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(54) Titre : METHODE DE MISE EN PLACE D'UN CATALYSEUR D'EPOXYDATION DANS UN REACTEUR, METHODE DE PREPARATION D'UN OXYDE OLEFINIQUE OU D'UN PRODUIT CHIMIQUE DERIVE D'UN OXYDE OLEFINIQUE, ET REACTEUR PERMETTANT D'EFFECTUER LADITE METHODE DE PREPARATION
(54) Title: A METHOD OF INSTALLING AN EPOXIDATION CATALYST IN A REACTOR, A PROCESS FOR THE PREPARATION OF AN OLEFIN OXIDE OR A CHEMICAL DERIVABLE FROM AN OLEFIN OXIDE, AND A REACTOR SUITABLE FOR SUCH A PROCESS

(57) **Abrégé/Abstract:**

The present invention relates to an improved epoxidation process and an improved epoxidation reactor. The present invention makes use of a reactor which comprises a plurality of microchannels. Such process microchannels may be adapted such that the epoxidation and optionally other processes can take place in the microchannels and that they are in a heat exchange relation with channels adapted to contain a heat exchange fluid. A reactor comprising such process microchannels is referred to as a "microchannel reactor". The invention provides a method of installing an epoxidation catalyst in a microchannel reactor. The invention also provides a process for the epoxidation of an olefin and a process for the preparation of a chemical derivable from an olefin oxide. The invention also provides a microchannel reactor.

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5 A METHOD OF INSTALLING AN EPOXIDATION CATALYST IN A REACTOR,
A PROCESS FOR THE PREPARATION OF AN OLEFIN OXIDE OR A
CHEMICAL DERIVABLE FROM AN OLEFIN OXIDE, AND A REACTOR
SUITABLE FOR SUCH A PROCESS

Abstract of the Invention

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epoxidation reactor. The present invention makes use of a reactor which comprises a
plurality of microchannels. Such process microchannels may be adapted such that the
epoxidation and optionally other processes can take place in the microchannels and
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exchange fluid. A reactor comprising such process microchannels is referred to as a
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Field of the Invention

10 The invention relates to a method of installing an epoxidation catalyst in a reactor. The invention also relates to a process for the epoxidation of an olefin. The invention also relates to a process for the preparation of a chemical derivable from an olefin oxide. In particular, such a chemical may be a 1,2-diol, a 1,2-diol ether, a 1,2-carbonate or an alkanol amine. The invention also relates to a reactor which is suitable for use in such a process.

15 Background of the Invention

Ethylene oxide and other olefin oxides are important industrial chemicals used as a feedstock for making such chemicals as ethylene glycol, propylene glycol, ethylene glycol ethers, ethylene carbonate, ethanol amines and detergents. One method for manufacturing an olefin oxide is by olefin epoxidation, that is the catalyzed partial oxidation of the olefin with oxygen yielding the olefin oxide. The olefin oxide so manufactured may be reacted with water, an alcohol, carbon dioxide, or an amine to produce a 1,2-diol, a 1,2-diol ether, a 1,2-carbonate or an alkanol amine. Such production of a 1,2-diol, a 1,2-diol ether, a 1,2-carbonate or an alkanol amine is generally carried out separately from the manufacture of the olefin oxide, in any case the two processes are normally carried out in separate reactors.

25 In olefin epoxidation, a feed containing the olefin and oxygen is passed over a bed of catalyst contained within a reaction zone that is maintained at certain reaction conditions. A commercial epoxidation reactor is generally in the form of a shell-and-tube heat exchanger, in which a plurality of substantially parallel elongated, relatively narrow tubes are filled with shaped catalyst particles to form a packed bed, and in which the shell contains a coolant. Irrespective of the type of epoxidation catalyst used, in commercial operation the internal tube diameter is frequently in the range of from 20 to 40 mm, and the number of tubes per reactor may range in the thousands, for example up to 12,000.

35 Olefin epoxidation is generally carried out with a relatively low olefin conversion and oxygen conversion. Recycle of unconverted olefin and oxygen is

normally applied in order to enhance the economics of the process. Generally the feed additionally comprises a large quantity of so-called ballast gas to facilitate operation outside the explosion limits. Ballast gas includes saturated hydrocarbons, in particular methane and ethane. As a consequence, recycling generally involves the handling of large quantities of process streams, which includes the unconverted olefin, unconverted oxygen and the ballast gas. The processing of the recycle stream as normally applied in an olefin epoxidation plant is also fairly complex, as it involves olefin oxide recovery, carbon dioxide removal, water removal and re-pressurizing. The use of ballast gas not only contributes to the cost of processing, it also reduces the epoxidation reaction rate.

The epoxidation catalyst generally contains the catalytically active species, typically a Group 11 metal (in particular silver) and promoter components, on a shaped carrier material. Shaped carrier materials are generally carefully selected to meet requirements of, for example, strength and resistance against abrasion, surface area and porosity. The shaped carrier materials are generally manufactured by sintering selected inorganic materials to the extent that they have the desired properties.

