DESCRIPTION

HEAT TRANSFER TUBE FOR MULTITUBULAR REACTOR

Technical Field

[0001]

The present invention is related to a heat transfer tube for a multitubular reactor.

Background Art

[0002]

A multitubular reactor using a fixed-bed catalyst is provided with a plurality of heat transfer tubes which are filled with catalyst and production of a substance which is desired (objective substance) is conducted by feeding a feed gas in the heat transfer tube to react the feed gas with the catalyst in the catalyst bed.

In the reaction of the feed gas and the catalyst in the catalyst bed, a reaction temperature at which the gas and the catalyst are reacted is important for producing the objective substance efficiently. Further, removal of heat generation during the reaction and preheating of the feed gas are made by means of a heated liquid (HTS) such as sodium nitrite and potassium nitrate in the multitubular reactor in order that the reaction temperature is optimal.

[0003]

As a method for preheating the feed gas, a method for raising a temperature by a heater or the like, a method for lengthening a length of a region from a reactor inlet to the catalyst bed (a preheating region) or the like has been employed.

However, problems of the former method are that the heater or the like is required to be installed on a feeding path of the feed gas, causing a high initial investment in facilities and that a running cost is increased due to operation of the heater or the like.

Further, a problem of the latter method is that the multitubular reactor itself is made large since the length of the heat transfer tube is required to be lengthen by the length corresponding to the length by which the preheating region is lengthen, resulting in the high initial investment in facilities.

In both methods, a large-scaled modification of facilities is required when the methods are applied to the existing facilities.

[0004]

Patent Literatures 1 and 2 disclose methods which can obtain the effect of preheating the feed gas more simply than the above-mentioned methods.

Patent Literatures 1 and 2 disclose a method for

producing methacrylic acid using a multitubular reactor, specifically a method for subjecting isobutylene and/or tertiary butanol to catalytic vapor-phase oxidation with molecular oxygen in a first-step reaction and a latter-step reaction, and a technique of preventing a trouble of occlusion of the catalyst layer in the latter-step reaction, which trouble is caused by an occlusive material contained in the former-step reaction product gas. In Patent Literatures 1 and 2, there is described that the insertion of an insert into the preheating region in the heat transfer tube of the multitubular reactor can give the preheating effect of the latter-step gas, whereby the effect of avoiding the occlusion and so on can be achieved.

[0005]

Further, in Patent Literatures 1 and 2, there is described that a void ratio of the preheating region into which the insert is inserted is preferably from 30 to 99 %, and preferably from 40 % to 99 % in the case where the insert is lodlike, and preferably from 50 % to 99 % in the case where the insert is platelike.

Furthermore, in Patent Literature 2, a straight rod, a zigzag rod, a spiral rod, a polygonal prism, a cylinder, and a circular column are exemplified as the form of the rodlike insert, and a ribbon, a zigzag plate and a spiral plate are exemplified as the platelike insert. In addition,

Patent Literature 2 describes that the platelike insert is not required to be a complete plate and may be a reticular plate.

[0006]

The employment of the techniques disclosed in Patent Literatures 1 and 2 makes it possible to obtain the effect of precluding the trouble of occlusion of the catalyst bed in the heat transfer tube upon production of methacrylic acid. The method for heating the feed gas more efficiently is sought for producing methacrylic acid stably for a longer time.

Background Art Literatures

Patent Literatures

[0007]

Patent Literature 1: JP 2-22242 A

Patent Literature 2: JP 1-165543 A

Summary of Invention

[0008]

In view of the above-described situations, the object of the present invention is to provide a heat transfer tube for a multitubular reactor which can heat a feed gas more efficiently.

[0009]

A heat transfer tube for a multitubular reactor of a first invention is a heat transfer tube including a gas supplying port at one end and a gas discharging port at the other end and a catalyst layer packed between the both ends, wherein an insert extending in an axial direction of the heat transfer tube is placed between the gas supplying port and the catalyst layer, which insert is characterized in that the insert includes a rod portion and a spiral portion which is provided on a side surface of the rod portion.

The heat transfer tube for a multitubular reactor of a second invention is characterized in that, in the first invention, the insert is configured so that a value obtained by dividing a projected area of the insert in the axial direction of the insert by a cross-sectional area of space of the heat transfer tube (that is, (a projected area of the insert in the axial direction of the insert)/(a cross-sectional area of space of the heat transfer tube) is from 0.80 to 0.99.

The heat transfer tube for a multitubular reactor of a third invention is characterized in that a pitch of the spiral potion of the insert is 50 mm or less in the first or the second invention.

The heat transfer tube for a multitubular reactor of a fourth invention is characterized in that, in the first, the second or the third invention is used in a fixed-bed

reactor for producing (meth)acrolein and/or (meth)acrylic acid by oxidizing propylene, isobutylene, t-butyl alcohol or a mixture thereof in a gas phase with a gas containing molecular oxygen.

[0010]

A heat transfer tube for a multitubular reactor of a fifth invention is a heat transfer tube including a gas supplying port at one end and a gas discharging port at the other end and a catalyst layer packed between the both ends, wherein an insert extending in an axial direction of the heat transfer tube is placed between the gas supplying port and the catalyst layer, which is characterized in that the insert is of a shape of zigzag sequence of plate portions having sloped surfaces inclined relative to the axial direction of the heat transfer tube, and the insert is configured such that a value obtained by dividing a projected area of the insert in the axial direction of the insert by a cross sectional area of space of the heat transfer tube (that is, (a projected area of the insert in the axial direction of the insert)/(a cross sectional area of space of the heat transfer tube) is from 0.5 to 0.7.

