monomer tank 1

and/or the polymerization tank 3. In a case where the raw material monomer tank 1 and/or the polymerization initiator tank 3 are provided with the jacket, they are preferably further provided with a stirring and/or mixing means for stirring the raw material monomer and/or the polymerization initiator in the tank in view of more effective temperature regulation. Further, at least one selected from the group consisting of the raw material monomer supply line 4, the polymerization initiator supply line 6 and the raw material supply line 9 may be provided with, for example, a jacket covering at least partially an outer surface of a line, a heat medium bath being able to allow at least a part of a line soaked, a heater/cooler with which a part of a line is replaced, and/or a trace pipe through which a cooling medium passes (the line provided with the jacket is understood as a double pipe), as the temperature regulating means, and such temperature regulating means can regulate a temperature of at least one selected from the raw material monomer, the polymerization initiator and a mixture thereof flowing through the line. As the heater/cooler, a heater/cooler having both a heating/cooling means and a mixing means, more specifically, ones having a function of dynamic mixing (e.g. screw mixer being able to heat/cool its cylinder) and ones having a function of static mixing (e.g. heat exchanger with a built-in static mixer) may be

used. In a case where the at least one selected from the group consisting of the raw material monomer supply line 4, the polymerization initiator supply line 6 and the raw material supply line 9 may be provided with a heater/cooler, the heater/cooler is provided to the line in any appropriate configuration, and the line parts other than the heater/cooler may be covered with a lagging to retain heat, or a jacket surrounding an outer surface of the line may be used in combination for cooling. By the use of the temperature regulating means provided to the tank and/or line as in the above, the temperature of the raw material mixture comprising the raw material monomer and the polymerization initiator supplied to the reactor 10 can be regulated.

[0020]

It is preferable that each of the members described in the above with reference to Fig. 1 is appropriately connected to a control means described below (not shown in the drawings) and construct the whole so as to enable the control means to control their operations. Thereby, in order to make the temperature of the outer surface of the reactor set for the jacket (temperature regulating means) correspond to the temperature in the reactor detected by the temperature sensor (temperature detecting means) T with respect to the reactor 10 (in the other words, in order to

achieve an adiabatic condition in the reactor 10), the supply amounts of the raw material monomer and the polymerization initiator to the reactor 10 can be adjusted by the operation of the pumps 5 and 7, or the temperature of the outer surface of the reactor set for the jacket 13 can be regulated.

[0021]

The jacket 13 surrounds almost the whole of the reactor 10 to appropriately heat or retain the heat of the reactor 10 by introducing steam, hot water, organic heat medium or the like from a heat medium supply route (not shown in the drawings). The temperature of the jacket 13 is able to be appropriately regulated with a temperature or pressure of the heat medium to be introduced. The heat medium introduced into the jacket 13 is removed from a heat medium discharge route (not shown in the drawings). The temperature and/or pressure of the jacket 13 are detected by a sensor such as a temperature sensor (not shown in the drawings) located on the heat medium discharge route. The point of location of a sensor such as the temperature sensor is not particularly limited, but it may be located, for example, on the heat medium supply route, or in the jacket 13. A jacket provided to a tank and/or line as the temperature regulating means may have the same constitution as that of the jacket 13. Although the embodiment is not

limited thereto, at least one selected from the raw material monomer supply line 4, the polymerization initiator supply line 6 and the raw material supply line 9 may be typically a double pipe, in which the internal space of the inner pipe is a flow path of a raw material monomer, a polymerization initiator or a mixture thereof, the space between the inner pipe and the outer pipe is a flow path of a heat medium (jacket).

[0022]

For the polymerization reaction in the reactor 10, it is required to proceed at a generally constant temperature in the reactor 10 in view of obtaining a polymer with a constant quality. Therefore, the above described temperature regulating means (jacket 13) is controlled at a constant temperature which has been set beforehand, so that the temperature inside the reactor 10 can be maintained at a generally constant temperature.

[0023]

The setting temperature of the above described temperature regulating means (jacket 13) is transmitted to a control means described below, to be used as data for determining whether control of the supply flow rate with the monomer supply means (pump 5) and/or the initiator supply means (pump 7) is necessary or not. The setting temperature of the above described temperature regulating

means (jacket 13) can be regulated by controlling the temperature or pressure of the above described heat medium.

[0024]

Examples of the control means include, for example, a control unit (not shown in the drawings) provided with CPU, ROM, RAM and so on.

The ROM of the control means is a device for storing a program which controls the pumps 5 and 7. The RAM of the control means is a device for temporary storing data of the temperatures in the reactor 10 detected by the temperature sensor T, data of the setting temperatures of the jacket 13, and data of the setting temperature of the temperature regulating means provided to a tank and/or line in order to execute the above program.

The CPU of the control means executes the program stored in the ROM based on data such as the data of the temperatures in the reactor 10 and the data of the setting temperatures of the jacket 13 stored in the above RAM so that the supply flow rates of the raw material monomer and/or the polymerization initiator to the reactor 10 is controlled by the monomer supply means (pump 5) and/or the initiator supply means (pump 7). Specifically, with respect to the jacket and/or the heater/cooler provided to a tank and/or line as a temperature regulating means, the CPU of the control means executes the program stored in the

ROM (which may be either a part of the above program or other program than the above program) based on data such as the data of the temperature in the reactor 10 and the data of the setting temperature of the jacket and/or the heater/cooler provided to the tank and/or line stored in the above RAM, and in the case of actually measuring, the temperature in the tank and/or line to adjust the setting temperatures of the jacket and/or the heater/cooler provided to the tank and/or line.

10 [0025]

An example of the control by the control means (control unit) will be described below.

When the temperature in the reactor 10 detected by the temperature sensor T exceeds the setting temperature of the jacket 13 as the temperature regulating means, the CPU executes the program in the ROM to control, for example, the pump 7 so as to decrease the supply flow rate of the polymerization initiator into the reactor 10. By conducting such control, polymerization heat generated in the reactor 10 can be decreased, and thereby the temperatures in the reactor 10 can be lowered.

