

DESCRIPTION

TITLE OF THE INVENTION:

CATALYST MOLDED ARTICLE, CATALYST MOLDED ARTICLE FOR PRODUCING METHACROLEIN AND/OR METHACRYLIC ACID, AND METHOD FOR PRODUCING METHACROLEIN AND/OR METHACRYLIC ACID

TECHNICAL FIELD

[0001]

The present invention relates to a catalyst molded body, a catalyst molded body for producing methacrolein and/or methacrylic acid, and a method for producing methacrolein and/or methacrylic acid.

BACKGROUND ART

[0002]

Conventionally, it has been known to produce methacrolein and/or methacrylic acid by a gas phase catalytic oxidation of isobutylene, tert-butyl alcohol, methyl-tert-butyl ether, or the like with molecular oxygen in the presence of a catalyst, and a catalyst having various compositions and shapes has been proposed as the catalyst, and further, a variety of catalyst production methods have been proposed.

[0003]

For example, Patent Document 1 proposes an extrusion-molded catalyst including at least molybdenum, bismuth, and iron, wherein a β -1,3-glucan and a liquid are added and kneaded. Patent Document 2 proposes a hollow cylindrical shaped catalyst body in which the ratio of an internal diameter and an external diameter of the cylindrical shape is set to a specific value. Patent Document 3 proposes a catalyst in which the cumulative pore volume and the volume of macropores, the specific surface area and the filling bulk density are adjusted. In addition, Patent Documents 4 and 5 propose that a specific step is added at the time of catalyst preparation.

PRIOR-ART DOCUMENT

PATENT DOCUMENT

[0004]

[Patent Document 1] JP 2002-282695 A

[Patent Document 2] JP 2016-538120 A

[Patent Document 3] JP 2010-188276 A

[Patent Document 4] JP 2008-155126 A

[Patent Document 5] JP H9-57105 A

SUMMARY OF THE INVENTION

TECHNICAL PROBLEMS

[0005]

However, according to studies conducted by the present inventors, it has been found that when methacrolein and/or methacrylic acid is produced by a gas phase oxidation reaction using the catalyst described in Patent Documents 1 to 5, the reaction rate of a raw material compound and the selectivity of methacrolein and/or methacrylic acid may not be sufficient.

MEANS FOR SOLVING THE PROBLEMS

[0006]

As a result of extensive studies in view of the above problems, the present inventors have found that, by using a specific catalyst molded body, methacrolein and/or methacrylic acid is obtained with a high selectivity while maintaining a high reaction rate of a raw material compound, and achieved the present invention. In other words, the gist of the present invention is as follows.

[0007]

[1] A catalyst molded body satisfying the following Formulae (1) and (2), when a density (g/cm^3) of the catalyst molded body is X, a filling bulk density (g/cm^3) of the catalyst molded body is Y, and a representative length (cm) of the catalyst molded body is Z:

$$0.700 (\text{g/cm}^3)^2 \leq (X \times Y) \leq 1.200 (\text{g/cm}^3)^2 \quad \text{Formula (1)}$$

$$0.050 \text{ g/cm}^2 \leq (X \times Z) \leq 0.130 \text{ g/cm}^2 \quad \text{Formula (2).}$$

[2] The catalyst molded body according to [1], wherein X is 1.300 g/cm^3 or more and 1.800 g/cm^3 or less.

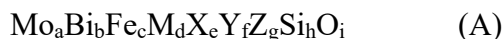
[3] The catalyst molded body according to [1] or [2], wherein Y is 0.500 g/cm^3 or more and 0.800 g/cm^3 or less.

[4] The catalyst molded body according to any one of [1] to [3], wherein Z is 0.030 cm or more and 0.100 cm or less.

[5] The catalyst molded body according to any one of [1] to [4], wherein the catalyst molded body contains molybdenum, bismuth, and iron as essential components.

[6] The catalyst molded body according to any one of [1] to [5], wherein the catalyst

molded body has a composition represented by the following Formula (A):



(In Formula (A), Mo, Bi, Fe, Si, and O represent molybdenum, bismuth, iron, silicon, and oxygen, respectively; M represents at least one element selected from the group consisting of cobalt and nickel; X represents at least one element selected from the group consisting of chromium, lead, manganese, calcium, magnesium, niobium, silver, barium, tin, tantalum, and zinc; Y represents at least one element selected from the group consisting of phosphorus, boron, sulfur, selenium, tellurium, cerium, tungsten, antimony, and titanium; Z represents at least one element selected from the group consisting of lithium, sodium, potassium, rubidium, cesium, and thallium; a, b, c, d, e, f, g, h, and i represent the atomic ratios of each element, and when $a = 12$, $b = 0.01-3$, $c = 0.01-5$, $d = 1-12$, $e = 0-8$, $f = 0-5$, $g = 0.001-2$, $h = 0-20$, and i is the oxygen atom ratio required to satisfy the valence of each above component).

[7] The catalyst molded body according to any one of [1] to [6], which is a catalyst molded body for producing methacrolein and/or methacrylic acid.

[8] The catalyst molded body according to any one of [1] to [7], which is a catalyst molded body for producing methacrolein and/or methacrylic acid from one or more compounds selected from isobutylene, tert-butyl alcohol and methyl-tert-butyl ether.

[9] A method for producing methacrolein and/or methacrylic acid, wherein one or more compounds selected from the group consisting of isobutylene, tert-butyl alcohol and methyl-tert-butyl ether are subjected to a gas phase catalytic oxidation with molecular oxygen in the presence of the catalyst molded body according to any one of [1] to [8].

EFFECT OF THE INVENTION

[0008]

According to the present invention, methacrolein and/or methacrylic acid can be produced with a high selectivity while maintaining a high reaction rate of a raw material compound.

DETAILED DESCRIPTION OF THE PRESENT INVENTION

[0009]

Hereinafter, one embodiment of the present invention will be described in detail, but the embodiment described below is an example (representative example) of one embodiment of the present invention, and the present invention is not limited to the following embodiment. Hereinafter, the embodiment in which methacrolein and/or methacrylic acid is produced using a catalyst molded body according to one embodiment

of the present invention will be described. It is noted that in the present invention, the catalyst molded body means a catalyst obtained by molding a catalyst component.

[0010]

<Catalyst Molded Body>

The catalyst molded body according to the present embodiment is a catalyst molded body satisfying the following Formulae (1) and (2), when a density (g/cm^3) of the catalyst molded body is X, a filling bulk density (g/cm^3) of the catalyst molded body is Y, and a representative length (cm) of the catalyst molded body is Z.