The heat transfer tube for a multitubular reactor of a sixth invention is characterized in that, in the first invention, the insert is formed by bending a plate material.

The heat transfer tube for a multitubular reactor of a

seventh invention is characterized in that, in the fifth or the sixth invention, an angle formed by adjacent plate portions is from 20° to 100°.

The heat transfer tube for a multitubular reactor of an eighth invention is characterized in that, in the fifth, the sixth, of the seventh invention is used in a fixed-bed reactor for producing (meth)acrolein and/or (meth)acrylic acid by oxidizing propylene, isobutylene, t-butyl alcohol or a mixture thereof in a gas phase with a gas containing molecular oxygen.

In the present description, the term "acrolein" conceptually includes methacrolein and the term "acrylic acid" conceptually includes methacrylic acid.

[0011]

According to the first invention, when the insert is disposed within the heat transfer tube, the rod portion of the insert is positioned near a center of the heat transfer tube and the spiral portion is placed between the rod portion and a tube wall (strictly, an inner surface of the tube wall) of the heat transfer tube. Then, since the gas flows along the spiral portion, a gas flow resistance due to the insertion of the insert is suppressed, while the contact between the gas and the tube wall can be improved. Thus, heat exchange efficiency can be significantly increased and therefore the temperature of the gas can be

increased in a short time. Further, the increase in the gas temperature in a short time makes it possible to shorten a distance between the gas supplying port and the catalyst bed, whereby the heat transfer tube can be shortened and/or an amount of the catalyst packed in the tube can be increased.

According to the second invention, since the insert is configured so that the value obtained by dividing the projected area of the insert in the axial direction by the cross-sectional area of space of the heat transfer tube is from 0.80 to 0.90, the heat exchange effectiveness can be improved, while the occlusion caused by the increase in the gas flow resistance due to the insertion of the insert is prevented.

According to the third invention, the heat exchange (or heat transfer) effectiveness is increased.

According to the fourth invention, since the distance between the gas supplying port and the catalyst bed can be shortened and/or an amount of the catalyst packed in the heat transfer tube can be increased, lifetime of the catalyst can be made longer. Thus, the productivity of (meth)acrolein and/or (meth)acrylic acid can be improved, while the running cost can be low.

[0012]

According to the fifth invention, the insert has the

sloped surfaces inclined relative to the axial direction and is configured such that the value obtained by the projected area of the insert in the axial direction by the cross-sectional direction of the space of the heat transfer tube is from 0.5 to 0.7. For this reason, the heat exchange effective can be improved, while the occlusion due to the increase in the gas flow resistance caused by the insertion of the insert is prevented. Further, since a gas flow toward the tube wall of the heat transfer tube is created by the sloped surfaces and therefore the gas and the tube wall are liable to contact, the heat exchange efficiency can be significantly increased and the gas temperature can be raised in a short time. Furthermore, when the gas temperature can be raised in a short time, the distance between the gas supplying port to the catalyst layer can be shortened, whereby the heat transfer tube can be shortened and the amount of the catalyst packed into the heat transfer tube can be increased.

According to the sixth invention, since the insert can be formed easily, the investment in facilities can be low.

According to the seventh invention, the occlusion due to the increase in resistance can be prevented, while the reduction in heat exchange efficiency can be prevented.

According to the eighth invention, since the distance between the gas supplying port and the catalyst layer can

be shortened and/or the amount of the catalyst packed into the heat transfer tube can be increased, the lifetime of the catalyst can be made longer. Therefore, the productivity of acrolein and/or acrylic acid can be improved, while the running cost can be low.

Brief Description of Drawings

[0013]

Fig. 1(A) is a partial schematic view illustrating a heat transfer tube P1 wherein an insert 10 is placed according to an embodiment of the present first invention and Fig. 1(B) is a partial enlarged view illustrating the insert 10.

Fig. 2 is a schematic cross-sectional view of the heat transfer tube P1 in which the inert 10 is inserted according to an embodiment of the present first invention.

Fig. 3(A) is a schematic view illustrating a multitubular reactor T and Fig. 3(B) is a schematic view illustrating a heat transfer tube P without the insert.

Fig. 4 is a schematic view illustrating a calculation model of an example and the calculation model of a comparative example.

Fig. 5 is a view showing results of numerical simulation of feed-gas temperature-rise effect given by the insert.

Fig. 6 is a view showing experimental results of the feed-gas temperature-rise effect given by the insert.

Fig. 7(A) is a partial schematic view illustrating a heat transfer tube P2 wherein an insert 100 is placed according to an embodiment of the present fifth invention and Fig. 7(B) is a partial enlarged view illustrating the insert 100.

Fig. 8 is a schematic cross-sectional view of the heat transfer tube P2 in which the inert 100 is inserted according to an embodiment of the present fifth invention.

Fig. 9 is a view showing experimental results for confirming the effect of the insert on overall heat transfer coefficient.

Fig. 10 is a schematic view illustrating a calculation model.

Fig. 11 is a view showing results of numerical simulation of feed-gas temperature-rise effect given by the insert.

Embodiments for Carrying Out the Invention

[0014]

Next, embodiments of the present invention are described with reference to drawings.