[0026]

On the other hand, when the temperature in the reactor 10 is below the setting temperature of the jacket 13, the CPU executes the program in the ROM to control, for example,

the pump 7 so as to increase the supply flow rate of the polymerization initiator into the reactor 10. By conducting such control, polymerization heat generated in the reactors 10 can be increased, and thereby the temperatures in the reactors 10 can be raised.

[0027]

For example, when the control over the pump 7 for the polymerization reaction in the reactor 10 results in remarkable decrease in the total supply flow rate into the reactor 10, it is preferable to not only control the pump 7 so as to decrease the supply flow rate of the polymerization initiator, but also to control the pump 5 so as to increase the supply flow rate of the raw material monomer at the same time.

Further, as another example of the control, the following control is noted. That is, when the temperature in the reactor 10 detected by the temperature sensor T exceeds the setting temperature of the jacket 13 as the temperature regulating means, the pump 5 is controlled to increase the supply flow rate of the raw material monomer, so that the relative supply flow rate of the polymerization initiator into the reactor 10 is decreased. By conducting such control, the temperature in the reactor 10 can also be lowered.

[0028]

A ratio of the supply flow rate of the raw material monomer and the supply flow rate of the polymerization initiator can be appropriately set depending on the kind of the polymer generated, the kind of the polymerization initiator used, and so on.

Also, degree of increase or decrease in the supply flow rate of the raw material monomer and/or the supply flow rate of the polymerization initiator can be appropriately set depending on the kind of the polymer generated, the kind of the polymerization initiator used, and so on. However, in a case what is supplied to the reactor 10 by the initiator supply means is not the polymerization initiator only, but the raw material monomer comprising the polymerization initiator, it is necessary to consider a content ratio of the polymerization initiator in the raw material monomer comprising polymerization initiator to control the supply flow rate of the polymerization initiator.

[0029]

The setting temperature of the jacket provided to the tank and/or line can be adjusted by controlling a flow rate and/or a temperature of a heat medium flowing in the jacket. The setting temperature of the heater/cooler provided to the tank and/or line, for example, when a heat exchanger is used as a heater/cooler (not shown in the drawings), can

generally be adjusted by controlling a flow rate and/or a temperature of a heat medium flowing in the heat exchanger. The tank and/or line may be optionally provided with a temperature sensor (a temperature detecting means) for detecting a temperature of the raw material monomer and/or the polymerization initiator stored in the tank and/or a temperature sensor (a temperature detecting means) for detecting a temperature of a fluid flowing through the line.

[0030]

Additionally, it is not necessary for this embodiment, but a preheater 21 and a devolatilizing extruder 23 may be located downstream of the effluent line 15. There may be a pressure adjusting valve (not shown in the drawings) provided between the preheater 21 and the devolatilizing extruder 23. An extruded object after devolatilization is discharged from a discharge line 25.

[0031]

As the preheater 21, any appropriate heater can be used as long as it is able to heat a viscous fluid. As the devolatilizing extruder 23, a single or multi screw devolatilizing extruder can be used.

[0032]

Further, there may be a recovery tank 27 for storing the raw material monomer which is separated and recovered from a volatile component (comprising unreacted raw

material, mainly) separated with the devolatilizing extruder 23.

[0033]

Next, a process for producing a polymer composition
5 conducted by using such apparatus will be described. In
this embodiment, a case of conducting continuous
polymerization of a methacrylic ester monomer, in the other
words, a case of producing a methacrylic ester based
polymer will be described as an example, although the
10 present invention is not limited thereto.

[0034]

- Preparation

At first, the raw material monomer, the polymerization
initiator and so on are prepared.

15 [0035]

As the raw material monomer, a raw material monomer
containing 50% by weight or more of methyl methacrylate is
used in this embodiment.

Examples of the raw material monomer are

- 20 - methyl methacrylate alone, or
- a mixture of not less than 50% by weight (preferably
not less than 70% by weight, more preferably not less than
90% by weight) of methyl methacrylate and not more than 50%
by weight (preferably not more than 30% by weight, more
25 preferably not more than 10% by weight) of other vinyl

monomer copolymerizable therewith (the sum of the amounts of methyl methacrylate and other vinyl monomer copolymerizable therewith is 100% by weight).

[0036]

5 Examples of copolymerizable vinyl monomer include monofunctional monomers having one double bond which is radical-polymerizable and multifunctional monomers having two or more double bonds which are radical-polymerizable. More specifically, examples of the monofunctional monomers
10 having one double bond which is radical-polymerizable include, for example, methacrylic esters such as ethyl methacrylate, n-propyl methacrylate, isopropyl methacrylate, n-butyl methacrylate, t-butyl methacrylate, sec-butyl methacrylate, isobutyl methacrylate, benzyl methacrylate,
15 and 2-ethylhexyl methacrylate; acrylic esters such as methyl acrylate, ethyl acrylate, propyl acrylate, butyl acrylate, and 2-ethylhexyl acrylate; unsaturated carboxylic acids or acid anhydrides thereof such as acrylic acid, methacrylic acid, maleic acid, itaconic acid, maleic acid
20 anhydride, and itaconic acid anhydride; hydroxy group-containing monomers such as 2-hydroxyethyl acrylate, 2-hydroxypropyl acrylate, monoglycerol acrylate, 2-hydroxyethyl methacrylate, 2-hydroxypropyl methacrylate, and monoglycerol methacrylate; nitrogen-containing monomers
25 such as acrylamide, methacrylamide, acrylonitrile,

methacrylonitrile, diacetoneacrylamide, and dimethylaminoethyl methacrylate; epoxy group-containing monomers such as allyl glycidyl ether, glycidyl acrylate, and glycidyl methacrylate; styrene based monomers such as styrene and α -methylstyrene. Examples of the multifunctional monomers having two or more double bonds which are radical-polymerizable include, for example, diesters of unsaturated carboxylic acids and glycols such as ethylene glycol dimethacrylate, and butane diol dimethacrylate; unsaturated carboxylic acid alkenyl esters such as allyl acrylate, allyl methacrylate, and allyl cinnamate; polybasic acid polyalkenyl esters such as diallyl phthalate, diallyl maleate, triallyl cyanurate, and triallyl isocyanurate; esters of unsaturated carboxylic acids and polyalcohols such as trimethylolpropane triacrylate; and divinylbenzene. The above described examples of copolymerizable vinyl monomer may be used alone or in combination of at least two of them.