[0011]

$$0.700 (\text{g/cm}^3)^2 \leq (X \times Y) \leq 1.200 (\text{g/cm}^3)^2 \quad \text{Formula (1)}$$

[0012]

$$0.050 \text{ g/cm}^2 \leq (X \times Z) \leq 0.130 \text{ g/cm}^2 \quad \text{Formula (2)}$$

[0013]

In the present invention, the density X of the catalyst molded body, the filling bulk density Y of the catalyst molded body, and the representative length Z of the catalyst molded body mean values calculated by the method described in the Examples described later.

[0014]

When the catalyst molded body satisfies the above Formulae (1) and (2), methacrolein and/or methacrylic acid can be produced with a high selectivity while maintaining a high reaction rate of a raw material. Although this mechanism is not clear, it is considered that according to the catalyst molded body satisfying the above Formulae (1) and (2), the catalyst molded body in which the density and the shape of the catalyst molded body are appropriately controlled is appropriately filled and thereby methacrolein and/or methacrylic acid can be produced with a high raw material reaction rate and a high selectivity.

[0015]

Among the above Formula (1), the value of $(X \times Y)$ is preferably $0.750 (\text{g/cm}^3)^2$ or more, more preferably $0.790 (\text{g/cm}^3)^2$ or more, further preferably $0.800 (\text{g/cm}^3)^2$ or more, from the viewpoint of improving the reaction rate of the raw material compound and improving the selectivity of the target product, while the value of $(X \times Y)$ is preferably $1.170 (\text{g/cm}^3)^2$ or less, more preferably $1.150 (\text{g/cm}^3)^2$ or less, and further preferably $1.100 (\text{g/cm}^3)^2$ or less, for improving the selectivity of methacrolein and/or methacrylic acid.

[0016]

Among the above Formula (2), the value of $(X \times Z)$ is preferably 0.055 g/cm^2 or

more, further preferably 0.060 g/cm^2 or more, particularly preferably 0.065 g/cm^2 or more, and most preferably 0.070 g/cm^2 or more, from the viewpoint of improving the reaction rate of the raw material compound and improving the selectivity of the target product, while the value of $(X \times Z)$ is preferably 0.125 g/cm^2 or less and further preferably 0.120 g/cm^2 or less, for improving the selectivity of methacrolein and/or methacrylic acid.

[0017]

The density X of the catalyst molded body is not particularly limited as long as the catalyst molded body satisfies the above Formulae (1) and (2), but the density X is preferably 1.300 g/cm^3 or more, further preferably 1.350 g/cm^3 or more, and particularly preferably 1.400 g/cm^3 or more, from the viewpoint of improving the reaction rate of the raw material compound and improving the selectivity of the target product, while the density X is preferably 1.800 g/cm^3 or less, further preferably 1.750 g/cm^3 or less, and particularly preferably 1.700 g/cm^3 or less, for improving the selectivity of the methacrolein and/or methacrylic acid.

[0018]

The filling bulk density Y of the catalyst molded body is not particularly limited as long as the catalyst molded body satisfies the above Formula (1), but the filling bulk density Y is preferably 0.500 g/cm^3 or more, further preferably 0.550 g/cm^3 or more, and particularly preferably 0.600 g/cm^3 or more, from the viewpoint of improving the reaction rate of the raw material compound and improving the selectivity of the target product, while the filling bulk density Y is preferably 0.800 g/cm^3 or less, further preferably 0.750 g/cm^3 or less, and particularly preferably 0.700 g/cm^3 or less, for improving the selectivity of methacrolein and/or methacrylic acid.

[0019]

The representative length Z of the catalyst molded body is not particularly limited as long as the catalyst molded body satisfies the above Formula (2), but the representative length Z is preferably 0.030 cm or more, further preferably 0.035 cm or more, particularly preferably 0.040 cm or more, and most preferably 0.050 cm or more, from the viewpoint of improving the reaction rate of the raw material compound and improving the selectivity of the target product, while the representative length Z is preferably 0.100 cm or less, further preferably 0.095 cm or less, and particularly preferably 0.090 cm or less, for improving the selectivity of methacrolein and/or methacrylic acid.

[0020]

The volume of the catalyst molded body is not particularly limited, but the volume is preferably 0.050 cm^3 or more, further preferably 0.060 cm^3 or more, and

particularly preferably 0.070 cm^3 or more, from the viewpoint of improving the reaction rate of the raw material compound and improving the selectivity of the target product, while the volume is preferably 1.000 cm^3 or less, more preferably 0.800 cm^3 or less, further preferably 0.500 cm^3 or less, and particularly preferably 0.300 cm^3 or less, for improving the selectivity of methacrolein and/or methacrylic acid. Note that, in the present invention, the volume of the catalyst molded body means, for example, a volume excluding a space portion when the catalyst molded body is in a hollow cylindrical shape.

[0021]

The outer surface area of the catalyst molded body is not particularly limited, but the outer surface area is preferably 0.500 cm^2 or more, further preferably 0.700 cm^2 or more, particularly preferably 0.800 cm^2 or more in order to produce methacrolein and/or methacrylic acid with high selectivity while maintaining a high reaction rate of the raw material compound, while the outer surface area is preferably 5.000 cm^2 or less, further preferably 4.000 cm^2 or less, and particularly preferably 3.000 cm^2 or less. The outer surface area of the catalyst molded body refers to the geometric surface area.

[0022]

The mass of the catalyst molded body is not particularly limited, but the mass is preferably 0.050 g or more, further preferably 0.080 g or more, and particularly preferably 0.100 g or more, from the viewpoint of improving the reaction rate of the raw material compound and improving the selectivity of the target product, while the mass is preferably 0.300 g or less, further preferably 0.280 g or less, and particularly preferably 0.250 g or less for improving the selectivity of methacrolein and/or methacrylic acid.

[0023]

The shape of the catalyst molded body is not particularly limited, and may be, for example, spherical, cylindrical, ring-shape, star-shape or the like. Among them, ring-shape is preferable.

[0024]

Further, the catalyst molded body may be supported on a carrier. In this case, silica, alumina, silica-alumina, magnesia, titania, silicon carbide, or the like can be used as the carrier. These may be used alone or in combination of two or more. It is noted that, when the carrier is used, silica is preferably used among the above. In the present invention, when the carrier is used to support the catalyst component, a catalyst molded body is referred to as a catalyst molded body including the carrier.

[0025]

The average diameter of the catalyst molded body is not particularly limited, but the average diameter is preferably 0.10 cm or more, further preferably 0.15 cm or more,

and particularly preferably 0.20 cm or more when the catalyst molded body has a ring-shape, cylindrical, or spherical shape in order to suppress the increase in pressure loss during the catalytic reaction and stably produce methacrolein and/or methacrylic acid over a long period of time, while the average diameter is preferably 1.00 cm or less, further preferably 0.90 cm or less, and particularly preferably 0.80 cm or less for improving the selectivity of methacrolein and/or methacrylic acid.