During the epoxidation, the catalyst is subject to a performance decline, which represents itself by a loss in activity of the catalyst and selectivity in the formation the desired olefin oxide. In response to the loss of activity, the epoxidation reaction temperature may be increased such that the production rate of the olefin oxide is maintained. The operation of commercial reactors is normally limited with respect to the reaction temperature and when the applicable temperature limit has been reached, the production of the olefin oxide has to be interrupted for an exchange of the existing charge of epoxidation catalyst for a fresh charge.

It would be of great value if improved epoxidation processes and improved epoxidation reactors would become available.

Summary of the Invention

The present invention provides such improved epoxidation processes and improved epoxidation reactors. Embodiments of the present invention make use of a reactor which comprises a plurality of microchannels ("process microchannels" hereinafter). The process microchannels may be adapted such that the epoxidation and optionally other processes can take place in the microchannels and that they are in a heat exchange relation with channels adapted to contain a heat exchange fluid ("heat

exchange channels” hereinafter). A reactor comprising process microchannels is referred to herein by using the term “microchannel reactor”. As used herein, the term “Group 11” refers to Group 11 of the Periodic Table of the Elements.

In an embodiment, the present invention provides a method of installing an
5 epoxidation catalyst in one or more process microchannels of a microchannel reactor, which method comprises
depositing a Group 11 metal or a cationic Group 11 metal component on at least a portion of the walls of the said process microchannels,
depositing one or more promoter components on at least a portion of the same walls prior
10 to, together with or subsequent to the deposition of the Group 11 metal or the cationic Group 11 metal component, and,
if a cationic Group 11 metal component is deposited, reducing at least a portion of the cationic Group 11 metal component.

In another embodiment, the invention provides a process for the epoxidation of an
15 olefin comprising
- installing an epoxidation catalyst in one or more process microchannels of a microchannel reactor by
depositing a Group 11 metal or a cationic Group 11 metal component on at least a portion of the walls of the said process microchannels;
20 depositing one or more promoter components on at least a portion of the same walls prior to, together with or subsequent to the deposition of the Group 11 metal or the cationic Group 11 metal component; and,
if a cationic Group 11 metal component is deposited, reducing at least a portion of the cationic Group 11 metal component, and
25 - reacting a feed comprising the olefin and oxygen in the presence of the epoxidation catalyst installed in the one or more process microchannels.

In another embodiment, the invention provides a process for the preparation of a 1,2-diol, a 1,2-diol ether, a 1,2-carbonate or an alkanol amine, which process comprises
- installing an epoxidation catalyst in one or more process microchannels of a
30 microchannel reactor by
depositing a Group 11 metal or a cationic Group 11 metal component on at least a portion of the walls of the said process microchannels;

depositing one or more promoter components on at least a portion of the same walls prior to, together with or subsequent to the deposition of the Group 11 metal or the cationic Group 11 metal component; and,

if a cationic Group 11 metal component is deposited, reducing at least a portion of the cationic Group 11 metal component,

- reacting a feed comprising the olefin and oxygen in the presence of the epoxidation catalyst installed in the one or more process microchannels to produce an olefin oxide, and

- converting the olefin oxide with water, an alcohol, carbon dioxide or an amine to form the 1,2-diol, 1,2-diol ether, 1,2-carbonate or alkanol amine.

In another embodiment, the invention provides a reactor suitable for the epoxidation of an olefin, which reactor is a microchannel reactor comprising one or more process microchannels having installed therein an epoxidation catalyst which comprises a Group 11 metal, wherein the quantity of the Group 11 metal is in the range of from 10 to 500 kg/m³ reactor volume, reactor volume being the total volume defined by the cross sectional area and the total length of the portions of the microchannels occupied by the epoxidation catalyst.

Description of the Drawings

FIG. 1 shows a schematic of a microchannel reactor and its main constituents.

FIG. 2 shows a schematic of a typical example of a repeating unit which comprises process microchannels and heat exchange channels and its operation when in use in the practice of the invention. A microchannel reactor of this invention may comprise a plurality of such repeating units.

Detailed Description of the Invention

The use of a microchannel reactor having a catalyst installed therein in accordance with this invention leads to one or more of the following advantages:

- the epoxidation catalyst does not involve the use a shaped carrier, which can eliminate the need for a step for producing a shaped carrier.

- quenching of the olefin oxide inside the process microchannel enables operation under conditions which may be within explosion limits when such conditions would be applied in a conventional shell-and-tube heat exchanger reactor. Such conditions may be achieved by contacting an oxygen rich feed component with an olefin rich feed component within the process microchannels, which oxygen rich feed component and olefin rich feed component are normally outside the explosion limits.

Quenching inside the process microchannels also decreases the formation of byproducts, such as aldehydes and carboxylic acids.