A heat transfer tube for multitubular reactor of the present invention is one wherein an insert for heating a

reaction gas before being supplied to catalyst is placed in the heat transfer tube in which the catalyst is packed, which tube is characterized by the shape of the insert.

[0015]

Firstly, the multitubular reactor and the heat transfer tube P (in this description, "p" collectively refers to the heat transfer tubes) are described with reference to Fig. 3 before describing the insert which is a characteristic of the present invention.

[0016]

In Fig. 3, the symbol "T" denotes the multitubular reactor. As shown in Fig. 3, many heat transfer tubes P are provided in this multitubular reactor T. Each heat transfer tube P is a hollow tube of which both ends are opened, inside which tube a fixed-bed catalyst is packed to form a catalyst layer C.

These heat transfer tubes P are constructed such that the feed gas BRG supplied to the multitubular reactor T is advanced from one ends of the heat transfer tubes P and this feed gas BRG reacts by means of the catalyst in the heat transfer tubes P and the produced objective substance ARG is discharged from the other ends of the heat transfer tubes P in the multitubular reactor T.

[0017]

Further, a liquid (HTS) such as heated sodium nitrite

or heated potassium nitrate is supplied around the heat transfer tubes P in the multitubular reactor T so that this HTS can remove the heat generation during the reaction and preheat the feed gas BRG.

[0018]

For this reason, when the feed gas BRG is supplied to the multitubular reactor T, the feed gas BRG is preheated and the preheated feed gas BRG is reacted by means of the catalyst to give the objective substance ARG and then this objective substance can be discharged from the multitubular reactor T.

[0019]

Next, the insert 10 which is a characteristic of the present first invention is described.

The heat transfer tube P1 for multitubular reactor of the present first invention (hereinafter merely referred to as a "heat transfer tube P1 of the present first invention") is constructed so that the preheating of the feed gas BRG is made between the gas supplying port into which the feed gas BRG is supplied and the catalyst layer C. Specifically, space is formed between the gas supplying port into which the feed gas BRG is supplied and the catalyst layer C, and the preheating of the feed gas BRG is carried out in this space (hereinafter referred to as a "preheating layer"). Further, the insert 10 is inserted in

the preheated layer of the heat transfer tube P1 of the present first invention so as to improve the preheating effect of the feed gas BRG.

[0020]

As shown in Fig. 1, the insert 10 inserted into the heat transfer tube P1 of the present first invention has a rod portion 11 which extends in an axial direction and a spiral portion 12 which is formed on a side surface of this rod portion 11. The spiral portion 12 is formed by winding a wire rod around the side surface of the rod portion 11 and fixed to the rod portion 11 by welding.

The method for forming the spiral portion 12 is not particularly limited and the spiral portion 12 may be formed by a technique of winding a member other than the wire rod, such as a plate member or a prismatic member, around the side surface of the rod portion 11.

[0021]

This insert 10 is placed so that one end of the rod portion 11 is put on the catalyst layer C. In other words, the insert 10 is placed in the preheating layer, not being connected or fixed to the heat transfer tube P1.

[0022]

Since this construction is employed, the gas flow from the gas supplying port to the catalyst layer C varies in the heat transfer tube P1 of the present first invention

wherein the insert 10 is placed in the preheating layer, due to the effect of the insert 10. In other words, since the rod portion 11 of the insert 10 is placed near the center of the heat transfer tube P1 and the spiral portion 12 is placed between the rod portion 11 and the tube wall of the heat transfer tube P1, the feed gas BRG supplied into the heat transfer tube P1 does not flow in parallel to the axial direction of the heat transfer tube P1, and flows along the spiral portion 12 between the rod portion 11 and the tube wall of the heat transfer tube P1. Then, the feed gas BRG flows near the tube wall of the heat transfer tube P1 of which temperature is made high, whereby the preheating of the feed gas BRG can be conducted efficiently. In addition, since the feed gas BRG flows along the spiral portion 12, the flow resistance of the feed gas BRG caused by the insertion of the insert 10 can be suppressed.

[0023]

In other words, the heat exchange efficiency between the feed gas BRG and the tube wall of the heat transfer tube P1 can be significantly increased and the temperature of the feed gas BRG can be increased in a short time, since the contact between the feed gas BRG and the tube wall can be improved in the heat transfer tube P1 of the present first invention with the flow resistance of the feed gas BRG suppressed.

Further, when the temperature of the feed gas BRG is increased in a short time, a distance between the gas supplying port and the catalyst layer C can be shortened, whereby the multitubular reactor T can be made smaller by shortening the heat transfer tube P1. Furthermore, when the length of the heat transfer tube P1 is the same, the preheating layer can be made short, whereby the amount of catalyst packed in the heat transfer tube P1 can be increased.

[0024]

A size of the insert 10 that is inserted in the heat transfer tube P1 of the present first invention is not particularly limited as long as the shape of the insert is as described above and can be placed in the preheating layer of the heat transfer tube P1.

However, when the insert 10 is formed so that a value obtained by dividing a projected area of the insert 10 in the axial direction by a cross-sectional area of space of the heat transfer tube P1 (the cross-sectional area of a hollow portion (a gas path) of the heat transfer tube P1) is from 0.80 to 0.99, space sufficient for smooth flow of the feed gas BRG can be ensured. Then, the heat exchange efficiency can be improved, while the occlusion due to the increase in the gas flow resistance caused by the insertion of the insert 10 is prevented.