[0026]

The average pore volume of the catalyst molded body is not particularly limited, but the average pore volume is preferably $0.100 \text{ cm}^3/\text{g}$ or more, further preferably $0.200 \text{ cm}^3/\text{g}$ or more, and particularly preferably $0.300 \text{ cm}^3/\text{g}$ or more, for improving the selectivity of methacrolein and/or methacrylic acid, while the average pore volume is preferably $1.000 \text{ cm}^3/\text{g}$ or less, further preferably $0.900 \text{ cm}^3/\text{g}$ or less, and particularly preferably $0.800 \text{ cm}^3/\text{g}$ or less in order to provide the catalyst molded body with adequate strength and stably produce methacrolein and/or methacrylic acid over a long period of time. The average pore volume of the catalyst molded body can be measured by a mercury intrusion measuring apparatus or the like.

[0027]

The catalyst component constituting the catalyst molded body is not particularly limited, but it is preferable that the catalyst composition is such that methacrolein and/or methacrylic acid can be produced from isobutylene, tert-butyl alcohol, methyl-tert-butyl ether, or the like. Specifically, it is preferable that the catalyst molded body is a composite oxide containing a plurality of catalyst components. Specifically, it is preferable that the catalyst molded body contains molybdenum, bismuth, and iron as catalyst components. Hereinafter, preferred embodiments of the catalyst molded body containing molybdenum, bismuth and iron as essential components will be described.

[0028]

The atomic ratio of bismuth to molybdenum is not particularly limited, but the atomic ratio of bismuth to 12 atoms of molybdenum is preferably 0.01 or more, further preferably 0.10 or more, particularly preferably 0.20 or more, for improving the selectivity of methacrolein and/or methacrylic acid, while the atomic ratio of bismuth to 12 atoms of molybdenum is preferably 3.00 or less, further preferably 2.00 or less, and particularly preferably 1.50 or less.

[0029]

The atomic ratio of iron to molybdenum is not particularly limited, but the atomic ratio of iron to 12 atoms of molybdenum is preferably 0.01 or more, further preferably 0.10 or more, particularly preferably 0.50 or more, for improving the selectivity of

methacrolein and/or methacrylic acid, while the atomic ratio of iron to 12 atoms of molybdenum is preferably 5.00 or less, further preferably 4.00 or less, and particularly preferably 3.00 or less.

[0030]

Further, the catalyst molded body may contain elements other than those described above. Such elements include, but are not limited to, cobalt, nickel, chromium, lead, manganese, calcium, magnesium, niobium, silver, barium, tin, tantalum, zinc, phosphorus, boron, sulfur, selenium, tellurium, cerium, tungsten, antimony, titanium, lithium, sodium, potassium, rubidium, cesium, or thallium.

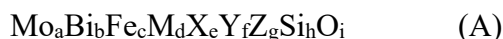
[0031]

Among them, it is preferable that the catalyst molded body contains at least one element selected from the group consisting of cobalt and nickel, and at least one element selected from the group consisting of lithium, sodium, potassium, rubidium, cesium, and thallium.

[0032]

Of these, it is particularly preferred that the catalyst molded body has a composition represented by the following Formula (A):

[0033]



(In Formula (A), Mo, Bi, Fe, Si, and O represent molybdenum, bismuth, iron, silicon, and oxygen, respectively; M represents at least one element selected from the group consisting of cobalt and nickel; X represents at least one element selected from the group consisting of chromium, lead, manganese, calcium, magnesium, niobium, silver, barium, tin, tantalum, and zinc; Y represents at least one element selected from the group consisting of phosphorus, boron, sulfur, selenium, tellurium, cerium, tungsten, antimony, and titanium; Z represents at least one element selected from the group consisting of lithium, sodium, potassium, rubidium, cesium, and thallium; a, b, c, d, e, f, g, h, and i represent the atomic ratios of each element, and when a = 12, b = 0.01-3, c = 0.01-5, d = 1-12, e = 0-8, f = 0-5, g = 0.001-2, h = 0-20, and i is the oxygen atom ratio required to satisfy the valence of each above component). Note that the composition of the catalyst is a value calculated from the raw material charge amount of each element.

[0034]

Note that M, X, Y, and Z in the above Formula (A) may each be composed of a plurality of elements. When M, X, Y, and Z are each composed of a plurality of elements, d, e, f, and g each represent the total atomic ratio of each element to 12 atoms of molybdenum.

[0035]

In Formula (A), M preferably contains cobalt and nickel.

[0036]

In Formula (A), X is preferably lead.

[0037]

In Formula (A), Y preferably contains at least one selected from the group consisting of phosphorus, antimony, and tungsten, and particularly preferably contains at least tungsten.

[0038]

In Formula (A), Z is preferably cesium.

[0039]

In Formula (A), b is more preferably 0.10 or more, further preferably 0.20 or more, while b is preferably 2.00 or less, and further preferably 1.50 or less.

[0040]

In Formula (A), c is more preferably 0.10 or more, further preferably 0.50 or more, while c is preferably 4.00 or less, and further preferably 3.00 or less.

[0041]

In Formula (A), d is more preferably 2.00 or more, further preferably 3.00 or more, while d is preferably 11.00 or less, and further preferably 10.00 or less.

[0042]

In Formula (A), e is more preferably 0.01 or more, further preferably 0.10 or more, while e is more preferably 5.00 or less, further preferably 2.00 or less, and particularly preferably 1.00 or less.

[0043]

In Formula (A), f is more preferably 0.10 or more, further preferably 0.50 or more, while f is preferably 4.0 or less, and further preferably 3.0 or less.

[0044]

In Formula (A), g is more preferably 0.01 or more, further preferably 0.10 or more, while g is preferably 1.50 or less, and further preferably 1.00 or less.

[0045]

When the catalyst molded body contains the silica carrier, h is more preferably 0.10 or more, further preferably 0.50 or more, while h is preferably 15.0 or less, and further preferably 12.00 or less in Formula (A).

[0046]

<Method for Producing Catalyst Molded Body>

Hereinafter, a method for producing a catalyst molded body used in the present

invention will be described.

[0047]

The method for producing a catalyst molded body according to the present invention is not particularly limited, and various known methods can be applied. For example, a method in which an aqueous slurry containing catalyst components such as molybdenum, bismuth and iron is produced and dried, and if necessary, pulverized and molded, followed by heat treatment to obtain a catalyst molded body can be mentioned.

[0048]

There is no particular limitation on the method of producing such an aqueous slurry, and various methods such as a known coprecipitation method and a known oxide mixing method can be used. Hereinafter, as a representative example, a production example of a catalyst molded body by the coprecipitation method will be described.