- 5 - the epoxidation within the process microchannels can advantageously be carried out at conditions of high total concentration of the olefin, oxygen and the olefin oxide, which can lead to a higher epoxidation rate and/or lower epoxidation reaction temperature. Lowering the epoxidation reaction temperature can lead to improved selectivity and improved catalyst life. Employing conditions of high total concentration of the olefin, oxygen and the olefin oxide can also eliminate the need of using a ballast gas, which provides more efficient processing and reduction of the costs of recycling.
- 10 - the epoxidation carried out in process microchannels may be operated at a high conversion level of oxygen or the olefin oxide. In particular when the process is carried out at a high olefin conversion level, it is advantageous to operate the epoxidation process in once-through operation, which implies that no recycle stream is applied. In addition, it is advantageous that in such case air may be fed to the process microchannels, instead of oxygen separated from air, which can eliminate the need for an air separation unit.
- 15 - carrying out the olefin epoxidation inside the process microchannels enables quenching inside the same process microchannels and conversion of the co-formed carbon dioxide with at least a portion of the produced olefin oxide, and optionally condensing a liquid, typically aqueous, mixture comprising unconverted olefin oxide and the 1,2-carbonate. In respect of its composition, a remaining gaseous stream which may comprise unconverted ethylene and oxygen is suitable for recycle. This can reduce the complexity of the further processing of product and recycle streams, eliminating the need for, for example, an olefin oxide recovering unit and a carbon dioxide removal unit.
- 20 - carrying out the olefin epoxidation inside the process microchannels enables conversion of the formed olefin oxide inside the same process microchannels to 1,2-diol, 1,2-diol ether, 1,2-carbonate or alkanol amine. This can eliminate the need for additional reactors for such further conversion. It can also eliminate the need for an olefin oxide recovering unit and/or a carbon dioxide removal unit, and it can reduce the need for heat exchanging equipment. Hence, it can reduce the complexity of the additional processing conventionally applied in a manufacturing plant, for example for product recovery. Conversion of the olefin oxide inside the process

microchannels also decreases the formation of byproducts, such as aldehydes and carboxylic acids.

Microchannel reactors suitable for use in this invention and their operation have been described in WO-A-2004/099113, WO-A-01/12312, WO-01/54812, 5 US-A-6440895, US-A-6284217, US-A-6451864, US-A-6491880, US-A-6666909, US-6811829, US-A-6851171, US-A-6494614, US-A-6228434 and US-A-6192596, which are incorporated herein by reference. Methods by which the microchannel reactor may be manufactured, loaded with catalyst and operated, as described in these references, may generally be applicable in the practice of the present invention.

10 With reference to FIG. 1, microchannel reactor 100 may be comprised of a process header 102, a plurality of process microchannels 104, and a process footer 108. The process header 102 provides a passageway for fluid to flow into the process microchannels 104. The process footer 108 provides a passageway for fluid to flow from the process microchannels 104.

15 The number of process microchannels contained in a microchannel reactor may be very large. For example, the number may be up to 10^5 , or even up to 10^6 or up to 2×10^6 . Normally, the number of process microchannels may be at least 10 or at least 100, or even at least 1000.

20 The process microchannels are typically arranged in parallel, for example they may form an array of planar microchannels. The process microchannels may have at least one internal dimension of height or width of up to 15 mm, for example from 0.05 to 10 mm, in particular from 0.1 to 5 mm, more in particular from 0.5 to 2 mm. The other internal dimension of height or width may be, for example, from 0.1 to 100 cm, in particular from 0.2 to 75 cm, more in particular from 0.3 to 50 cm. The length of 25 the process microchannels may be, for example, from 1 to 500 cm, in particular from 2 to 300 cm, more in particular from 3 to 200 cm, or from 5 to 100 cm.

The microchannel reactor 100 additionally comprises heat exchange channels (not shown in FIG. 1) which are in heat exchange contact with the process microchannels 104. The heat exchange channels may also be microchannels. The 30 microchannel reactor is adapted such that heat exchange fluid can flow from heat exchange header 110 through the heat exchange channels to heat exchange footer 112. The heat exchange channels may be aligned to provide a flow in a co-current, counter-current or, preferably, cross-current direction, relative to a flow in the process

microchannels 104. The cross-current direction is as indicated by arrows 114 and 116.

The heat exchange channels may have at least one internal dimension of height or width of up to 15 mm, for example from 0.05 to 10 mm, in particular from 0.1 to 5 mm, more in particular from 0.5 to 2 mm. The other internal dimension of height or width may be, for example, from 0.1 to 100 cm, in particular from 0.2 to 75 cm, more in particular from 0.3 to 50 cm. The length of the heat exchange channels may be, for example, from 1 to 500 cm, in particular from 2 to 300 cm, more in particular from 3 to 200 cm, or from 5 to 100 cm.

The separation between a process microchannel 104 and the next adjacent heat exchange channel may be in the range of from 0.05 mm to 5 mm, in particular from 0.2 to 2 mm.

In some embodiments of this invention, there is provided for first heat exchange channels and second heat exchange channels, or first heat exchange channels, second heat exchange channels and third heat exchange channels, or even up to fifth heat exchange channels, or even further heat exchange channels. Thus, in such cases, there is a plurality of sets of heat exchange channels, and accordingly there may be a plurality of heat exchange headers 110 and heat exchange footers 112, whereby the sets of heat exchange channels may be adapted to receive heat exchange fluid from a heat exchange header 110 and to deliver heat exchange fluid into a heat exchange footer 112.

The process header 102, process footer 108, heat exchange header 110, heat exchange footer 112, process microchannels 104 and heat exchange channels may independently be made of any construction material which provides sufficient strength, dimensional stability and heat transfer characteristics to permit operation of the processes in accordance with this invention. Suitable construction materials include, for example, steel (for example stainless steel and carbon steel), monel, titanium, copper, glass and polymer compositions. The kind of heat exchange fluid is not material to the present invention and the heat exchange fluid may be selected from a large variety. Suitable heat exchange fluids include steam, water, air and oils. In embodiments of the invention which include a plurality of sets of heat exchange channels, such sets of heat exchange channels may operate with different heat exchange fluids or with heat exchange fluids having different temperatures.