[0025]

Further, a pitch "a" (see Fig. 1(A)) of the spiral portion 12 of the insert 10 is not particularly limited. When the pitch "a" is too large, the feed gas BRG passes through the preheating layer rapidly, resulting in possibility that the heat exchange is not made sufficiently.

Therefore, the pitch a of the spiral portion 12 is preferably 50 mm or less. Such a pitch can sufficiently ensure the time for which the feed gas BRG passes through the preheating layer, and therefore the heat exchange effectiveness can be improved.

[0026]

Next, the insert 100 which is a characteristic of the present fifth invention is described.

The heat transfer tube P2 for for multitubular reactor of the present fifth invention (hereinafter merely referred to as a "heat transfer tube 100 of the present fifth invention") is constructed so that the preheating of the feed gas BRG is made between the gas supplying port into which the feed gas BRG is supplied and the catalyst layer C. Specifically, space is formed between the gas supplying port into which the feed gas BRG is supplied and the catalyst layer C, and the preheating of the feed gas BRG is carried out in this space (hereinafter referred to as a "preheating layer"). Further, the insert 100 is inserted

in the preheated layer of the heat transfer tube P2 of the present fifth invention so as to improve the preheating effect of the feed gas BRG.

[0027]

As shown in Fig. 7, the insert 100 inserted into the heat transfer tube p2 of the present fifth invention is a member which is a plate member which extends in the axial direction and has a plurality of sloped surfaces 100f which are inclined relative to the axial direction of the heat transfer tube. Further, the insert 100 is formed so that a value obtained by dividing a projected area of the insert 100 in the axial direction by a cross-sectional area of the heat transfer tube P2 (the cross-sectional area of a hollow portion (a gas path) of the heat transfer tube P2) is from 0.5 to 0.7.

[0028]

For example, as shown in Fig. 8, when a width L1 of the plate member constituting the insert 100 is from 14 mm to 20 mm, and a length L3 of each of the plate portions (see Fig. 7) is from 14 mm to 28 mm, an angle θ formed by adjacent plate portions (see Fig. 7) is made to be from 30° to 90°. Such construction makes a length L2 which is a length of the insert 100 viewed from the axial direction to be from 14 mm to 20 mm. Then, when the inner diameter D1 of the heat transfer tube P2 is from 20 mm to 30 mm, the

value obtained by dividing the projected area of the insert 100 (L1xL2) in the axial direction by the cross-sectional area of space of the heat transfer P2 can be made to be from 0.5 to 0.7.

[0029]

Further, as shown in Fig. 7, this insert 100 is placed so that one end thereof is put on the catalyst layer C. In other words, the inert 100 is placed in the preheating layer, not being connected or fixed to the heat transfer tube P2.

[0030]

Since this construction is employed, the gas flow from the gas supplying port to the catalyst layer C varies in the heat transfer tube P2 of the present fifth invention wherein the insert 100 is placed in the preheating layer, due to the effect of the insert 100. In other words, since the insert 100 has a plurality of sloped surfaces 100f inclined relative to the axial direction of the heat transfer tube P2, the feed gas BRG supplied into the heat transfer tube P2 does not flow in parallel to the axial direction of the heat transfer tube P2, and flows towards the tube wall of the heat transfer tube P2. Then, the majority of the feed gas BRG can be brought into contact with the tube wall of the heat transfer tube P2, whereby the preheating of the feed gas BRG can be conducted

efficiently. In addition, since the feed gas BRG flows along the sloped surfaces 100f, the flow resistance of the feed gas BRG caused by the insertion of the insert 100 can be suppressed.

[0031]

In other words, the heat exchange efficiency between the feed gas BRG and the tube wall of the heat transfer tube P2 can be significantly increased and the temperature of the feed gas BRG can be increased in a short time, since the contact between the feed gas BRG and the tube wall can be improved in the heat transfer tube P2 of the present fifth invention with the flow resistance of the feed gas BRG suppressed.

Further, when the temperature of the feed gas BRG is increased in a short time, a distance between the gas supplying port and the catalyst layer C can be shortened, whereby the multitubular reactor T can be made smaller by shortening the heat transfer tube P2. Furthermore, when the length of the heat transfer tube P2 is the same, the preheating layer can be made short, whereby the amount of catalyst packed in the heat transfer tube P2 can be increased.

[0032]

The insert 100 inserted into the heat transfer tube P2 of the present fifth invention may be formed using any

method and any material, as long as the insert 100 is a member consisting of a plurality of plate portions having sloped surfaces 100f and the value obtained by dividing the projected area of the insert 100 in the axial direction by the cross-sectional area of space of the heat transfer tube P2 is from 0.5 to 0.7.

For example, the insert 100 can be formed by bending (or folding) a plate member such that the sloped surfaces 100f are continued zigzag. If the insert 100 is formed by this method, the insert 100 can be formed easily and therefore the investment in facilities can be low.

[0033]

Further, the angle θ formed by the adjacent plate portions in the insert 100 (see Fig. 7), for example, the bending angle θ in the case where the insert 100 is formed by bending the plate member, is not particularly limited. However, the angle θ is preferably from 20° to 100°, and more preferably 30° to 90° in order to prevent the occlusion due to the increase in flow resistance caused by the disposition of the insert 100 in the heat transfer tube P2 and to maintain the heat exchange efficiency high to some extent.