[0049]

The raw material of the catalyst component used for producing the aqueous slurry is not particularly limited, and an oxide, a chloride, a hydroxide, a sulfate, a nitrate, a carbonate, an acetate, an ammonium salt, or a mixture thereof of each element constituting the catalyst component can be used. For example, ammonium paramolybdate, molybdenum trioxide, or the like can be used as a raw material of molybdenum. As a raw material of bismuth, bismuth oxide, bismuth nitrate, or the like can be used. As a raw material of iron, ferric nitrate, ferric oxide, or the like can be used. These may be used alone or in combination of two or more. Known compounds can also be used for other elements. Note that the ratio of each raw material may be appropriately adjusted so as to obtain a desired catalyst composition.

[0050]

In addition, when two or more kinds of raw materials are used, it is preferable to mix each raw material while stirring to produce the aqueous slurry. By performing mixing while stirring, it can be prepared with good reproducibility.

[0051]

The stirring method is not particularly limited, and known stirring apparatuses such as a rotary type stirrer, such as a rotary blade stirrer and a high-speed rotary shear stirrer (homogenizer), a pendulum type linear motion type stirrer, a shaker for stirring each container, a vibration type stirrer using ultrasonic waves, and the like can be cited, but it is preferable to use a rotary type stirrer such as a rotary blade stirrer or a high-speed rotary shear stirrer (homogenizer) because the intensity of the stirring apparatus can be easily adjusted and the stirring apparatus is industrially simple. The rotation speed of a stirring blade or a rotating blade in the rotary type stirrer may be appropriately adjusted

to such an extent that inconvenience such as scattering of the liquid does not occur by taking into consideration the shape of the container, the stirring blade, the baffle plate, and the like, and the liquid amount, and the like.

[0052]

Further, after mixing each raw material, it is preferable to perform heat treatment.

[0053]

As a condition of the heat treatment, there is no particular limitation, but the treatment temperature is preferably 70 °C or more, further preferably 80 °C or more, while the treatment temperature is preferably 180 °C or less, particularly preferably 150 °C or less. Further, the heating time is preferably 10 minutes or more, while the upper limit for the heating treatment time is not particularly limited, but is preferably 5 hours or less. By performing the heat treatment, a high-performance catalyst can be prepared with good reproducibility.

[0054]

Although a catalyst molded body can be produced by molding described later the obtained aqueous slurry, it is preferable to dry the aqueous slurry before molding to make it into a state of a dried powder to perform molding. There is no particular limitation on the method of drying the aqueous slurry, and examples thereof include a method of drying using a spray dryer, a method of drying using a slurry dryer, a method of drying using a drum dryer, a method of pulverizing a lumpy dried product obtained by evaporation to dryness, and the like. As for the drying condition, for example, when drying using a spray dryer, the inlet temperature is preferably 100 to 500 °C, more preferably 110 to 300 °C. In addition, the outlet temperature is preferably 100 to 300 °C, more preferably 105 to 200 °C. By performing drying under the above conditions, a dried product having a constant bulk density can be obtained with good reproducibility, and a catalyst molded body satisfying the requirements in the present invention can be produced with good reproducibility.

[0055]

In some cases, the dried powder thus obtained contains a salt of such as nitric acid derived from a catalyst raw material or the like. Therefore, it is preferable to calcine the dried powder in order to desorb such a salt, but when the dried powder is molded and then calcined, the salt may decompose and the strength of the catalyst molded body may decrease. Therefore, it is preferable to perform calcination before molding the dried powder. In addition, for the purpose of desorbing the salt, the primary calcination of the dried powder may be performed, and then the secondary calcination for forming a final catalyst active point structure may be performed after performing molding described later.

Further, the primary calcination and the secondary calcination may be performed, followed by molding.

[0056]

Each calcination temperature is preferably from 200 to 600 °C, more preferably from 300 to 550 °C. By performing calcination at a temperature in this range, a high-performance catalyst molded body tends to be obtained. The time for continuing the heat treatment after reaching the predetermined temperature is not particularly limited, but in order to produce a catalyst molded body having high productivity and high performance, it is preferable to perform the heat treatment in the range of 0.5 to 15 hours. It is more preferably carried out in the range of 1 to 10 hours, and further preferably carried out in the range of 1.5 to 8 hours.

[0057]

In addition, it is preferable that the bulk density of the dried powder immediately before molding is 0.500 (g/ml) to 1.500 (g/ml). By using the dried powder in the range of this value, a catalyst molded body having a filling bulk density described above can be produced with good reproducibility.

The bulk density of the dried powder can be appropriately adjusted by controlling the temperature and time at the time of performing drying. Generally, the higher the temperature at the time of drying and the longer the drying time, the larger the bulk density of the dried powder, and the lower the temperature at the time of drying and the shorter the drying time, the smaller the bulk density tends to be.

[0058]

In addition, in the present invention, in order for the catalyst molded body to satisfy Formulae (1) and (2), it is necessary to adjust the mass, the bulk density, the outer surface area, and the volume of the catalyst molded body, but for these, the conditions at the time of molding may be appropriately adjusted by a known method.

[0059]

There is no particular limitation on the method of molding the dried powder before or after calcination, and various molding methods such as tableting molding, extrusion molding, and supporting on a carrier can be used.

In addition, at the time of molding, for the purpose of controlling the specific surface area, the pore volume and the pore distribution of the molded product, increasing the mechanical strength, and increasing the handling property at the time of molding, an inorganic compound such as graphite or diatomaceous earth, an inorganic fiber such as glass fiber, ceramic fiber or carbon fiber, and an organic binder such as methyl cellulose, ethyl cellulose, carboxymethyl cellulose, hydroxyethyl cellulose, hydroxyethyl methyl

cellulose, hydroxypropyl cellulose, hydroxypropyl methyl cellulose, or the like may be added. These may be used alone or in combination of two or more.

The amount of the organic binder to be used is appropriately selected depending on the type and size of the particles, the type of the liquid, and the like, but is usually 0.05 to 15 parts by mass, preferably 0.1 to 10 parts by mass, with respect to 100 parts by mass of the particles. As the amount of the organic binder added increases, the moldability tends to be improved, and as the amount decreases, the post-treatment such as heat treatment after molding tends to be simpler.

[0060]

In order to produce a catalyst molded body satisfying Formula (1), it is necessary to control the density and the filling property of the catalyst. Although the density of the catalyst can be adjusted, for example, by the amount of a liquid added to the dried powder or by the pressure at the time of extrusion molding the kneaded product in the case of wet extrusion molding, it is preferable to control the extrusion pressure of the kneaded product together because the filling property of the catalyst may be deteriorated when controlled only by the amount of the liquid added to the dried powder. The pressure during extrusion can be adjusted by the die opening ratio of the extruder and the discharge rate of the kneaded product discharged therefrom. In the case of tableting molding, it can be adjusted by the density and compression pressure of the dried powder. The filling property of the catalyst can be adjusted, for example, by an additive material added at the time of molding in addition to the density of the catalyst.

In the present invention, the die opening ratio of the extruder is defined as follows.