A microchannel reactor according to the invention may comprise a plurality of repeating units comprising one or more process microchannels and one or more heat exchange channels. Reference is now made to FIG. 2, which shows a typical repeating unit and its operation.

5 Process microchannels 210 have an upstream end 220 and a downstream end 230 and may comprise of a first section 240 which may contain a catalyst (not drawn), for example an epoxidation catalyst. First section 240 may be in heat exchange contact with first heat exchange channel 250, allowing heat exchange between first section 240 of process microchannel 210 and first heat exchange channel 250. The
10 repeating unit may comprise first feed channel 260 which ends into first section 240 through one or more first orifices 280. Typically one or more first orifices 280 may be positioned downstream relative to another first orifice 280. During operation, feed comprising the olefin and oxygen may enter into first section 240 of process microchannel 210 through an opening in upstream end 220 and/or through first feed
15 channel 260 and one or more first orifices 280.

 Process microchannels 210 may comprise a second section 340 which may or may not be adapted to contain a catalyst. Second section 340 may or may not contain a catalyst, as described herein. Second section 340 is positioned downstream of first section 240. Second section 340 may be in heat exchange contact with second heat
20 exchange channel 350, allowing heat exchange between second section 340 of process microchannel 210 and second heat exchange channel 350. In some embodiments second section 340 is adapted to quench olefin oxide obtained in and received from first section 240 by heat exchange with a heat exchange fluid in second heat exchange channel 350. Quenching may be achieved in one or more stages by the presence of a
25 plurality of second heat exchange channels 350, for example two or three or four. Such a plurality of second heat exchange channels 350 may be adapted to contain heat exchange fluids having different temperatures, in particular such that in downstream direction of second section 340 heat exchange takes place with a second heat exchange channel 350 containing a heat exchange fluid having a lower temperature.
30 The repeating unit may comprise second feed channel 360 which ends into second section 340 through one or more second orifices 380. During operation, feed may enter into second section 340 from upstream in process microchannel 210 and through second feed channel 360 and one or more second orifices 380. Typically one or more second orifices 380 may be positioned downstream relative to another second orifice

380. In embodiments in which second section 340 is adapted for accommodating conversion of olefin oxide to 1,2-diol, 1,2-diol ether, 1,2-carbonate or alkanol amine, feed entering during operation through second feed channel 360 and one or more second orifices 380 may comprise water, the alcohol, carbon dioxide or the amine.

5 Also, catalyst may be fed through second feed channel 360 and one or more second orifices 380. If desirable, a separate set of second feed channel (not drawn) with one or more second orifices (not drawn) may be present in order to accommodate separate feeding of feed and catalyst.

10 The first and second feed channels 260 or 360 in combination with first and second orifices 280 or 380, whereby one or more first or second orifices 280 or 380 are positioned downstream to another first or second orifice 280 or 380, respectively, allow for replenishment of a reactant. Replenishment of a reactant is a feature in some embodiments of this invention.

15 Process microchannels 210 may comprise an intermediate section 440, which is positioned downstream of first section 240 and upstream of second section 340. Intermediate section 440 may be in heat exchange contact with third heat exchange channel 450, allowing heat exchange between intermediate section 440 of process microchannel 210 and third heat exchange channel 450. In some embodiments, for example in embodiments in which second section 340 is adapted for accommodating
20 conversion of olefin oxide to 1,2-diol, 1,2-diol ether, 1,2-carbonate or alkanol amine, intermediate section 440 is adapted to quench olefin oxide obtained in and received from first section 240 by heat exchange with a heat exchange fluid in third heat exchange channel 450. Quenching may be achieved in stages by the presence of a plurality of
25 third heat exchange channels 450, for example two or three or four. Such a plurality of third heat exchange channels 450 may be adapted to contain heat exchange fluids having different temperatures, in particular such that in downstream direction of intermediate section 440 heat exchange takes place with a third heat exchange channel 450 containing a heat exchange fluid having a lower temperature.

30 The feed channels may be microchannels. They may have at least one internal dimension of height or width of up to 15 mm, for example from 0.05 to 10 mm, in particular from 0.1 to 5 mm, more in particular from 0.5 to 2 mm. The other internal dimension of height or width may be, for example, from 0.1 to 100 cm, in particular from 0.2 to 75 cm, more in particular from 0.3 to 50 cm. The length of the feed

channels may be, for example, from 1 to 250 cm, in particular from 2 to 150 cm, and more particularly from 3 to 100 cm, or from 5 to 50 cm.

The length of the sections of the process microchannels may be selected independently of each other, in accordance with, for example, the heat exchange capacity needed or the quantity of catalyst which may be contained in the section. The lengths of the sections are preferably at least 1 cm, or at least 2 cm, or at least 5 cm. The lengths of the sections are preferably at most 250 cm, or at most 150 cm, or at most 100 cm, or at most 50 cm. Other dimensions of the sections are dictated by the corresponding dimensions of process microchannel 210.

