[CLAIMS]

What is claimed is:

[Claim 1]

A production method of one or both of (meth)acrolein and (meth)acrylic acid using a heat-exchange-type reaction vessel comprising a reaction tube at an inner part, the production method comprising

causing an oxidation reaction of a raw material supplied to the reaction tube while circulating a heat medium to an outer side of the reaction tube to produce one or both of (meth)acrolein and (meth)acrylic acid,

wherein the reaction tube comprises i layers, which are a plurality of catalyst layers having different catalyst charging amounts per unit volume, in a longitudinal direction of the reaction tube, provided that i is an integer of 2 or more, and

the oxidation reaction satisfies Expression (1),

$\xi \leq 0.275$ (mol·K/h/W) (1)

provided that the following formula is satisfied,

[Math. 1]

$$\xi = \mathrm{F} \times \left(m1 / \sum_{j=1}^{i} mj \right) \Big/ (U \times A) \cdots (*)$$

in Formula (*), m1 is a catalyst charging amount (kg) in a first catalyst layer from a raw material inlet side of the reaction tube; mj is a catalyst charging amount (kg) in a j-th catalyst layer from the raw material inlet side of the reaction tube; j is an integer of 1 or more and i or less; F is a supply amount (mol/h) of the raw material supplied to the reaction tube; A is an inner surface area (m^2) of the reaction tube with which the first catalyst layer from the raw material inlet side of the reaction tube comes into contact; and U is an overall heat transfer coefficient ($W/m^2/K$) based on an inner surface area of a portion in the reaction tube

with which both the first catalyst layer and the heat medium come into contact, and wherein U is obtained by using a methodology, which requires:

supplying an inert gas composed of 21% by volume of oxygen and 79% by volume of nitrogen to the reaction tube at a temperature 50 °C lower than that of the heat medium circulating the outer side of the reaction tube;

measuring temperature distribution across minute region, which is defined as a region obtained by dividing the reaction tube into 20 mm in the longitudinal direction; and

calculating a temperature change dT1 according to Formula (***),

dT1 = U × inner surface area (m^2) of reaction tube with which minute region is in contact with × dT2/[mass flow rate (g/s) of inert gas × specific heat (J/g/K) of inert gas] (***),

obtaining a value of U by using a least squares method to find the value at which the calculated dT1 coincides with the actual measured value of dT1; and

calculating U as an arithmetic mean value obtained for 20 adjacent minute regions, provided that at least one of the 20 minute regions has a temperature difference dT2 of 3 °C or higher, where dT2 is the difference between the heat medium temperature and the average temperature in the minute region.

[Claim 2]

The production method according to Claim 1,

wherein the oxidation reaction further satisfies Expression (1'),

$0.002 \leq \xi \leq 0.275$ (mol·K/h/W) (1').

[Claim 3]

The production method according to Claim 1,

wherein the oxidation reaction further satisfies Expression (1"),

$\xi \leq 0.24$ (mol·K/h/W) (1").

[Claim 4]

The production method according to Claim 1,

wherein the oxidation reaction further satisfies Expression (2),

$0.25 \leq \tau \leq 0.5$ (2)

provided that the following formula is satisfied,

[Math. 2]

$$\tau = m1 \Big/ \sum_{k=1}^{i} mk \cdots (**)$$

in Formula (**), m1 is the catalyst charging amount (kg) in the first catalyst layer from the raw material inlet side of the reaction tube; mk is a catalyst charging amount (kg) in a k-th catalyst layer from the raw material inlet side of the reaction tube; and k is an integer of 1 or more and i or less.

[Claim 5]

The production method according to Claim 4,

wherein the oxidation reaction further satisfies Expression (2'),

$0.26 \leq \tau \leq 0.5$ (2').

[Claim 6]

The production method according to Claim 1,

wherein i as the number of the catalyst layers of the reaction tube is 2 to 4.

[Claim 7]

The production method according to Claim 1,

wherein U is 40 to 400 ($W/m^2/K$).

[Claim 8]

The production method according to Claim 1,

wherein U is 50 to 300 ($W/m^2/K$).

[Claim 9]

The production method according to Claim 1,

wherein F is 1 to 20 (mol/h).

[Claim 10]

The production method according to Claim 1,

wherein F is 2.5 to 15 ($mol \cdot h^{-1}$).

[Claim 11]

The production method according to Claim 1,

wherein A is 0.03 to 0.6 (m^2).

[Claim 12]

The production method according to any one of Claims 1 to 11,

wherein the raw material is at least one selected from propylene, isobutylene, tert-butanol, and methyl tert-butyl ether, and

one or both of the (meth)acrolein and the (meth)acrylic acid are (meth)acrolein and (meth)acrylic acid.

[Claim 13]

The production method according to any one of Claims 1 to 11,

wherein the raw material is (meth)acrolein, and

one or both of the (meth)acrolein and the (meth)acrylic acid are (meth)acrylic acid.

[Claim 14]

The production method according to Claim 12,

wherein a catalyst having a formulation represented by Formula (I) is used in the catalyst layer,

$$Mo_{a1}Bi_{b1}Fe_{c1}M_{d1}X_{e1}Y_{f1}Z_{g1}Si_{h1}O_{i1} \quad (I)$$

in Formula (I), Mo, Bi, Fe, Si, and O each represent molybdenum, bismuth, iron, silicon, and oxygen; 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; and a1, b1, c1, d1, e1, f1, g1, h1, and i1 represent an atomic ratio of each of the elements, provided that when a1 = 12, b1 = 0.01 to 3, c1 = 0.01 to 5, d1 = 1 to 12, e1 = 0 to 8, f1 = 0 to 5, g1 = 0.001 to 2, h1 = 0 to 20, and i1 is an atomic ratio of oxygen required to satisfy a valence of the element.

[Claim 15]

The production method according to Claim 13,

wherein a catalyst having a formulation represented by Formula (II) is used in the catalyst layer,

$$P_{a2}Mo_{b2}V_{c2}Cu_{d2}X_{e2}Y_{f2}Z_{g2}O_{h2} \quad (II)$$

in Formula (II), P, Mo, V, Cu, and O each represent phosphorus, molybdenum, vanadium, copper, and oxygen; X represents at least one element selected from the group consisting of antimony, bismuth, arsenic, germanium, zirconium, tellurium, silver, selenium, silicon, tungsten, and boron; Y represents at least one element selected from the group consisting of iron, zinc, chromium, magnesium, tantalum, cobalt, manganese, barium, gallium, cerium, and lanthanum; Z represents at least one element selected from the group consisting of potassium, rubidium, cesium, and thallium; and a2, b2, c2, d2, e2, f2, g2, and h2 represent an atomic ratio of each of the elements, provided that when b2 = 12, a2 = 0.5 to 3, c2 = 0.01 to 3, d2 = 0.01 to 2, e2 = 0 to 3, f2 = 0 to 3, g2 = 0.01 to 3, and h2 is an atomic ratio

of oxygen required to satisfy a valence of the element.