Die opening ratio of extruder (%) = (total area of opening of die/cylinder cross-sectional area of extruder) × 100

In addition, in order to produce a catalyst molded body satisfying Formula (2), it is necessary to control the density and the shape of the catalyst. The shape of the catalyst can be adjusted by the shape or the like of the die at the time of molding.

[0061]

<Method for Producing Methacrolein and/or Methacrylic Acid>

The catalyst according to the present embodiment is effective in producing methacrolein and/or methacrylic acid. Specifically, in the presence of the catalyst according to the present embodiment, isobutylene, tert-butyl alcohol, methyl-tert-butyl ether, or the like as a reaction raw material is subjected to a gas phase catalytic oxidation with molecular oxygen, whereby methacrolein and/or methacrylic acid can be produced. Incidentally, the reaction is usually carried out by a fixed bed reaction. In addition, one

kind of catalyst may be used, or two or more kinds may be used in combination. Further, if necessary, the catalyst may be diluted with an inert carrier such as silica, alumina, silica-alumina, silicon carbide, titania, magnesia, ceramic balls, or stainless-steel and used. Further, catalysts having different compositions and activities may be mixed and used. Again, it may be diluted with an inert carrier.

[0062]

As the fixed bed reactor, a tube type reactor by heat medium circulation is preferably used. Although a single tube type in which only one reaction tube is provided may be used, it is preferable to use a multi-tube type reactor having a plurality of reaction tubes, industrially. In this case, it is preferable that the reactor has a heat removal facility. The length of the reaction tube may be appropriately selected depending on the amount to be produced, from a few centimeters to a few meters.

[0063]

The total concentration of isobutylene, tert-butyl alcohol, and methyl-tert-butyl ether, which are the reaction raw materials in the raw material gas, is preferably set to 1 to 20% by volume, although there is no particular limitation.

[0064]

It is preferable that the raw material gas contains water vapor in addition to the reaction raw materials and molecular oxygen. The concentration of water vapor in the raw material gas is preferably 1 to 45% by volume. It is preferable that the water vapor is also diluted with an inert gas such as nitrogen or carbon dioxide and used.

[0065]

The total molar ratio of isobutylene, tert-butyl alcohol, and methyl-tert-butyl ether as the raw materials to oxygen molecules is not particularly limited, but is preferably 50 mol% or more, and on the other hand, is preferably 300 mol% or less. Note that, although the molecular oxygen may be pure oxygen gas, industrially, it is preferable to be air. The reaction pressure during the reaction is not particularly limited and is used from normal pressure to several atmospheres. In addition, the reaction temperature is preferably in the range of 250 to 450 °C.

[0066]

Although the reaction is preferably carried out by a fixed bed reactor of a tube type, the tube diameter of the reaction tube to be used is preferably 5 mm or more, and further preferably 10 mm or more, in order to uniformly fill the catalyst molded body and stably produce methacrolein and/or methacrylic acid for a long period of time, and on the other hand, it is preferably 100 mm or less, particularly preferably 80 mm or less, in order to efficiently remove heat generated by the reaction and stably produce methacrolein

and/or methacrylic acid.

[0067]

The contact time of the raw materials to be supplied to the reaction tube to the catalyst is preferably 0.1 seconds or more, further preferably 1.0 second or more, in order to obtain a sufficient conversion rate, and on the other hand, it is preferably 10.0 seconds or less, and further preferably 8.0 seconds or less, in order to obtain a high selectivity.

EXAMPLES

[0068]

Hereinafter, the present invention will be described using Examples, but the present invention is not limited to these Examples. In the present invention, the reaction rate of isobutylene and the selectivity of the produced methacrolein and methacrylic acid are defined as follows, respectively.

[0069]

Reaction rate of isobutylene (%) = (number of moles of isobutylene reacted/number of moles of isobutylene supplied) $\times$ 100

[0070]

Selectivity of methacrolein (%) = (number of moles of methacrolein produced/number of moles of isobutylene reacted) $\times$ 100

[0071]

Selectivity of methacrylic acid (%) = (number of moles of methacrylic acid produced/number of moles of isobutylene reacted) $\times$ 100

[0072]

The density X (g/cm³) of the catalyst molded body, the filling bulk density Y (g/cm³) of the catalyst molded body, and the representative length Z (cm) of the catalyst molded body were calculated by the following formula, respectively.

[0073]

Density X (g/cm³) of catalyst molded body = mass (g) of catalyst molded body/volume (cm³) of catalyst molded body

Note that, when the catalyst molded body had a geometrically shaped hollow portion, it was defined that the volume of the catalyst molded body did not include the hollow portion.

[0074]

The filling bulk density Y (g/cm³) of the catalyst molded body was calculated from the total weight of the catalyst molded body when the catalyst molded body was filled into a 100-ml graduated cylinder by a method based on JIS-K 7365. Specifically, it

was calculated by the following formula.

[0075]

Filling bulk density Y (g/cm^3) = total weight (g) of catalyst molded body when filled/100 (ml)

[0076]

Representative length Z (cm) of catalyst molded body = volume (cm^3) of catalyst molded body/outer surface area (cm^2) of catalyst molded body

[0077]

[Example 1]

To 600 parts by mass of pure water, 300 parts by mass of ammonium paramolybdate was dissolved. Subsequently, 7.44 parts by mass of ammonium paratungstate, 13.8 parts by mass of cesium nitrate, 16.4 parts by mass of antimony trioxide, and 19.8 parts by mass of bismuth trioxide were added while stirring the solution, and the mixture was warmed to 50 °C (Liquid A). Separately, to 600 parts by mass of pure water, 125.9 parts by mass of ferric nitrate, 45.3 parts by mass of nickel nitrate, 272.0 parts by mass of cobalt nitrate, 18.8 parts by mass of lead nitrate, and 3.4 parts by mass of 85% phosphoric acid were sequentially added and dissolved, and the mixture was warmed to 30 °C (Liquid B). Under stirring, Liquid B was added to Liquid A to obtain an aqueous slurry, which was heated at 90 °C for 2 hours to proceed the reaction, and then heated to 103 °C and heated for 2 hours.

The slurry containing the raw material compound of the catalyst component thus obtained was dried under the conditions of an inlet temperature of 180 °C and an outlet temperature of 120 °C using a spray dryer to obtain a spherical dried powder having an average particle diameter of 60 μm . The resulting dried powder was calcined at 300 °C for 1 hour, followed by calcination at 515 °C for 3 hours to obtain a catalyst calcined powder. The resulting catalyst calcined powder had a bulk density of 1.00 (g/cm^3).