The microchannel reactor of this invention may be manufactured using known techniques, for example conventional machining, laser cutting, molding, stamping and etching and combinations thereof. The microchannel reactor of this invention may be manufactured by forming sheets with features removed which allow passages. A stack of such sheets may be assembled to form an integrated device, by using known techniques, for example diffusion bonding, laser welding, cold welding, diffusion brazing, and combinations thereof. The microchannel reactor of this invention comprises appropriate headers, footers, valves, conduit lines, and other features to control input of reactants, output of product, and flow of heat exchange fluids. These are not shown in the drawings, but they can be readily provided by those skilled in the art. Also, there may be further heat exchange equipment (not shown in the drawings) for temperature control of feed, in particular for heating feed or feed components, before it enters the process microchannels, or for temperature control of product, in particular for quenching product, after it has left the process microchannels. Such further heat exchange equipment may be integral with the microchannel reactor, but more typically it will be separate equipment. These are not shown in the drawings, but they can be readily provided by those skilled in the art. Heat integration may be applied, for example by using reaction heat of the epoxidation process for heating feed components, or for other heating purposes.

Typically, the epoxidation catalysts are solid catalysts under the conditions of the epoxidation reaction. The catalysts may form a coating on at least a portion of the wall of the designated section of the process microchannels. The skilled person will understand that the coating will be positioned on the interior wall of the process microchannels. Additionally, one or more of the catalysts may be in the form of a

coating on inserts which may be placed in the designated section of the process microchannels.

The epoxidation catalyst which may be used in this invention is typically a catalyst which comprises one or more Group 11 metals. The Group 11 metals may be selected from the group consisting of silver and gold. Preferably, the Group 11 metal comprises silver. In particular, the Group 11 metal comprises silver in a quantity of at least 90 %w, more in particular at least 95 %w, for example at least 99 %w, or at least 99.5 %w, calculated as the weight of silver metal relative to the total weight of the Group 11 metal, as metal. Typically, the epoxidation catalyst additionally comprises one or more promoter components. More typically, the epoxidation catalyst comprises the Group 11 metal, one or more promoter components and additionally one or more components comprising one or more further elements. In some embodiments, the epoxidation catalyst may comprise a carrier material on which the Group 11 metal, any promoter components and any components comprising one or more further elements may be deposited. Suitable promoter components and suitable components comprising one or more further elements and suitable carrier materials may be as described hereinafter.

In an embodiment, the present invention provides a method of installing an epoxidation catalyst in one or more process microchannels of a microchannel reactor, which method comprises depositing one or more Group 11 metals or one or more cationic Group 11 metal components on at least a portion of the walls of the said process microchannels, depositing one or more promoter components on at least the same walls prior to, together with or subsequent to the deposition of the Group 11 metal(s) or the cationic Group 11 metal component(s), and, if a cationic Group 11 metal component is deposited, reducing at least a portion of the cationic Group 11 metal component(s).

Group 11 metal may be deposited on at least a portion of the walls of the process microchannels by contacting the walls with a liquid containing dispersed Group 11 metal, for example a Group 11 metal sol, and removing the liquid, for example by evaporation, while leaving Group 11 metal on the wall. Such deposition may be carried out more than once, for example two times or three times, to accomplish the deposition of a desired amount of Group 11 metal. The quantity of Group 11 metal in such liquid may be in the range of from 1 to 30 %w, in particular from 2 to 15 %w, relative to the weight of the liquid. The liquid may comprise additives, such as dispersants and stabilizers. Such additives may be removed after

the removal of the liquid, by heating for example at a temperature of from 100 to 300 °C, in particular from 150 to 250 °C, in an inert atmosphere, for example in nitrogen or argon, or in an oxygen containing atmosphere, for example air or a mixture comprising oxygen and argon.

5 As an alternative, or in addition, Group 11 metal may be deposited on at least a portion of the walls of the process microchannels by vapor deposition techniques known in the art.

 A cationic Group 11 metal component may be deposited on at least a portion of the walls of the process microchannels by contacting the walls with a liquid
10 mixture comprising the cationic Group 11 metal component, and removing a liquid component of the liquid mixture. A reducing agent may be applied prior to, together with or after the deposition of cationic Group 11 metal component. Typically, the liquid mixture may comprise the cationic Group 11 metal component and a reducing agent, in which case removing the liquid and performing reduction of at least a
15 portion of the cationic Group 11 metal component may be accomplished simultaneously. Such deposition may be carried out more than once, for example two times or three times, to accomplish the deposition of a desired amount of Group 11 metal. The cationic Group 11 metal component includes, for example a non-complexed or complexed Group 11 metal salt, in particular, a cationic Group 11
20 metal-amine complex. Contacting the walls with a liquid mixture comprising a cationic Group 11 metal-amine complex and a reducing agent may be followed by heating at a temperature of from 100 to 300 °C, in particular from 150 to 250 °C, in an inert atmosphere, for example in nitrogen or argon, or in an oxygen containing atmosphere, for example air or a mixture comprising oxygen and argon. The heating
25 will, in general, effect the reduction of at least a portion of the cationic Group 11 metal-amine complex. Examples of cationic Group 11 metal-amine complexes are cationic Group 11 metal complexed with a monoamine or a diamine, in particular a 1,2-alkylene diamine. Examples of suitable amines are ethylene diamine, 1,2-propylene diamine, 2,3-butylene diamine, and ethanol amine. Higher amines may be
30 used, such as, for example, triamines, tetraamines, and pentaamines. Examples of reducing agents are oxalates, lactates and formaldehyde. The quantity of Group 11 metal in such liquid mixture may be in the range of from 1 to 40 %w, in particular from 2 to 30 %w, calculated as the weight of the Group 11 metal relative to the weight of the liquid mixture. For further particulars of liquid mixtures comprising