[0037]

As the polymerization initiator, for example, a radical initiator is used in this embodiment.

Examples of the radical initiator include azo compounds such as azobisisobutyronitrile, azobisdimethylvaleronitrile, azobiscyclohexanenitrile, 1,1'-azobis(1-acetoxy-1-phenylethane), dimethyl 2,2'-

azobisisobutylate, and 4,4'-azobis-4-cyanovaleric acid;
organic peroxides such as benzoyl peroxide, lauroyl
peroxide, acetyl peroxide, caprylyl peroxide, 2,4-
dichlorobenzoyl peroxide, isobutyl peroxide, acetyl
5 cyclohexylsulfonyl peroxide, t-butyl peroxy-pivalate, t-
butyl peroxyneodecanoate, t-butyl peroxyneoheptanoate, t-
butyl peroxy-2-ethylhexanoate, 1,1-di(t-
butylperoxy)cyclohexane, 1,1-di(t-butylperoxy)-3,3,5-
trimethylcyclohexane, 1,1-di(t-hexylperoxy)-3,3,5-
10 trimethylcyclohexane, diisopropyl peroxydicarbonate,
diisobutyl peroxydicarbonate, di-sec-butyl
peroxydicarbonate, di-n-butyl peroxydicarbonate, bis(2-
ethylhexyl) peroxydicarbonate, bis(4-t-butylcyclohexyl)
peroxydicarbonate, t-amyl peroxy-2-ethylhexanoate, 1,1,3,3-
15 tetramethyl butyl peroxy-ethylhexanoate, 1,1,2-trimethyl
propyl peroxy-2-ethylhexanoate, t-butyl peroxy isopropyl
monocarbonate, t-amyl peroxy isopropyl monocarbonate, t-
butyl peroxy-2-ethylhexyl carbonate, t-butyl peroxy allyl
carbonate, t-butyl peroxy isopropyl carbonate, 1,1,3,3-
20 tetramethyl butyl peroxy isopropyl monocarbonate, 1,1,2-
trimethyl propyl peroxy isopropyl monocarbonate, 1,1,3,3-
tetramethyl butyl peroxy isononate, 1,1,2-trimethyl propyl
peroxy-isononate, and t-butyl peroxybenzoate.

These polymerization initiators may be used alone or
25 in combination of at least two of them.

[0038]

The polymerization initiator is selected depending on the kinds of the polymer to be produced and the raw material monomer used. For example, although the present invention is not particularly limited, as the polymerization initiator (radical initiator), those of which τ/θ (-) is, for example not more than 0.1, preferably not more than 0.02, more preferably not more than 0.01 can be used, wherein τ (second) represents a half-life of the polymerization initiator at the polymerization temperature, and θ (second) represents an average residence time in a reactor. When the value of τ/θ is not more than the above value, a polymerization reaction can be effectively initiated because the polymerization initiator is sufficiently decomposed (thus, generating a radical) in a reactor.

The supply amount of the polymerization initiator (radical initiator) is not particularly limited, but generally 0.001 to 1% by weight with respect to the raw material monomer (the raw material monomer eventually supplied to the reactor 10).

[0039]

In addition to the raw material monomer and the polymerization initiator described above, any appropriate other component(s), for example, a chain transfer agent, a

mold release agent, a rubbery polymer such as butadiene and styrene-butadiene rubber (SBR), a thermal stabilizing agent, and an ultraviolet absorbing agent may be used. The chain transfer agent is used for adjusting a molecular weight of a produced polymer. The mold release agent is used for improving moldability of a resin composition obtained from the polymer composition. The thermal stabilizing agent is used for preventing a produced polymer from thermal degradation. The ultraviolet absorbing agent is used for preventing a produced polymer from being degraded by ultraviolet rays.

[0040]

As to the chain transfer agent, either monofunctional or polyfunctional chain transfer agent can be used. More specifically, examples thereof include alkyl mercaptans such as n-propyl mercaptan, isopropyl mercaptan, n-butyl mercaptan, t-butyl mercaptan, n-hexyl mercaptan, n-octyl mercaptan, 2-ethylhexyl mercaptan, n-dodecyl mercaptan, and t-dodecyl mercaptan; aromatic mercaptans such as phenyl mercaptan and thiocresol; mercaptans having 18 or less carbons such as ethylene thioglycol; polyalcohols such as ethylene glycol, neopentyl glycol, trimethylolpropane, pentaerythritol, dipentaerythritol, tripentaerythritol, and sorbitol; those of which hydroxyl group is esterified with thioglycolic acid or 3-mercaptopropionic acid, 1,4-

dihydronaphthalene, 1,4,5,8-tetrahydronaphthalene, β -terpinene, terpinolene, 1,4-cyclohexadiene, hydrogen sulfide and so on. These may be used alone or in combination of at least two of them.

5 [0041]

The supply amount of the chain transfer agent is not particularly limited since it varies depending on the kind of the chain transfer agent used and so on. For example, in a case of using mercaptans, it is preferably 0.01 to 3%
10 by weight, and more preferably 0.05 to 1% by weight with respect to the raw material monomer (the raw material monomer eventually supplied to the reactor 10).

[0042]

Examples of the mold release agents are not
15 particularly limited, but include esters of higher fatty acids, higher fatty alcohols, higher fatty acids, higher fatty acid amides, metal salts of higher fatty acids and so on. As the mold release agent, only one kind or two or more kinds thereof may be used.