[0078]

After well mixing 400 parts by mass of the obtained catalyst calcined powder and 12 parts by mass of methyl cellulose powder as an organic binder component, 166 parts by mass of pure water was added and kneaded until a clay-like substance was obtained by a kneader. Next, the obtained amorphous kneaded product was formed into a ring-shape having an outer diameter of 0.64 cm, an inner diameter of 0.25 cm, and a length of 0.50 cm using a piston type extruder having a die opening ratio of 1.2%. The discharge rate of the kneaded product was adjusted to 250 mm/s. The obtained molded body was dried using a hot air dryer at 110 °C, thereby obtaining a catalyst molded body 1.

[0079]

The obtained catalyst molded body 1 had the density X of 1.322 (g/cm³), the filling bulk density Y of 0.692 (g/cm³), the representative length Z of 0.070 (cm), and the value of (X × Y) was 0.915 (g/cm³)² and the value of (X × Z) was 0.093 (g/cm³). The composition excluding the organic binder component that was distilled off under the conditions of oxygen and the catalyst reaction temperature was Mo₁₂Bi_{0.6}Fe_{2.2}Co_{6.6}Ni_{1.1}Pb_{0.4}P_{0.2}W_{0.2}Sb_{0.8}Cs_{0.5}. Note that the composition of the catalyst molded body 1 is a value calculated from the raw material charge amount of each element.

[0080]

(Production of Methacrolein and Methacrylic Acid)

The obtained catalyst molded body 1 was filled in a stainless-steel reaction tube having an inner diameter of 20 mm, and a height of 500 mm in an amount of 10 g, and reacted using a raw material gas containing 5% by volume of isobutylene, 12% by volume of oxygen, 10% by volume of water vapor, and 73% by volume of nitrogen at a contact time of 3.6 seconds, and a reaction temperature of 340 °C under normal pressure. As a result of the reaction, the reaction rate of isobutylene was 94.7%, the selectivity of methacrolein was 85.4%, and the selectivity of methacrylic acid was 3.6%. In addition, the reaction rate and selectivity were stable and did not decrease in the continuous reaction for 200 hours.

[0081]

[Example 2]

After well mixing 400 parts by mass of the catalyst calcined powder obtained by the same method as in Example 1 and 12 parts by mass of methyl cellulose powder as an organic binder component, 156 parts by mass of pure water was added and kneaded until a clay-like substance was obtained by a kneader. Next, the obtained amorphous kneaded product was formed into a ring-shape having an outer diameter of 0.63 cm, an inner diameter of 0.30 cm, and a length of 0.50 cm using a piston type extruder having a die opening ratio of 1.1%. The discharge rate of the kneaded product was adjusted to 250 mm/s. The obtained molded body was dried using a hot air dryer at 110 °C, thereby obtaining a catalyst molded body 2.

[0082]

The obtained catalyst molded body 2 had the density X of 1.463 (g/cm³), the filling bulk density Y of 0.713 (g/cm³), the representative length Z of 0.062 (cm), and the value of (X × Y) was 1.043 (g/cm³)² and the value of (X × Z) was 0.092 (g/cm²).

Using the obtained catalyst molded body 2, the production of methacrolein and methacrylic acid was carried out in the same manner as in Example 1. The reaction results

are shown in Table 1. In addition, the reaction rate and selectivity were stable and did not decrease in the continuous reaction for 200 hours.

[0083]

[Example 3]

After well mixing 400 parts by mass of the catalyst calcined powder obtained by the same method as in Example 1 and 12 parts by mass of methyl cellulose powder as an organic binder component, 168 parts by mass of pure water was added and kneaded until a clay-like substance was obtained by a kneader. Next, the obtained amorphous kneaded product was formed into a ring-shape having an outer diameter of 0.65 cm, an inner diameter of 0.22 cm, and a length of 0.50 cm using a piston type extruder having a die opening ratio of 1.2%. The discharge rate of the kneaded product was adjusted to 250 mm/s. The obtained molded body was dried using a hot air dryer at 110 °C, thereby obtaining a catalyst molded body 3.

[0084]

The obtained catalyst molded body 3 had the density X of 1.323 (g/cm³), the filling bulk density Y of 0.734 (g/cm³), the representative length Z of 0.070 (cm), and the value of $(X \times Y)$ was 0.971 (g/cm³)² and the value of $(X \times Z)$ was 0.092 (g/cm²). Using the obtained catalyst molded body 3, the production of methacrolein and methacrylic acid was carried out in the same manner as in Example 1. The reaction results are shown in Table 1. In addition, the reaction rate and selectivity were stable and did not decrease in the continuous reaction for 200 hours.

[0085]

[Comparative Example 1]

After well mixing 400 parts by mass of the catalyst calcined powder obtained by the same method as in Example 1 and 12 parts by mass of methyl cellulose powder as an organic binder component, 156 parts by mass of pure water was added and kneaded until a clay-like substance was obtained by a kneader. Next, the obtained amorphous kneaded product was formed into a columnar shape having an outer diameter of 0.64 cm and a length of 0.50 cm using a piston type extruder having a die opening ratio of 1.4%. The discharge rate of the kneaded product was adjusted to 250 mm/s. The obtained molded body was dried using a hot air dryer at 110 °C, thereby obtaining a catalyst molded body 4.

[0086]

The obtained catalyst molded body 4 had the density X of 1.409 (g/cm³), the filling bulk density Y of 0.882 (g/cm³), the representative length Z of 0.098 (cm), and the value of $(X \times Y)$ was 1.242 (g/cm³)² and the value of $(X \times Z)$ was 0.137 (g/cm²).

Using the obtained catalyst molded body 4, the production of methacrolein and methacrylic acid was carried out in the same manner as in Example 1. The reaction results are shown in Table 1.

[0087]

[Comparative Example 2]

After well mixing 400 parts by mass of the catalyst calcined powder obtained by the same method as in Example 1 and 12 parts by mass of methyl cellulose powder as an organic binder component, 156 parts by mass of pure water was added and kneaded until a clay-like substance was obtained by a kneader. Next, the obtained amorphous kneaded product was formed into a columnar shape having an outer diameter of 0.64 cm and a length of 0.40 cm using a piston type extruder having a die opening ratio of 1.4%. The discharge rate of the kneaded product was adjusted to 250 mm/s. The obtained molded body was dried using a hot air dryer at 110 °C, thereby obtaining a catalyst molded body 5. The obtained catalyst molded body 5 had the density X of 1.406 (g/cm³), the filling bulk density Y of 0.888 (g/cm³), the representative length Z of 0.089 (cm), and the value of (X × Y) was 1.249 (g/cm³)² and the value of (X × Z) was 0.125 (g/cm²).

[0088]

Using the obtained catalyst molded body 5, the production of methacrolein and methacrylic acid was carried out in the same manner as in Example 1. The reaction results are shown in Table 1.