cationic Group 11 metal-amine complex and a reducing agent, reference may be made to US-A-5380697, US-A-5739075, EP-A-266015, and US-B-6368998, which are incorporated herein by reference.

In some embodiments, Group 11 metal or cationic Group 11 metal component
5 may be deposited on at least a portion of the walls of the process microchannels before the microchannel reactor is manufactured by assembling sheets, as described hereinbefore. In such embodiments portions of the walls on which no Group 11 metal is to be deposited may be shielded by a temporary coating. In other embodiments, Group 11 metal or cationic Group 11 metal component may be deposited on at least a
10 portion of the walls of the process microchannels after they have been formed by assembling the sheets described hereinbefore. In such embodiments, inserts may be placed temporarily in the sections of the microchannels where no Group 11 metal is to be deposited on the walls.

In some embodiments, Group 11 metal or cationic Group 11 metal component
15 may be deposited on at least a portion of the walls of the process microchannels wherein the said walls are at least partly covered with a carrier material, and Group 11 metal or cationic Group 11 metal component is deposited on or in the carrier material, suitably by using an impregnation method. The said walls may be at least partly covered with the carrier material by wash coating, prior to or after assembling the
20 process microchannels. Particulars of suitable carrier materials are as specified hereinafter.

In some embodiments, the walls of the process microchannels on which Group
11 metal or cationic Group 11 metal component may be deposited are at least partly roughened or corrugated. Roughening or corrugation may provide grooves and
25 elevations, so that the roughened or corrugated wall surface is effectively enlarged, for example, by a factor of from 0.5 to 10, or from 1 to 5, relative to the surface area of the roughened or corrugated wall surface as defined by its outer dimensions. This can increase the adhesion of the epoxidation catalyst deposited on the wall, and it will effect that more epoxidation catalyst surface can contribute in catalyzing the
30 epoxidation reaction. Roughening and corrugation may be achieved by methods known in the art, for example by etching or by applying abrasive power.

In some embodiments, the said deposition of Group 11 metal or cationic Group 11 metal component, with subsequent reduction, will yield a Group 11 metal mirror positioned on the walls of the process microchannels, and in other

embodiments this will yield discrete Group 11 metal particles, for example in the form of spheres. In yet other embodiments, a combination of a mirror and discrete particles will be yielded. Such morphology differences are not essential in the practice of the present invention.

5 One or more promoter components may be deposited on at least a portion of the same walls of the process microchannels as the walls on which Group 11 metal or cationic Group 11 metal component is deposited. The deposition of promoter components may be effected prior to, together with or subsequent to the deposition of Group 11 metal or cationic Group 11 metal component. Particulars of such promoter
10 components, including suitable quantities thereof, are disclosed hereinafter. Suitable methods of depositing the promoter components may include, for example, contacting the walls with a liquid mixture comprising one or more of the promoter components to be deposited and a diluent, and removing the diluent while leaving at least a portion of the promoter component(s). In particular in embodiments in which the walls of the
15 process microchannels are covered with a carrier materials, the liquid mixture may be kept in contact with the walls for a period of time before removing the diluent, for example for up to 10 hours, in particular for 0.25 to 5 hours, and the temperature may be up to 95 °C, in particular in the range of from 10 to 80 °C. Suitable liquids typically comprise the promoter component(s) dissolved or dispersed in an aqueous
20 liquid, for example water or an aqueous organic diluent, such as for example a mixture of water and one or more of methanol, ethanol, propanol, isopropanol, acetone or methyl ethyl ketone. The deposition may be carried out more than once, for example two times or three times, to accomplish the deposition of a desired amount of promoter component. Alternatively, different promoter components may
25 be deposited in different deposition steps.

In addition to one or more promoter components, one or more components comprising one or more further elements may be deposited on at least a portion of the same walls of the process microchannels as the walls on which Group 11 metal or cationic Group 11 metal component is deposited. The deposition of components
30 comprising the further elements may be effected prior to, together with or subsequent to the deposition of Group 11 metal or cationic Group 11 metal component, and prior to, together with or subsequent to the deposition of the promoter components. Particulars of the components comprising the further elements, including suitable quantities thereof, are disclosed hereinafter. Suitable methods of depositing the components comprising the

further elements include, for example, contacting the walls with a liquid mixture comprising one or more of the components to be deposited and a diluent, and removing the diluent while leaving at least a portion of the component(s). In particular in embodiments in which the walls of the process microchannels are covered with a carrier materials, the liquid mixture may be kept in contact with the walls for a period of time before removing the diluent, for example for up to 10 hours, in particular for 0.25 to 5 hours, and the temperature may be up to 95 °C, in particular in the range of from 10 to 80 °C. Suitable liquids typically comprise the component(s) dissolved or dispersed in an aqueous liquid, for example water or an aqueous organic diluent, such as for example a mixture of water and one or more of methanol, ethanol, propanol, isopropanol, tetrahydrofuran, ethylene glycol, ethylene glycol dimethyl ether, diethylene glycol dimethyl ether, dimethylformamide, acetone or methyl ethyl ketone. The deposition may be carried out more than once, for example two times or three times, to accomplish the deposition of a desired amount of the components. Alternatively, different components comprising a further element may be deposited in different deposition steps.