20 [0043]

Examples of the esters of higher fatty acids specifically include, for example, saturated fatty acid alkyl esters such as methyl laurate, ethyl laurate, propyl laurate, butyl laurate, octyl laurate, methyl palmitate,
25 ethyl palmitate, propyl palmitate, butyl palmitate, octyl

palmitate, methyl stearate, ethyl stearate, propyl stearate, butyl stearate, octyl stearate, stearyl stearate, myristyl myristate, methyl behenate, ethyl behenate, propyl behenate, butyl behenate, octyl behenate; unsaturated fatty acid alkyl esters such as methyl oleate, ethyl oleate, propyl oleate, butyl oleate, octyl oleate, methyl linoleate, ethyl linoleate, propyl linoleate, butyl linoleate, octyl linoleate; saturated fatty acid glycerides such as lauric monoglyceride, lauric diglyceride, lauric triglyceride, palmitic monoglyceride, palmitic diglyceride, palmitic triglyceride, stearic monoglyceride, stearic diglyceride, stearic triglyceride, behenic monoglyceride, behenic diglyceride, behenic triglyceride; unsaturated fatty acid glycerides such as oleic monoglyceride, oleic diglyceride, oleic triglyceride, linolic monoglyceride, linolic diglyceride, linolic triglyceride. Among them, methyl stearate, ethyl stearate, butyl stearate, octyl stearate, stearic monoglyceride, stearic diglyceride, stearic triglyceride, and son on are preferred.

[0044]

Examples of the higher fatty alcohols specifically include, for example, saturated fatty (or aliphatic) alcohols such as lauryl alcohol, palmityl alcohol, stearyl alcohol, isostearyl alcohol, behenyl alcohol, myristyl alcohol, cetyl alcohol; unsaturated fatty (or aliphatic)

alcohols such as oleyl alcohol, linolyl alcohol. Among them, stearyl alcohol is preferred.

[0045]

Examples of the higher fatty acids specifically
5 include, for example, saturated fatty acids such as caproic acid, caprylic acid, capric acid, lauric acid, myristic acid, palmitic acid, stearic acid, arachidic acid, behenic acid, lignoceric acid, 12-hydroxyoctadecanoic acid; unsaturated fatty acids such as palmitoleic acid, oleic
10 acid, linoleic acid, linolenic acid, cetoleic acid, erucic acid, ricinoleic acid.

[0046]

Examples of the higher fatty acid amides specifically include, for example, saturated fatty acid amides such as
15 lauric acid amide, palmitic acid amide, stearic acid amide, behenic acid amide; unsaturated fatty acid amides such as oleic acid amide, linoleic acid amide, erucic acid amide; amides such as ethylene-bis-lauric acid amide, ethylene-bis-palmitic acid amide, ethylene-bis-stearic acid amide,
20 N-oleyl stearamide. Among them, stearic acid amide and ethylene-bis-stearic acid amide are preferred.

[0047]

Examples of the metal salts of higher fatty acids include, for example, sodium salts, potassium salts,
25 calcium salts and barium salts of the above-described

higher fatty acids, and so on.

[0048]

A used amount of the mold release agent is preferably adjusted in a range from 0.01 to 1.0 part by weight, and
5 more preferably adjusted in a range from 0.01 to 0.50 part by weight, with respect to 100 parts by weight of a polymer contained in a polymer composition to be obtained.

[0049]

Examples of the thermal stabilizing agent are not
10 particularly limited, but include, for example, phosphorous-based thermal stabilizing agent and organic disulfide compounds. Among them, the organic disulfide compounds are preferable. As the thermal stabilizing agent, only one kind or two or more kinds thereof may be used.

15 [0050]

Examples of the phosphorus-based thermal stabilizing agent include, for example, tris(2,4-di-*t*-butylphenyl)phosphite, 2-[[2,4,8,10-tetrakis(1,1-dimethylethyl)dibenzo[d,f][1,3,2]dioxaphosphepine-6-yl]oxy]-*N,N*-bis[2-[[2,4,8,10-tetrakis(1,1-dimethylethyl)dibenzo[d,f][1,3,2]dioxaphosphepine-6-yl]oxy]-ethyl]ethanamine, diphenyl tridecyl phosphite, triphenyl phosphite, 2,2-methylenebis(4,6-di-*tert*-butylphenyl)octylphosphite, bis(2,6-di-*tert*-butyl-4-methylphenyl)pentaerythritol diphosphite, and so on. Among
25

them, 2,2-methylenebis(4,6-di-tert-butylphenyl)octylphosphite is preferred.

[0051]

Examples of the organic disulfide compounds include,
5 for example, dimethyl disulfide, diethyl disulfide, di-n-propyl disulfide, di-n-butyl disulfide, di-sec-butyl disulfide, di-tert-butyl disulfide, di-tert-amyl disulfide, dicyclohexyl disulfide, di-tert-octyl disulfide, di-n-dodecyl disulfide, di-tert-dodecyl disulfide, and so on.
10 Among them, di-tert-alkyl disulfide is preferred, and di-tert-dodecyl disulfide is more preferred.

[0052]

A used amount of the thermal stabilizing agent is preferably 1 to 2,000 ppm by weight with respect to a
15 polymer contained in a polymer composition to be obtained. On molding a polymer composition (more specifically, a resin composition after devolatilization) to prepare a molded article from the polymer composition of the present invention, a molding temperature is set at a higher
20 temperature for the purpose of improving its molding processability in some cases. Use of the thermal stabilizing agent is effective for such case.

[0053]

As the kinds of the ultraviolet absorbing agent, a
25 benzophenone-based ultraviolet absorbing agent, a

cyanoacrylate-based ultraviolet absorbing agent, a benzotriazole-based ultraviolet absorbing agent, a malonic ester-based ultraviolet absorbing agent, an oxalic anilide-based ultraviolet absorbing agent and so on are exemplified.

5 These ultraviolet absorbing agents may be used alone or in combination of at least two of them. Among them, the benzotriazole-based ultraviolet absorbing agent, the malonic ester-based ultraviolet absorbing agent, and the oxalic anilide-based ultraviolet absorbing agent are
10 preferable.