[0034]

(Application to production of (meth)acrolein and/or (meth)acrylic acid)

When the heat transfer tube P of any one of the embodiments according to the present inventions is used in a fixed-bed reactor for producing (meth)acrolein and/or (meth)acrylic acid by oxidizing propylene, isobutylene, t-butyl alcohol or a mixture thereof with a gas containing molecular oxygen in a gas phase, the productivity of (meth)acrolein and/or (meth)acrylic acid can be improved and the running cost can be made low. Therefore, the heat transfer tube P (including P1 and P2) of the present invention is suitable for the production of (meth)acrolein and/or (meth)acrylic acid.

[0035]

The reasons why the productivity of (meth)acrolein and/or (meth)acrylic acid can be increased and the running cost can be made low by using the heat transfer tube P of the embodiment according to the present invention are as follows.

[0036]

The catalyst used upon the production of (meth)acrolein and/or (meth)acrylic acid has a characteristic that the lifetime of the catalyst can be made longer when an amount of catalyst packed in the heat transfer tube P is increased.

In the case of the heat transfer tube P of the embodiment according to the present invention, even if the

distance between the gas supplying port and the catalyst layer C is shortened, the temperature of the feed gas BRG supplied into the catalyst layer C can be preheated to an appropriate temperature, whereby the length of the catalyst C can be made longer by shortening the length of the preheating layer. In other words, the amount of catalyst which is packed in the heat transfer tube P can be increased.

Thus, since the use of the heat transfer tube P of the embodiment according to the present invention makes it possible to increase the amount of the catalyst packed in the heat transfer tube P and increase the lifetime of the catalyst, the productivity of (meth)acrolein and/or (meth)acrylic acid can be improved with the running cost low.

Examples

Example 1

[0037]

The feed gas preheating effect given by the use of the heat transfer tube for multitubular reactor of the present first invention was confirmed by numerical simulation.

In the numerical simulation, the feed gas temperature change in the preheating layer was calculated by varying a diameter of the rod portion and the pitch of the spiral

portion in the case where the insert of the present invention (which has the rod portion and the spiral portion was inserted in the preheating layer of the heat transfer tube. For the purpose of comparison, the feed gas temperature change in the preheating layer was also calculated in the case where a corrugated plate was inserted as the insert.

[0038]

The simulation was conducted by steady calculation using a two-dimensional axial symmetry or a three-dimensional model by a simulation soft (Fluent Ver6.3.26), assuming that the feed gas is an incompressible fluid. As the turbulent model, k-omega (sst) model was used.

[0039]

The calculation conditions are described below.

(1) Conditions for the feed gas

(i) The feed gas was assumed to be a mixture gas of methacrolein, oxygen, nitrogen and so on, and the following values were employed as estimated data at 270 °C for properties of the gas.

Density of gas: 0.6517 kg/m^3

Viscosity of gas: 2.646×10^{-5} Pa·s

Specific heat of gas: 1188 J/(kg·K)

Thermal conductivity of gas: 0.0414 W/(m·K)

(ii) The conditions for the feed gas at inlet (that is,

at gas supplying port)

Flow rate of gas: 0.0004954 kg/s (23 NL/min)

Gas inlet temperature: 250 °C

(2) Conditions for catalyst layer

The catalyst layer was assumed to be solid having the following properties.

Density: 7930 kg/m^3

Specific heat: 590 J/(m·K)

Thermal conductivity: 16.7 W/(m·K)

(3) Conditions for heat transfer tube

Material of heat transfer tube: Stainless steel (SUS304)

Inner diameter of heat transfer tube: 25 mm

Wall thickness of heat transfer tube: 1.0 mm

Length of preheating layer: 315 mm

Wall temperature of preheating layer: 280 °C

(4) Conditions for insert

(i) Example (Working example)

The insert of example was made a construction wherein the wire rod is wound into a spiral around the side surface of the rod portion. Further, it was assumed that the insert was placed so that the central axis of the rod portion coincided with the central axis of the heat transfer tube (see Fig. 4(A)).

In this Example, the calculation was carried out for

four patterns of inserts which were different in combination of the rod portion and the spiral portion.

a. Rod portion

Material: Stainless steel (SUS 304)

Length: 215 mm

Shaft diameter d1: 11.6 mm, 13.6 mm, 14.6 mm

b. Spiral portion

Material: Stainless steel (SUS 304)

Length: 215 mm

Diameter: 5.0 mm

Pitch: 12.5 mm, 25.0 mm

(ii) Comparative Example

The insert of a comparative example had a construction of corrugated plate formed by bending a plate material. Further, it was assumed that the insert was placed so that the central axis of the insert coincided with the central axis of the heat transfer tube (see Fig. 4(B)).

Material: Stainless steel (SUS 304)

Total length: 215 mm

Width of plate material: 17.0 mm

Pitch of bending: 34.0 mm

Bending angle: 90°

It should be noted that the physical properties of the insert were those at 20 °C for both of the example and the comparative example.

[0040]

The results are shown in Fig. 5. In a graph of Fig. 5, a horizontal axis denotes a depth of the preheating layer (a distance from the gas supplying port) and a vertical axis denotes the gas temperature.