[0089]

[Comparative Example 3]

After well mixing 400 parts by mass of the catalyst calcined powder obtained by the same method as in Example 1 and 12 parts by mass of methyl cellulose powder as an organic binder component, 164 parts by mass of pure water was added and kneaded until a clay-like substance was obtained by a kneader. Next, the obtained amorphous kneaded product was formed into a columnar shape having an outer diameter of 0.64 cm and a length of 0.50 cm using a piston type extruder having a die opening ratio of 1.4%. The discharge rate of the kneaded product was adjusted to 250 mm/s. The obtained molded body was dried using a hot air dryer at 110 °C, thereby obtaining a catalyst molded body 6.

[0090]

The obtained catalyst molded body 6 had the density X of 1.351 (g/cm³), the filling bulk density Y of 0.824 (g/cm³), the representative length Z of 0.098 (cm), and the value of (X × Y) was 1.113 (g/cm³)² and the value of (X × Z) was 0.132 (g/cm²).

Using the obtained catalyst molded body 6, the production of methacrolein and

methacrylic acid was carried out in the same manner as in Example 1. The reaction results are shown in Table 1.

[0091]

[Example 4]

After well mixing 400 parts by mass of the catalyst calcined powder obtained by the same method as in Example 1 and 12 parts by mass of methyl cellulose powder as an organic binder component, 178 parts by mass of pure water was added and kneaded until a clay-like substance was obtained by a kneader. Next, the obtained amorphous kneaded product was formed into a ring-shape having an outer diameter of 0.63 cm, an inner diameter of 0.30 cm, and a length of 0.50 cm using a piston type extruder having a die opening ratio of 1.1%. The discharge rate of the kneaded product was adjusted to 250 mm/s. The obtained molded body was dried using a hot air dryer at 110 °C, thereby obtaining a catalyst molded body 7.

The obtained catalyst molded body 7 had the density X of 1.306 (g/cm³), the filling bulk density Y of 0.612 (g/cm³), the representative length Z of 0.062 (cm), and the value of $(X \times Y)$ was 0.799 (g/cm³)² and the value of $(X \times Z)$ was 0.081 (g/cm²).

Using the obtained catalyst molded body 7, the production of methacrolein and methacrylic acid was carried out in the same manner as in Example 1. The reaction results are shown in Table 1. In addition, the reaction rate and selectivity were stable and did not decrease in the continuous reaction for 200 hours.

[0092]

[Example 5]

After well mixing 400 parts by mass of the catalyst calcined powder obtained by the same method as in Example 1 and 12 parts by mass of methyl cellulose powder as an organic binder component, 148 parts by mass of pure water was added and kneaded until a clay-like substance was obtained by a kneader. Next, the obtained amorphous kneaded product was formed into a ring-shape having an outer diameter of 0.63 cm, an inner diameter of 0.30 cm, and a length of 0.50 cm using a piston type extruder having a die opening ratio of 1.1%. The discharge rate of the kneaded product was adjusted to 250 mm/s. The obtained molded body was dried using a hot air dryer at 110 °C, thereby obtaining a catalyst molded body 8.

The obtained catalyst molded body 8 had the density X of 1.514 (g/cm³), the filling bulk density Y of 0.750 (g/cm³), the representative length Z of 0.062 (cm), and the value of $(X \times Y)$ was 1.135 (g/cm³)² and the value of $(X \times Z)$ was 0.094 (g/cm²).

Using the obtained catalyst molded body 8, the production of methacrolein and methacrylic acid was carried out in the same manner as in Example 1. The reaction results

are shown in Table 1. In addition, the reaction rate and selectivity were stable and did not decrease in the continuous reaction for 200 hours.

[0093]

[Example 6]

After well mixing 400 parts by mass of the catalyst calcined powder obtained by the same method as in Example 1 and 12 parts by mass of methyl cellulose powder as an organic binder component, 164 parts by mass of pure water was added and kneaded until a clay-like substance was obtained by a kneader. Next, the obtained amorphous kneaded product was formed into a ring-shape having an outer diameter of 0.63 cm, an inner diameter of 0.10 cm, and a length of 0.50 cm using a piston type extruder having a die opening ratio of 1.4%. The discharge rate of the kneaded product was adjusted to 250 mm/s. The obtained molded body was dried using a hot air dryer at 110 °C, thereby obtaining a catalyst molded body 9.

The obtained catalyst molded body 9 had the density X of 1.351 (g/cm³), the filling bulk density Y of 0.775 (g/cm³), the representative length Z of 0.087 (cm), and the value of $(X \times Y)$ was 1.047 (g/cm³)² and the value of $(X \times Z)$ was 0.117 (g/cm²).

Using the obtained catalyst molded body 9, the production of methacrolein and methacrylic acid was carried out in the same manner as in Example 1. The reaction results are shown in Table 1. In addition, the reaction rate and selectivity were stable and did not decrease in the continuous reaction for 200 hours.

[0094]

[Example 7]

After well mixing 400 parts by mass of the catalyst calcined powder obtained by the same method as in Example 1 and 12 parts by mass of methyl cellulose powder as an organic binder component, 158 parts by mass of pure water was added and kneaded until a clay-like substance was obtained by a kneader. Next, the obtained amorphous kneaded product was formed into a ring-shape having an outer diameter of 0.63 cm, an inner diameter of 0.10 cm, and a length of 0.50 cm using a piston type extruder having a die opening ratio of 1.4%. The discharge rate of the kneaded product was adjusted to 250 mm/s. The obtained molded body was dried using a hot air dryer at 110 °C, thereby obtaining a catalyst molded body 10.

The obtained catalyst molded body 10 had the density X of 1.413 (g/cm³), the filling bulk density Y of 0.815 (g/cm³), the representative length Z of 0.087 (cm), and the value of $(X \times Y)$ was 1.152 (g/cm³)² and the value of $(X \times Z)$ was 0.122 (g/cm²).

Using the obtained catalyst molded body 10, the production of methacrolein and methacrylic acid was carried out in the same manner as in Example 1. The reaction results

are shown in Table 1. In addition, the reaction rate and selectivity were stable and did not decrease in the continuous reaction for 200 hours.

[0095]

[Comparative Example 4]

After well mixing 400 parts by mass of the catalyst calcined powder obtained by the same method as in Example 1 and 12 parts by mass of methyl cellulose powder as an organic binder component, 192 parts by mass of pure water was added and kneaded until a clay-like substance was obtained by a kneader. Next, the obtained amorphous kneaded product was formed into a ring-shape having an outer diameter of 0.63 cm, an inner diameter of 0.30 cm, and a length of 0.50 cm using a piston type extruder having a die opening ratio of 1.1%. The discharge rate of the kneaded product was adjusted to 250 mm/s. The obtained molded body was dried using a hot air dryer at 110 °C, thereby obtaining a catalyst molded body 9.

The obtained catalyst molded body 9 had the density X of 1.216 (g/cm³), the filling bulk density Y of 0.552 (g/cm³), the representative length Z of 0.062 (cm), and the value of $(X \times Y)$ was 0.671 (g/cm³)² and the value of $(X \times Z)$ was 0.075 (g/cm²).