[0054]

Examples of the benzophenone-based ultraviolet absorbing agent include, for example, 2,4-dihydroxybenzophenone, 2-hydroxy-4-methoxybenzophenone, 2-
15 hydroxy-4-methoxybenzophenone-5-sulfonic acid, 2-hydroxy-4-octyloxybenzophenone, 4-dodecyloxy-2-hydroxybenzophenone, 4-benzyloxy-2-hydroxybenzophenone, 2,2'-dihydroxy-4,4'-dimethoxybenzophenone, and so on.

[0055]

20 Examples of the cyanoacrylate-based ultraviolet absorbing agent include, for example, ethyl 2-cyano-3,3-diphenylacrylate, 2-ethylhexyl 2-cyano-3,3-diphenylacrylate, and so on.

[0056]

25 Examples of the benzotriazole-based ultraviolet

absorbing agent include, for example, 2-(2-hydroxy-5-methylphenyl)-2H-benzotriazole, 5-chloro-2-(3,5-di-tert-butyl-2-hydroxyphenyl)-2H-benzotriazole, 2-(3-tert-butyl-2-hydroxy-5-methylphenyl)-5-chloro-2H-benzotriazole, 2-(3,5-
5 di-tert-pentyl-2-hydroxyphenyl)-2H-benzotriazole, 2-(3,5-di-tert-butyl-2-hydroxyphenyl)-2H-benzotriazole, 2-(2H-benzotriazol-2-yl)-4-methyl-6-(3,4,5,6-tetrahydrophthalimidylmethyl)phenol, 2-(2-hydroxy-5-tert-octylphenyl)-2H-benzotriazole, and so on.

10 [0057]

As to the malonic ester-based ultraviolet absorbing agent, 2-(1-aryl alkylidene)malonates are generally used, and examples thereof include dimethyl 2-(p-methoxybenzylidene)malonate and so on.

15 [0058]

As to the oxalic anilide-based ultraviolet absorbing agent, 2-alkoxy-2'-alkyloxalic anilides are generally used, and examples thereof include 2-ethoxy-2'-ethyloxalic anilide and so on.

20 [0059]

A used amount of the ultraviolet absorbing agent is preferably 5 to 1,000 ppm by weight with respect to a polymer contained in a polymer composition to be obtained.

[0060]

25 In the raw material monomer tank 1, the raw material

monomer (methyl methacrylate alone or a mixture of methyl methacrylate and other vinyl monomer copolymerizable therewith) as described above is appropriately prepared (together with other component(s) such as the chain transfer agent as the case may be). In the polymerization initiator tank 3, the polymerization initiator as described above is appropriately prepared with the raw material monomer if necessary (together with other component(s) such as the chain transfer agent as the case may be). The polymerization initiator tank 3 may store the polymerization initiator alone or in the form of the mixture of the raw material monomer and the polymerization initiator (may further comprise other component(s) such as the chain transfer agent as the case may be).

[0061]

• Polymerization Step

The raw material monomer and the polymerization initiator are continuously supplied to the reactor 10 through the supply port 11a from the raw material monomer tank 1 and the polymerization initiator tank 3 as the supply source(s) of the raw material monomer and the polymerization initiator. More specifically, the raw material monomer is continuously supplied from the raw material monomer tank 1 by the pump 5, and the polymerization initiator (preferably, the mixture of the

raw material monomer and the polymerization initiator, which is also simply referred to as the polymerization initiator herein) is supplied from the polymerization initiator tank 3 by the pump 7, and they merge together through the raw material supply line 9 into the reactor 10 via the supply port 11a. In the present invention, the raw material mixture comprising the raw material monomer and the polymerization initiator supplied to the reactor 10 is a mixture comprising the raw material monomer, the polymerization initiator, and other component(s) such as the chain transfer agent, as the case may be, eventually supplied to the reactor 10.

[0062]

For supplying the polymerization initiator to the reactor 10, when the mixture of the raw material monomer and the polymerization initiator is prepared in the polymerization initiator tank 3 and supplied therefrom, it is preferable to adjust a ratio A:B in a range from 80:20 to 98:2 wherein A represents the supply flow rate (kg/h) of the raw material monomer from the raw material monomer tank 1, and B represents the supply flow rate (kg/h) of the mixture of the raw material monomer and the polymerization initiator (of which content ratio of the polymerization initiator is 0.002 to 10% by weight) from the polymerization initiator tank 3.

[0063]

In the process of the present invention, the temperature of the raw material mixture comprising the raw material monomer and the polymerization supplied to the reactor 10 is adjusted to -50 to -10°C. The temperature is preferably -40 to -15°C. When the temperature is lower than -50°C, a moisture in the raw material monomer or the polymerization initiator and other additives may precipitate. When the temperature is higher than -10°C, the polymerization temperature should be raised to increase a polymerization rate and, thereby, thermal stability and heat resistance of the finally obtained resin composition may decrease. Further, when the temperature is higher than -10°C and the polymerization temperature is lowered, the reactor should be cooled with a jacket and so on. In this case, gelation may occur due to local cooling to lessen the quality of the finally obtained resin composition.

[0064]

The temperature of the raw material mixture comprising the raw material monomer and the polymerization supplied to the reactor 10 may be adjusted, as described in the above, by the temperature regulating means provided to a tank and/or line. A process for supplying the raw material mixture at -50 to -10°C to the reactor 10 includes, more

specifically, (I) a process which comprises adjusting the respective temperatures of the raw material monomer stored in the raw material monomer tank 1 and the polymerization initiator stored in the polymerization initiator tank 3 are set to -50 to -10°C, maintain the temperatures of the raw material monomer supply line 4, the polymerization initiator supply line 6 and the raw material supply line 9 by a temperature regulating means and/or a lagging, and supplying the raw material mixture comprising the raw material monomer and the polymerization initiator to the reactor 10; (II) a process which comprises regulating the respective temperatures of the raw material monomer stored in the raw material monomer tank 1 and the polymerization initiator stored in the polymerization initiator tank 3, and the respective supply flow rates of the raw material monomer and the polymerization initiator so that the temperature of the raw material mixture at the supply port 11a of the reactor 10 becomes -50 to -10°C; (III) a process which comprises controlling the setting temperature of the temperature regulating means provided to at least one selected from the group consisting of the raw material monomer supply line 4, the polymerization initiator supply line 6 and the raw material supply line 9 so that the temperature of the raw material mixture at the supply port 11a of the reactor 10 becomes -50 to -10°C; (IV) a process

which comprises regulating the respective temperatures of the raw material monomer stored in the raw material monomer tank 1 and the polymerization initiator stored in the polymerization initiator tank 3, and the respective supply flow rates of the raw material monomer and the polymerization initiator or controlling the setting temperature of the temperature regulating means provided to at least one selected from the group consisting of the raw material monomer supply line 4, the polymerization initiator supply line 6 and the raw material supply line 9 so that the temperature of the raw material mixture at the supply port 11a of the reactor 10 becomes -50 to -10°C. In the processes according to the above (II), (III) and (IV), it is preferable that the temperature of the raw material mixture is actually measured in the vicinity of the supply port 11a of the reactor 10 by the temperature detecting means detecting the temperature in the raw material supply line 9.