As shown in Fig. 5, the calculation was made for Case 2 (shaft diameter d1: 13.6 mm (diameter d2: 23.6 mm), pitch : 25.0 mm), Case 3 (shaft diameter d1: 13.6 mm (diameter d2: 23.6 mm), pitch: 12.5 mm), Case 4 (shaft diameter d1 : 13.6 mm (diameter d2: 24.6 mm), pitch: 25.0 mm) and Case 5 (shaft diameter d1: 13.6 mm (diameter d2: 22.6 mm), pitch: 25.0 mm) with respect to the inserts of the working examples.

In the respective cases excluding Case 5, the values obtained by dividing the projected area of the insert in the axial direction by the cross-sectional area of space of the heat transfer tube are in a range of from 0.80 to 0.99 and it can be confirmed that the temperature of the feed gas can be raised earlier in the respective cases compared to Comparative example (Case 1).

[0041]

On the other hand, in Case 5, the value obtained by dividing the projected area of the insert in the axial direction by the cross-sectional area of space of the heat transfer tube is 0.746 which is smaller than the lower

limit of the above-mentioned range and it can be confirmed that the temperature rising in Case 5 is the same as that of Comparative example.

[0042]

From the above results, it was confirmed that the insert of the present first invention wherein the wire rod is wound into a spiral around the side surface of the rod portion shows an excellent effect of raising the feed gas temperature when the value obtained by dividing the projected area of the insert in the axial direction by the cross-sectional area of space of the heat transfer tube is in the range of from 0.80 to 0.99.

Example 2

[0043]

The feed gas preheating effect which is given by the use of the heat transfer tube for multitubular reactor of the present first invention was confirmed by an experiment.

In the experiment, the change in the feed gas temperature in the preheating layer was calculated for three pattern, one being a case wherein the insert of the present first invention (the insert which has the rod portion and the spiral portion) was inserted in the preheating layer of the heat transfer layer (this working example), one being a case wherein a corrugated plate was

placed as the insert in the preheating layer (Comparative example 1), and one being a case wherein no insert was placed in the preheating layer (Comparative example 2).

[0044]

The heat transfer tube, the insert and the feed gas used in the experiment were as follows.

In the experiment, HTS flowed around the heat transfer tube and the temperature of HTS was 280 °C.

[0045]

(1) Feed gas (a mixture gas of methacrolein, oxygen, nitrogen and so on)

Flow rate of gas: 1400 NL/hr

Gas inlet temperature: 236.5 °C

(2) Heat transfer tube

Material of heat transfer tube: Stainless steel (SUS304)

Inner diameter of heat transfer tube: 25 mm

Wall thickness of heat transfer tube: 1.0 mm

Length of heat transfer tube: 3 m

Length of preheating layer: 315 mm

Wall temperature of the preheating layer: 280 °C

(3) Conditions for the insert

(i) Example (Working example)

The insert of this example was made a construction wherein the wire rod was wound into a spiral around the

side surface of the rod portion and the diameter d2 of the insert was 23 mm (see Fig. 2 and Fig. 4(A)).

a. Rod portion

Material: Stainless steel (SUS 304)

Length: 200 mm

Shaft diameter d1: 12.5 mm

b. Spiral portion

Material: Stainless steel (SUS 304)

Diameter: 5 mm

Pitch: 50 mm

(ii) Comparative example 1

The insert of Comparative example 1 was made a corrugated construction which was formed by bending a plate material (see Fig. 4(B)).

Material: Stainless steel (SUS 304)

Total length: 300 mm

Width of plate material: 17 mm

Pitch of bending: 34 mm

Bending angle: 90°

[0046]

The results are shown in Table 1 and Fig. 6. In a graph of Fig. 6, a horizontal axis denotes a depth of the preheating layer and a vertical axis denotes the gas temperature.

Table 1

Depth of Preheating Layer	Example	Comp. Ex. 1	Comp. Ex. 2
mm	°C	°C	°C
0	236.5	236.5	236.5
25	246.8		
50	254.6	245.9	238.3
75	260.6		
100	265.2	254.7	239.3
125	268.7		
150	271.4	263.6	242.1
175	273.4		
200	275.0	268.9	243.0
250		272.8	243.3
300		276.1	241.8

As shown in Fig. 6, it can be confirmed that when the insert of the working example is placed, the feed gas temperature can be raised earlier compared to the case where the insert of Comparative example 1 was placed and the case where no insert was placed.

Further, it can be confirmed that when the distances (distances from the gas supplying ports) at which the same temperature is obtained are compared, the distance can be significantly reduced in the working example compared to Comparative example 1.

[0047]

From the above results, it can be confirmed the insert of the working example wherein the wire rod is wound into a

spiral around the side surface of the rod portion is effective for raising the temperature of the feed gas earlier.

Example 3

[0048]

The feed gas preheating effect which is given by the use of the heat transfer tube for multitubular reactor of the present fifth invention was confirmed by an experiment.

In the experiment, the overall heat transfer coefficient was confirmed when the insert of the present fifth invention (the insert of a corrugated plate formed such that the value obtained by dividing the projected area of the insert in the axial direction by the cross-sectional area of space of the heat transfer tube was in a range of from 0.5 to 0.7) was inserted in the preheating layer of the heat transfer tube (Examples 3-1 and 3-2).

For the purpose of comparison, the overall heat transfer coefficient was confirmed for the case where no insert was placed (Comparative example 3-1) and the case where the insert of a conventional corrugated plate (wherein the value obtained by the projected area of the insert in the axial direction by the cross-sectional area of space in the heat transfer tube was 0.41) was placed (Comparative Example 3-2).