Using the obtained catalyst molded body 11, the production of methacrolein and methacrylic acid was carried out in the same manner as in Example 1. The reaction results are shown in Table 1. In addition, the reaction rate and selectivity were stable and did not decrease in the continuous reaction for 200 hours.

[0096]

[Comparative Example 5]

After well mixing 400 parts by mass of the catalyst calcined powder obtained by the same method as in Example 1 and 12 parts by mass of methyl cellulose powder as an organic binder component, 170 parts by mass of pure water was added and kneaded until a clay-like substance was obtained by a kneader. Next, the obtained amorphous kneaded product was formed into a columnar shape having an outer diameter of 0.66 cm and a length of 0.55 cm using a piston type extruder having a die opening ratio of 1.7%. The discharge rate of the kneaded product was adjusted to 250 mm/s. The obtained molded body was dried using a hot air dryer at 110 °C, thereby obtaining a catalyst molded body 10.

The obtained catalyst molded body 10 had the density X of 1.278 (g/cm³), the filling bulk density Y of 0.791 (g/cm³), the representative length Z of 0.103 (cm), and the value of $(X \times Y)$ was 1.011 (g/cm³)² and the value of $(X \times Z)$ was 0.132 (g/cm²).

Using the obtained catalyst molded body 12, the production of methacrolein and methacrylic acid was carried out in the same manner as in Example 1. The reaction results

are shown in Table 1. In addition, the reaction rate and selectivity were stable and did not decrease in the continuous reaction for 200 hours.

[0097]

Table 1

	Density X (g/cm ³)	Filling Bulk Density Y (g/cm ³)	Representative Length Z (cm)	Value of X * Y ((g/cm ³) ²)	Value of X * Z (g/cm ²)	Volume (cm ³)	Reaction Rate of Isobutylene (%)	Selectivity of Methacrolein (%)	Selectivity of Methacrylic Acid (%)	Total Selectivity (%)	Total Yield (%)
Example 1	1.322	0.692	0.070	0.915	0.093	0.136	94.7	85.4	3.6	89.0	84.3
Example 2	1.463	0.713	0.062	1.043	0.091	0.120	95.3	85.0	3.5	88.5	84.3
Example 3	1.323	0.734	0.070	0.971	0.092	0.117	95.1	84.4	3.7	88.1	83.8
Comparative Example 1	1.409	0.882	0.098	1.242	0.137	0.161	95.0	81.7	4.2	85.9	81.6
Comparative Example 2	1.406	0.888	0.089	1.249	0.125	0.129	95.0	82.2	4.2	86.4	82.1
Comparative Example 3	1.351	0.824	0.098	1.113	0.132	0.161	95.4	81.9	4.2	86.1	82.1
Example 4	1.306	0.612	0.062	0.799	0.081	0.120	94.3	85.5	3.5	89.0	83.9
Example 5	1.514	0.750	0.062	1.135	0.094	0.120	95.4	84.3	3.4	87.7	83.7
Example 6	1.351	0.775	0.087	1.047	0.117	0.152	95.1	84.2	3.7	87.9	83.6
Example 7	1.413	0.815	0.087	1.152	0.122	0.152	95.4	83.7	3.8	87.5	83.5
Comparative Example 4	1.216	0.552	0.062	0.671	0.075	0.120	91.8	85.7	3.5	89.2	81.9
Comparative Example 5	1.278	0.791	0.103	1.011	0.132	0.188	95.0	82.5	4.1	86.6	82.3

[0098]

From the above results, it can be seen that, by satisfying the above Formulae (1) and (2), it is possible to produce methacrolein and/or methacrylic acid with a high selectivity maintaining a high reaction rate of the raw material compound.

CLAIMS

1. A catalyst molded body for producing methacrolein and/or methacrylic acid from one or more compounds selected from isobutylene, tert-butyl alcohol, and methyl-tert-butyl ether satisfying the following Formulae (1) and (2), when a density (g/cm^3) of the catalyst molded body is X, a filling bulk density (g/cm^3) of the catalyst molded body is Y, and a representative length (cm) of the catalyst molded body is Z:

$$0.700 (\text{g/cm}^3)^2 \leq (X \times Y) \leq 1.200 (\text{g/cm}^3)^2 \quad \text{Formula (1)}$$

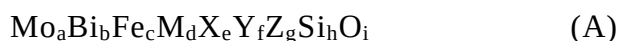
$$0.050 \text{ g/cm}^2 \leq (X \times Z) \leq 0.130 \text{ g/cm}^2 \quad \text{Formula (2),}$$

wherein X is 1.300 g/cm^3 or more and 1.800 g/cm^3 or less, Y is 0.500 g/cm^3 or more and 0.815 g/cm^3 or less, and Z is 0.062 cm or more and 0.100 cm or less,

wherein Z is calculated by the following formula:

representative length Z (cm) of the catalyst molded body = volume (cm^3) of the catalyst molded body/outer surface area (cm^2) of the catalyst molded body, and

wherein the catalyst molded body has a composition represented by the following Formula (A):



(In Formula (A), Mo, Bi, Fe, Si, and O represent molybdenum, bismuth, iron, silicon, and oxygen, respectively; M represents at least one element selected from the group consisting of cobalt and nickel; X represents at least one element selected from the group consisting of chromium, lead, manganese, calcium, magnesium, niobium, silver, barium, tin, tantalum, and zinc; Y represents at least one element selected from the group consisting of phosphorus, boron, sulfur, selenium, tellurium, cerium, tungsten, antimony, and titanium; Z represents at least one element selected from the group consisting of lithium, sodium, potassium, rubidium, cesium, and thallium; a, b, c, d, e, f, g, h, and i represent the atomic ratios of each element, wherein when $a = 12$, $b = 0.01-3$, $c = 0.01-5$, $d = 1-12$, $e = 0-8$, $f = 0-5$, $g = 0.001-2$, and $h = 0-20$, and i is the oxygen atom ratio required to satisfy the valence of each above component).

2. The catalyst molded body according to claim 1, wherein X is 1.300 g/cm^3 or more and 1.700 g/cm^3 or less.

3. The catalyst molded body according to claim 1 or 2, wherein Y is 0.600 g/cm^3 or more and 0.815 g/cm^3 or less.

4. The catalyst molded body according to any one of claims 1 to 3, wherein Z is 0.062 cm or more and 0.090 cm or less.

5. A method for producing methacrolein and/or methacrylic acid, wherein one or more compounds selected from the group consisting of isobutylene, tert-butyl alcohol

and methyl-tert-butyl ether are subjected to a gas phase catalytic oxidation with molecular oxygen in the presence of the catalyst molded body according to any one of claims 1 to 4.