[0065]

The raw material mixture comprising the raw material monomer and the polymerization initiator supplied to the reactor 10 as described in the above are subjected to continuous polymerization, continuous bulk polymerization in this embodiment (in other words, polymerization with no solvent). This polymerization step has only to proceed the

polymerization reaction partway, and a polymer composition (or polymerization syrup) is continuously taken from the effluent port 11b of the reactor 10.

[0066]

5 The continuous polymerization can be conducted under a condition in which the reactor is filled with the reaction mixture while substantially no gas phase is present (hereinafter referred to as a fully filled condition). This is especially suitable for the continuous bulk
10 polymerization. The fully filled condition can prevent beforehand the problems such as that gel adheres to and grows on the inner surface of the reactor, and that this gel is immixed into the reaction mixture to degrade quality of a polymer composition obtained in the end. Further, the
15 fully filled condition enables all of the inner volume of the reactor to be used as a reaction space, and thereby a high productivity can be attained.

[0067]

By locating the effluent port 11b of the reactor 10 at
20 the reactor's top as in this embodiment, the fully filled condition is conveniently realized simply by conducting the supply to and the taking from the reactor 10, continuously. It is especially suitable for continuous polymerization of a methacrylic ester based monomer that the effluent port is
25 located at the reactor's top.

[0068]

Further, the continuous polymerization may be conducted under an adiabatic condition (condition with substantially no heat transfer to or from outside of the reactor). This is especially suitable for the continuous bulk polymerization. The adiabatic condition can prevent beforehand the problems such as that gel adheres to and grows on the inner surface of the reactor, and that this gel is immixed into the reaction mixture to degrade quality of a polymer composition obtained in the end. Further, the adiabatic condition enables the polymerization reaction to become stable, and self regulating characteristics for suppressing a runaway reaction can be brought about.

[0069]

The adiabatic condition can be realized by making the temperature of the inside of the reactor 10 and the temperature of the outer surface thereof generally equal to each other. More specifically, this can be realized, with the use of the above described control means (not shown in the drawings), by adjusting the supply amounts of the raw material monomer and the polymerization initiator to the reactor 10 with operating the pumps 5 and 7 such that the temperature of the outer surface of the reactor 10 set for the jacket (temperature regulating means) 13 and the temperature in the reactor 10 detected by the temperature

sensor (temperature detecting means) T correspond to each other. It is not preferable to set the temperature of the outer surface of the reactor much higher than the temperature in the reactor since it adds extra amount of heat into the reactor. The smaller the difference between the temperature in the reactor and the temperature of the outer surface of the reactor is, the better it is. More specifically, it is preferable to adjust the temperature difference within the range of $\pm 5^{\circ}\text{C}$.

10 [0070]

The heat generated in the reactor 10 such as polymerization heat and stirring heat is generally carried away on taking the polymer composition from the reactor 10. The amount of the heat carried away by the polymer composition is determined by the flow rate and the specific heat of the polymer composition, and the temperature of the polymerization reaction.

[0071]

The temperature for the continuous polymerization is understood as the temperature in the reactor 10 (detected by the temperature sensor T). The continuous polymerization is conducted, for example, at a temperature in the range of 120 to 150°C , more preferably at a temperature in the range of 130 to 150°C . It is noted, however, that the temperature in the reactor may change

according to various conditions until it reaches a static state.

[0072]

The pressure for the continuous polymerization is understood as the pressure in the reactor 10. This pressure is a pressure not less than a vapor pressure of the raw material monomer at the temperature in the reactor to prevent gas of the raw material monomer from generating in the reactor, and is generally about 1.0 to 2.0 MPa in gauge pressure.

[0073]

A time period subjected to the continuous polymerization is understood as an average residence time in the reactor 10. The average residence time in the reactor 10 can be set according to the productivity of the polymer in the polymer composition and so on, and is not particularly limited, but, for example, from 15 minutes to 6 hours. The average residence time in the reactor 10 can be adjusted by using the pumps 5 and 7 to change the supply amount (supply flow rate) of the raw material monomer or the like to the reactor 10. However, the average residence time depends in a large part on the inner volume of the reactor 10.

[0074]

As described in the above, the polymer composition is

continuously taken from the effluent port 11b of the reactor 10. The obtained polymer composition comprises the generated polymer and the unreacted raw material monomer, and may further comprise the unreacted polymerization initiator, decomposed substances of the polymerization initiator, and so on.

[0075]

Although this embodiment is not limited thereto, the polymerization rate in the polymer composition is, for example, 30 to 90% by weight. The polymerization rate in the polymer composition generally corresponds to the content ratio of the polymer in the polymer composition. A higher polymerization rate results in higher productivity of the polymer but the viscosity of the polymer composition becomes high and, thereby strong power of stirring is needed. A lower polymerization rate results in lower productivity of the polymer and, thereby larger efforts are needed to recover unreacted raw material monomers. Thus, it is preferable to set an appropriate polymerization rate as a target or a criterion.

[0076]

In general, the following tendency is observed: the higher the polymerization temperature, the lower the syndiotacticity of the obtained polymer, the lower the heat resistance of a resin composition obtained in the end.