[0049]

The overall heat transfer coefficient is an index of a heat exchange amount per unit time and unit area, and was calculated using the following formulae 1 and 2 based on the respective experimentally-determined values of a gas inlet temperature, a gas outlet temperature, a gas flow rate, and a specific heat of gas. Specifically, Q is calculated using formula 2 and then U (overall heat exchange coefficient) is calculated using the calculated Q and formula 1.

Formula 1: $Q = UA\ln\Delta T$

Q: Heat exchange amount (kcal/hr)

U: Overall heat exchange coefficient ($kcal/hr/m^2/K$)

A: Surface area of exterior surface of heat transfer tube (m^2)

ΔT: Difference between the gas inlet temperature and the gas outlet temperature (K)

Formula 2: $Q = mc\Delta T$

m: Gas flow rate (L/min)

c: Specific heat of gas (kcal/L/K)

[0050]

The heat transfer tube, the insert and the gas which were used in the experiment are as follows. In the

experiment, HTS flowed around the heat transfer tube and the temperature of HTS was 280 °C.

[0051]

(1) Gas (air)

Flow rate of gas: 23 L/min

Gas inlet temperature: 25 °C

(2) Heat transfer tube

Material of heat transfer tube: Stainless steel (SUS 304)

Inner diameter of heat transfer tube: 30 mm

Wall thickness of heat transfer tube: 1.0 mm

Length of heat transfer tube: 3 m

Length of preheating layer: 300 mm

(3) Conditions for insert

The inserts in Examples 3-1 and 3-2 and Comparative example 3-1 were made a corrugated plate structure formed by bending the plate material (see Fig. 7).

The inserts were placed so that the central axes thereof coincided with the central axis of the heat transfer tube.

Further, in this experiment, the catalyst was not packed in the heat transfer tube. Therefore, each insert was placed on a mesh which was placed at an upper end of the catalyst layer assuming that the catalyst was packed in the heat transfer tube in all examples and comparative

example.

(i) Examples (Working examples)

a. Example 1

Material: Stainless steel (SUS 304)

Total length: 200 mm

Thickness of plate: 0.5 mm

Width of plate: 20.4 mm

Pitch of bending: 40.8 mm

Bending angle: 90°

b. Example 2

Material: Stainless steel (SUS 304)

Total length: 200 mm

Thickness of plate: 0.5 mm

Width of plate: 20.4 mm

Pitch of bending: 10.9 mm

Bending angle: 30°

(ii) Comparative example

Material: Stainless steel (SUS 304)

Total length: 300 mm

Thickness of plate: 0.5 mm

Width of plate: 17.0 mm

Pitch of bending: 24.0 mm

Bending angle: 90°

[0052]

The results are shown in Fig. 9.

As shown in Fig. 9, the overall heat transfer coefficients in Examples 3-1 and 3-2 are about four times the overall heat transfer coefficient in Comparative example 3-1. From this fact, it can be confirmed that the insertion of the insert of the present fifth invention can significantly increase the overall heat coefficient. Further, the overall heat transfer coefficients in Examples 3-1 and 3-2 are about 1.5 times or more the overall heat transfer coefficient in Comparative example 3-2.

From the results, the inserts of these working examples in Example 3 are very effective for raising the temperature of the feed gas earlier since the insertion of the inserts of these working examples into the preheating layer can significantly increase the overall heat transfer coefficient.

Example 4

[0053]

The feed gas preheating effect by the use of the heat transfer tube for multitubular reactor of the present fifth invention was confirmed by numerical simulation.

In the numerical simulation, the feed gas temperature change in the preheating layer was calculated in the case where the insert of the present fifth invention (which was a corrugated plate formed such that the value obtained by

dividing the projected area of the insert in the axial direction by the cross-sectional area of space of the heat transfer tube is in a range of from 0.5 to 0.7) was inserted in the preheating layer of the heat transfer tube. For the purpose of comparison, the feed gas temperature change in the preheating layer was calculated in the case where no insert was inserted (Comparative example 4-1) and the case where a conventional corrugated plate (wherein the value obtained by dividing the projected area of the heat transfer tube in the axial direction by the cross-sectional area of space of the heat transfer tube was 0.41) was placed (Comparative example 4-2).

[0054]

The simulation was conducted by steady calculation using a two-dimensional axial symmetry or a three-dimensional model by a simulation soft (Fluent Ver6.3.26), assuming that the feed gas is an incompressible fluid. As the turbulent model, k-omega (SST) model was used.

[0055]

The other calculation conditions are described below.

(1) Conditions for the feed gas

(i) The feed gas was assumed to be a mixture gas of methacrolein, nitrogen, oxygen and so on, and the following values were employed as estimated data at 270 °C for properties of the gas.

Density of gas: 0.6517 kg/m^3

Viscosity of gas: 2.646×10^{-5} Pa·s

Specific heat of gas: 1188 J/(kg·K)

Thermal conductivity of gas: 0.0414 W/(m·K)

(ii) The conditions for the feed gas at inlet (that is, at gas supplying port)

Flow rate of gas: 1400 NL/hr

Gas inlet temperature: 254 °C

(2) Conditions for catalyst layer

Density: 7930 kg/m^3

Specific heat: 590 J/(m·K)

Thermal conductivity: 16.7 W/(m·K)

(3) Conditions for heat transfer tube

Material of heat transfer tube: Stainless steel (SUS304)

Inner diameter of heat transfer tube: 25 mm

Wall thickness of heat transfer tube: 1.0 mm

Length of preheating layer: 315 mm

Wall temperature of the preheating layer: 280 °C

(4) Conditions for insert

The inserts were made a corrugated structure formed by bending a plate member in Examples 4-1 and 4-2 and Comparative example 4-1 (see Fig. 10). Further, it was assumed that the inserts were placed so that the central axes thereof coincided with the central axis of the heat

transfer tube and the properties of the insert were those at 20 °C for the examples and the comparative example.