The total quantity of Group 11 metal present in the first section of the process microchannels is not material to the invention, and may be selected within wide ranges. Typically, the total quantity of Group 11 metal may be in the range of from 10 to 500 kg/m³, more typically from 50 to 400 kg/m³, in particular from 100 to 300 kg/m³ reactor volume, wherein reactor volume is the total volume defined by the cross sectional area and the total length of the portions of the process microchannels which is occupied by the epoxidation catalyst, by presence of a packed bed and/or by the presence of the epoxidation catalyst on the wall. For the avoidance of doubt, the reactor volume so defined does not include portions of the process microchannel which do not comprise epoxidation catalyst. In embodiments of the invention wherein the feed comprises the olefin and oxygen in a total quantity of at least 50 mole-%, the total quantity of Group 11 metal may be in the range of from 5 to 250 kg/m³, more typically from 20 to 200 kg/m³, in particular from 50 to 150 kg/m³ reactor volume, as defined hereinbefore.

The carrier materials which may be used in this invention may be natural or artificial inorganic materials and they may include refractory materials, silicon carbide, clays, zeolites, charcoal and alkaline earth metal carbonates, for example calcium carbonate. Preferred are refractory materials, such as alumina, magnesia, zirconia and silica. The most preferred material is α -alumina. Typically, the carrier

material comprises at least 85 %w, more typically at least 90 %w, in particular at least 95 %w α -alumina, frequently up to 99.9 %w α -alumina, relative to the weight of the carrier. Other components of the α -alumina may comprise, for example, silica, alkali metal components, for example sodium and/or potassium components, and/or alkaline earth metal components, for example calcium and/or magnesium components.

The surface area of the carrier material may suitably be at least 0.1 m²/g, preferably at least 0.3 m²/g, more preferably at least 0.5 m²/g, and in particular at least 0.6 m²/g, relative to the weight of the carrier; and the surface area may suitably be at most 10 m²/g, preferably at most 5 m²/g, and in particular at most 3 m²/g, relative to the weight of the carrier. "Surface area" as used herein is understood to relate to the surface area as determined by the B.E.T. (Brunauer, Emmett and Teller) method as described in Journal of the American Chemical Society 60 (1938) pp. 309-316. High surface area carrier materials, in particular when they are an α -alumina optionally comprising in addition silica, alkali metal and/or alkaline earth metal components, provide improved performance and stability of operation.

The water absorption of the carrier material is typically in the range of from 0.2 to 0.8 g/g, preferably in the range of from 0.3 to 0.7 g/g. A higher water absorption may be in favor in view of a more efficient deposition of Group 11 metal, promoter components and components comprising one or more elements. As used herein, water absorption is as measured in accordance with ASTM C20, and water absorption is expressed as the weight of the water that can be absorbed into the pores of the carrier, relative to the weight of the carrier.

The particulate carrier material may have a pore size distribution such that pores with diameters in the range of from 0.2 to 10 μ m represent at least 70 % of the total pore volume. Such relatively narrow pore size distribution can contribute to one or more of the activity, selectivity and longevity of the catalyst. Longevity may be in respect of maintaining the catalyst activity and/or maintaining the selectivity. As used herein, the pore size distribution and the pore volumes are as measured by mercury intrusion to a pressure of 3.0×10^8 Pa using a Micromeritics Autopore 9200 model (130° contact angle, mercury with a surface tension of 0.473 N/m, and correction for mercury compression applied).

Preferably, the pore size distribution is such that the pores with diameters in the range of from 0.2 to 10 μ m represent more than 75 %, in particular more than

80 %, more preferably more than 85 %, most preferably more than 90 % of the total pore volume. Frequently, the pore size distribution is such that the pores with diameters in the range of from 0.2 to 10 μm represent less than 99.9 %, more frequently less than 99 % of the total pore volume.

5 Preferably, the pore size distribution is such that the pores with diameters in the range of from 0.3 to 10 μm represent more than 75 %, in particular more than 80 %, more preferably more than 85 %, most preferably more than 90 %, in particular up to 100 %, of the pore volume contained in the pores with diameters in the range of from 0.2 to 10 μm .

10 Typically, the pore size distribution is such that pores with diameters less than 0.2 μm represent less than 10 %, in particular less than 5 %, of the total pore volume. Frequently, the pores with diameters less than 0.2 μm represent more than 0.1 %, more frequently more than 0.5 % of the total pore volume.

15 Typically, the pore size distribution is such that pores with diameters greater than 10 μm represent less than 20 %, in particular less than 10 %, more in particular less than 5 %, of the total pore volume. Frequently, the pores with diameters greater than 10 μm represent more than 0.1 %, in particular more than 0.5 % of the total pore volume.