Therefore, it is preferable to conduct a polymerization at a low temperature to obtain a resin composition having high heat resistance. However, if continuous polymerization is conducted at lower temperatures with the use of the conventional continuous polymerization apparatus (Patent Literatures 1 and 2), a long time is required to achieve the desired polymerization rate. Therefore, it requires a larger reactor, furthermore larger space to realize a longer average residence time, so that it is not efficient. In addition, when the average residence time is longer than necessary, the generation of the oligomer such as dimer and trimer is increased, thereby heat resistance of the resin composition obtained from the polymer composition may be decreased.

[0077]

In addition, the amount of the polymerization initiator can be set depending on other factors such as a polymerization temperature, a desired polymerization rate, and an average residence time, and so on. The lower the polymerization temperature or the shorter the average residence time, the larger the amount of the polymerization initiator required for achieving the desired polymerization rate. However, the larger the amount of the polymerization initiator, the larger the remained amount of the terminal part which consists of a unstable unsaturated bond and at

which polymerization is stopped (terminal polymer) in the polymer composition, as a result, the thermal stability of the finally obtained resin composition tends to be decreased. Also, the much higher the polymerization temperature, the larger the generated amount of the terminal part which consists of an unsaturated bond derived from the polymerization initiator and at which polymerization is stopped (terminal polymer) in the polymer composition, as a result, the thermal stability of the finally obtained resin composition tends to be decreased.

[0078]

Especially when continuous polymerization is conducted under an adiabatic condition, since substantially no heat is transferred to or from the outside of the reactor, the polymerization temperature necessarily increases with heat generation due to polymerization. Thus, in continuous polymerization under an adiabatic condition, the polymerization rate is determined based on the temperature difference between a supply temperature of the raw material mixture supplied to the reactor and the polymerization temperature and, as a result, the temperature difference becomes larger, the polymerization rate becomes higher. Therefore, when the supply temperature of the raw material mixture supplied to the reactor is higher than -10°C , the polymerization temperature should be raised in order to

obtain the desired polymerization rate and, thereby, thermal stability and heat resistance of the finally obtained resin composition tend to be decreased.

[0079]

5 According to this embodiment, in order to achieve the desired polymerization rate and low polymerization temperature in the reactor 10, simultaneously, the control of the temperature regulating means provided to a tank and/or line are conducted so that the temperature of the
10 raw material mixture comprising the raw material monomer and the polymerization initiator supplied to the reactor 10 becomes -10 to -50°C, thereby it is possible to produce the polymer composition having superior thermal stability and heat resistance with high productivity.

15 [0080]

• Devolatilization Step

As described in the above, the polymer composition (polymerization syrup) taken from the effluent port 11b of the reactor 10 may comprise the unreacted raw material
20 monomer and polymerization initiator and so on, in addition to the generated polymer. Although this embodiment is not limited thereto, such polymer composition is preferably subjected to, for example, devolatilization to separate and recover the raw material monomer.

25 [0081]

More specifically, the polymer composition is transferred to the preheater 21 through the effluent line 15. The polymer composition in the preheater 21 is added with a part or all of an amount of heat necessary to volatilize the volatile component which is mainly composed of the unreacted raw material monomer. Then, the polymer composition is transferred to the devolatilizing extruder 23 via the pressure adjusting valve (not shown in the drawings), and the volatile component is at least partially removed in the devolatilizing extruder, and a residual extruded object is formed into pellets and discharged from the discharge line 25. Thereby, the resin composition comprising a methacrylic ester based polymer is produced in the form of the pellets.

[0082]

As a method for transferring the above polymer composition, a method described in JP H04-48802 B is preferable. As a method of using a devolatilizing extruder, methods described in, for example, JP H03-49925 A, JP S51-29914 B, JP S52-17555 B, JP H01-53682 B, JP S62-89710 A and so on are preferable.

[0083]

Further, during or after devolatilization of the polymer composition in the devolatilizing extruder described above, the polymer composition or the extruded

object can be added with a mold release agent such as higher alcohols and higher fatty acid esters, an ultraviolet absorbing agent, a thermal stabilizing agent, a colorant, an antistatic agent and so on, in order to
5 incorporate them into the resin composition, if necessary.

[0084]

The volatile component removed in the devolatilizing extruder 23 consists primarily of the unreacted raw material monomer and includes impurities; e.g., impurities
10 originally contained in the raw material monomer, additives used if necessary, volatile by-product(s) generated in the process of polymerization, oligomer such as dimer and trimer, decomposed substances of the polymerization initiator, and so on. In general, a larger amount of the
15 impurities make the obtained resin composition colored, which is not preferable. Then, the volatile component removed in the devolatilizing extruder 23 (which consists primarily of the unreacted raw material monomer and includes impurities as described above) may be passed
20 through a monomer recovery column (not shown in the drawings), and treated by means of distillation, adsorption and so on in the monomer recovery column to remove the impurities from the above described volatile component. Thereby, the unreacted raw material monomer can be
25 recovered with high purity, so that it can be suitably

reused as the raw material monomer for polymerization. For example, continuous distillation is conducted in the monomer recovery column to recover the unreacted raw material monomer with high purity as a distillate liquid from the top of the monomer recovery column, and it may be transferred and recycled to the raw material monomer tank 1 after it is reserved in the recovery tank 27 once, or it may be transferred and recycled to the raw material monomer tank 1 without being reserved in the recovery tank 27. On the other hand, the impurities removed in the monomer recovery column may be disposed as a waste.

[0085]

In order to prevent the recovered raw material monomer from causing the polymerization reaction in the recovery tank 27 and/or the raw material monomer tank 1, it is preferable that a polymerization inhibitor exists in the recovery tank 27 or the raw material monomer tank 1 at a ratio of, for example, 2 to 8 ppm by weight with respect to the raw material monomer, and more preferably, in addition to this, an oxygen concentration in a gas phase in the recovery tank 27 or the raw material monomer tank 1 is set at 2 to 8% by volume. If the recovered raw material monomer is wanted to be preserved in the recovery tank 27 for a long time, it is preferable to reserve it at a low temperature of, for example, 0 to 5°C.