(i) Examples (Working examples)

a. Example 4-1

Material: Stainless steel (SUS 304)

Total length: 200 mm

Thickness of plate: 0.5 mm

Width of plate: 20.4 mm

Pitch of bending: 40.8 mm

Bending angle: 90°

b. Example 4-2

Material: Stainless steel (SUS 304)

Total length: 200 mm

Thickness of plate: 0.5 mm

Width of plate: 20.4 mm

Pitch of bending: 10.9 mm

Bending angle: 30°

(ii) Comparative example 4-1

Material: Stainless steel (SUS 304)

Total length: 300 mm

Thickness of plate: 0.5 mm

Width of plate: 17.0 mm

Pitch of bending: 34.0 mm

Bending angle: 90°

[0056]

The results are shown in Table 2 and Fig. 11. In a graph of Fig. 11, a horizontal axis denotes a depth of the preheating layer (a distance from the gas supplying port) and a vertical axis denotes the gas temperature.

Table 2

Depth of Preheating Layer	Comp. Ex. 4-1	Comp. Ex. 4-2	Example 4-1	Example 4-2
mm	°C	°C	°C	°C
0	254.0	254.0	254.0	254.0
50	256.0	258.0	260.2	260.7
100	257.8	261.4	264.7	265.4
150	259.4	264.1	268.0	268.8
200	260.9	266.4	270.4	271.1
250	262.3	268.3	272.2	272.8
300	263.5	269.8	273.5	274.0

As shown in Fig. 6, in Examples 4-1 and 4-2, the feed gas temperature can be raised to the same temperature at about one third of the depth (100 mm) as that at 300 mm in Comparative example 4-1. Further, when comparing the examples with Comparative example 4-2, the feed gas temperature can be raised to the same temperature at about two third of the depth (200 mm) as that at 300 mm in Comparative example 4-2.

From the results, it is confirmed that the feed gas temperature can be raised earlier by disposing the insert of the present fifth invention in the preheating layer.

[0057]

From the results, it can be confirmed that when the insert is configured zigzag such that the value obtained by dividing the projected area of the insert in the axial direction by the cross-sectional area of space of the heat transfer tube is from 0.5 to 0.7, the effect of raising the feed gas temperature is high.

Industrial Applicability

[0058]

The heat transfer tube for multitubular reactor according to the present invention is suitable for the heat transfer tube for the fixed-bed reactor wherein the objective substance is produced using the fixed-bed catalyst (such as a catalyst used for producing (meth)acrolein and/or (meth)acrylic acid) of which life time can be longer by increasing the amount of the catalyst packed into the heat transfer tube.

CLAIMS

1. A heat transfer tube for a multitubular reactor comprising a gas supplying port at one end and a gas discharging port at the other end and a catalyst layer packed between the both ends, wherein

an insert extending in an axial direction of the heat transfer tube is placed between the gas supplying port and the catalyst layer, and

the insert comprises a rod portion and a spiral portion which is provided on a side surface of the rod portion.

2. The heat transfer tube for a multitubular reactor according to claim 1, wherein the insert is configured so that a value obtained by dividing a projected area of the insert in the axial direction of the insert by a cross-sectional area of space of the heat transfer tube is from 0.80 to 0.99.

3. The heat transfer tube for a multitubular reactor according to claim 1, wherein a pitch of the spiral potion of the insert is 50 mm or less.

4. The heat transfer tube for a multitubular reactor

according to any one of claims 1 to 3, which is used in a fixed-bed reactor for producing (meth)acrolein and/or (meth)acrylic acid by oxidizing propylene, isobutylene, t-butyl alcohol or a mixture thereof in a gas phase with a gas containing molecular oxygen.

5. A heat transfer tube for a multitubular reactor comprising a gas supplying port at one end and a gas discharging port at the other end and a catalyst layer packed between the both ends, wherein

an insert extending in an axial direction of the heat transfer tube is placed between the gas supplying port and the catalyst layer,

the insert is of a shape of zigzag sequence of plate portions having sloped surfaces inclined relative to the axial direction of the heat transfer tube, and

the insert is configured such that a value obtained by dividing a projected area of the insert in the axial direction of the insert by a cross sectional area of space of the heat transfer tube is from 0.5 to 0.7.

6. The heat transfer tube for a multitubular reactor according to claim 5, wherein the insert is formed by bending a plate material.

7. The heat transfer tube for a multitubular reactor according to claim 5, wherein an angle formed by adjacent plate portions is from 20° to 100°.

8. The heat transfer tube for a multitubular reactor according to any one of claims 5 to 7, which is used in a fixed-bed reactor for producing (meth)acrolein and/or (meth)acrylic acid by oxidizing propylene, isobutylene, t-butyl alcohol or a mixture thereof in a gas phase with a gas containing molecular oxygen.