[0086]

In this embodiment, the continuous bulk polymerization apparatus used to conduct the continuous bulk polymerization is described. However, the continuous polymerization apparatus of the present invention is not limited thereto, and may be used to conduct continuous solution polymerization. In such embodiment, since a solvent is used for the solution polymerization, the continuous polymerization apparatus is provided, in addition to a similar configuration to the continuous polymerization apparatus described in the above with reference to Fig. 1, with a solvent tank and a supply line and a pump (supply means) associated with the solvent tank to supply the solvent to a certain reactor for conducting the solution polymerization. The solvent tank and the supply line and the pump (supply means) associated with the solvent tank are not particularly limited, those similar to conventionally used ones can be used. The solvent can be supplied to the reactor for conducting the solution polymerization after being mixed with the raw material monomer and/or the polymerization initiator so as to make the temperature of the raw material mixture comprising the raw material monomer, the polymerization initiator and the solvent -50 to -10°C. The continuous solution polymerization is conducted similarly to the polymerization

step described in the above with reference to Fig. 1, except that the solvent is used in the polymerization reaction. As to the solvent, it is appropriately selected according to the raw material monomer of the solution polymerization reaction and so on, and not particularly limited, but examples thereof include toluene, xylene, ethyl benzene, methyl isobutyl ketone, methyl alcohol, ethyl alcohol, octane, decane, cyclohexane, decalin, butyl acetate, pentyl acetate, and so on. A ratio C:D is, for example, 70:30 to 95:5, and preferably 80:20 to 90:10, but not limited thereto, wherein C represents a supply flow rate (kg/h) of the raw material monomer to the reactor, and D represents a supply flow rate (kg/h) of the solvent to this certain reactor.

[0087]

The process for producing the polymer composition of the present invention is hereinbefore described through the embodiment of the present invention in detail. According to the present invention, a continuous polymerization is conducted in which the temperature of the raw material mixture comprising the raw material monomer and the polymerization initiator supplied to the reactor 10 is set to -50 to -10°C, thereby it becomes possible to control the syndiotacticity of the polymer contained in the finally obtained resin composition to more efficiently produce the

polymer composition suitable for obtaining a resin composition having high heat resistance and thermal stability.

[0088]

5 The present invention is not limited to the above embodiment, and various modifications can be made. For example, two or more reactors can be used to conduct the polymerization in two or more stages in series. Further, the process for producing the polymer composition of the
10 present invention is continuously conducted preferably by using the continuous polymerization apparatus of the present invention, but it may be conducted in a batch method.

[0089]

15 The polymer composition produced by the process of the present invention is preferably used as a material for a molded article, and the molded article obtained therefrom has an advantage of having high heat resistance and thermal stability. For example, the polymer composition produced
20 by the process of the present invention (more specifically, the resin composition after devolatilization) is molded alone or together with any suitable other component(s) according to any molding process such as injection molding and extrusion molding to prepare a molded article. The
25 polymer composition produced by the process of the present

invention is preferably used for preparing a molded article by injection molding, and it is possible to prepare a molded article with good moldability and prevent silver streaks from occurring. Especially, since the resin composition comprising a methacrylic ester based polymer has a superior transparency, the molded article prepared from it by injection molding has high transparency and less occurrence of silver streaks and good moldability, and therefore it is preferably utilized as a material for a light guide plate, which is used as a member of a backlight unit for various types of liquid crystal displays, or for vehicle members such as a rear lamp cover, a head lamp cover, a visor, a meter panel, and so on. In particular, it is preferably used as a light guide plate.

[0090]

Injection molding can be conducted by filling (injecting into) a mold having a certain thickness with at least the polymer composition produced by the process of the present invention in a molten state, followed by cooling, and then thus molded article is released from the mold. More specifically, the molded article can be prepared by, for example, supplying a molding machine from a hopper with the polymer composition produced by the process of the present invention (more specifically, the resin composition after devolatilization) alone or in

combination with any other suitable components, retracting and rotating a screw to measure the resin composition in a cylinder of the molding machine, melting the resin composition in the cylinder, filling a mold (e.g., metal mold) with the molten resin composition under pressure, holding the pressure for a certain time period until the mold is sufficiently cooled, opening the mold to eject the molded article therefrom.

[0091]

Thus, according to another aspect of the present invention, there is also provided a molded article prepared from the polymer composition produced by the process of the present invention. It is noted that conditions for preparing the molded article of the present invention from the polymer composition (for example, in a case of injection molding, a temperature for melting a molding material, a temperature of a mold to which the molding material is injected, a pressure to be held after the mold is filled with the molding material, and so on) can be appropriately set and are not specifically limited.

Industrial Applicability

[0092]

The process of the present invention can produce an methacrylic polymer composition suitable for obtaining

resin compositions with high quality demanded in a wide variety of applications (for example, a polymer composition having superior properties such as heat resistance and thermal stability, and less immixed with impurities).

CLAIMS

1. A process for producing a methacrylic polymer composition, which comprises

supplying a raw material mixture comprising a raw
5 material monomer containing 50% by weight or more of methyl methacrylate and a polymerization initiator into a reactor of a complete mixing type through a supply port thereof;

subjecting to continuous polymerization in the reactor; and

10 taking an obtained polymer composition from an effluent port thereof,

wherein a temperature of the raw material mixture supplied into the reactor is -50°C to -10°C .

2. The process for producing a methacrylic polymer
15 composition according to claim 1, wherein the effluent port of the reactor is located at a top of the reactor.

3. The process for producing a methacrylic polymer composition according to claim 1 or 2, wherein the continuous polymerization is conducted under an adiabatic
20 condition.

4. The process for producing a methacrylic polymer composition according to any one of claims 1 to 3, wherein a polymerization temperature in the continuous polymerization is 120°C to 150°C .

25 5. The process for producing a methacrylic polymer

composition according to any one of claims 1 to 4, wherein the continuous polymerization is continuous bulk polymerization.

6. A molded article which is prepared by the process
5 according to any one of claims 1 to 5.

7. The molded article according to claim 6, which is a light guide plate.