20 The particulate carrier material has typically a d_{50} in the range of from 0.1 to 100 μm , in particular from 0.5 to 50 μm . As used herein, the average particle size, referred to herein as " d_{50} ", is as measured by a Horiba LA900 particle size analyzer and represents a particle diameter at which there are equal spherical equivalent volumes of particles larger and particles smaller than the stated average particle size. The method of measurement includes dispersing the particles by ultrasonic treatment, thus breaking up
25 secondary particles into primary particles. This sonification treatment is continued until no further change in the d_{50} value is noticed, which typically requires 5 minute sonification when using the Horiba LA900 particle size analyzer. Preferably, the particulate carrier material comprises particles having dimensions such that they pass an ASTM sieve with openings sized at most 50 %, in particular 30 % of the smallest
30 dimension of the process microchannel.

The epoxidation catalyst which comprises one or more Group 11 metals dispersed on a carrier material exhibits appreciable catalytic activity when the Group 11 metal content is at least 10 g/kg, relative to the weight of the catalyst. Preferably,

the catalyst comprises Group 11 metal in a quantity of from 50 to 500 g/kg, more preferably from 100 to 400 g/kg.

The promoter component may comprise one or more elements selected from rhenium, tungsten, molybdenum, chromium, and mixtures thereof. Preferably the promoter component comprises, as one of its elements, rhenium.

The promoter component may typically be present in the epoxidation catalyst in a quantity of at least 0.05 mmole/kg, more typically at least 0.5 mmole/kg, and preferably at least 1 mmole/kg, calculated as the total quantity of the element (that is rhenium, tungsten, molybdenum and/or chromium) relative to the weight of Group 11 metal. The promoter component may be present in a quantity of at most 250 mmole/kg, preferably at most 50 mmole/kg, more preferably at most 25 mmole/kg, calculated as the total quantity of the element relative to the weight of Group 11 metal. The form in which the promoter component may be deposited is not material to the invention. For example, the promoter component may suitably be provided as an oxide or as an oxyanion, for example, as a rhenate, perrhenate, or tungstate, in salt or acid form.

When the epoxidation catalyst comprises a rhenium containing promoter component, rhenium may typically be present in a quantity of at least 0.5 mmole/kg, more typically at least 2.5 mmole/kg, and preferably at least 5 mmole/kg, in particular at least 7.5 mmole/kg, calculated as the quantity of the element relative to the weight of Group 11 metal. Rhenium is typically present in a quantity of at most 25 mmole/kg, preferably at most 15 mmole/kg, more preferably at most 10 mmole/kg, in particular at most 7.5 mmole/kg, on the same basis.

Further, when the epoxidation catalyst comprises a rhenium containing promoter component, the catalyst may preferably comprise a rhenium copromoter, as a further component deposited on the carrier. Suitably, the rhenium copromoter may be selected from components comprising an element selected from tungsten, chromium, molybdenum, sulfur, phosphorus, boron, and mixtures thereof. Preferably, the rhenium copromoter is selected from components comprising tungsten, chromium, molybdenum, sulfur, and mixtures thereof. It is particularly preferred that the rhenium copromoter comprises, as an element, tungsten.

The rhenium copromoter may typically be present in a total quantity of at least 0.05 mmole/kg, more typically at least 0.5 mmole/kg, and preferably at least 2.5 mmole/kg, calculated as the element (i.e. the total of tungsten, chromium,

molybdenum, sulfur, phosphorus and/or boron), relative to the weight of Group 11 metal. The rhenium copromoter may be present in a total quantity of at most 200 mmole/kg, preferably at most 50 mmole/kg, more preferably at most 25 mmole/kg, on the same basis. The form in which the rhenium copromoter may be deposited is not material to the invention. For example, it may suitably be provided as an oxide or as an oxyanion, for example, as a sulfate, borate or molybdate, in salt or acid form.

The epoxidation catalyst preferably comprises Group 11 metal, the promoter component, and a component comprising a further element. Eligible further elements may be selected from the group of nitrogen, fluorine, alkali metals, alkaline earth metals, titanium, hafnium, zirconium, vanadium, thallium, thorium, tantalum, niobium, gallium and germanium and mixtures thereof. Preferably the alkali metals are selected from lithium, potassium, rubidium and cesium. Most preferably the alkali metal is lithium, potassium and/or cesium. Preferably the alkaline earth metals are selected from calcium and barium. Typically, the further element is present in the epoxidation catalyst in a total quantity of from 0.05 to 2500 mmole/kg, more typically from 0.25 to 500 mmole/kg, calculated as the element on the weight of Group 11 metal. The further elements may be provided in any form. For example, salts of an alkali metal or an alkaline earth metal are suitable.

As used herein, the quantity of alkali metal present in the epoxidation catalyst is deemed to be the quantity insofar as it can be extracted from the epoxidation catalyst with de-ionized water at 100 °C. The extraction method involves extracting a 10-gram sample of the catalyst three times by heating it in 20 ml portions of de-ionized water for 5 minutes at 100 °C and determining in the combined extracts the relevant metals by using a known method, for example atomic absorption spectroscopy.

As used herein, the quantity of alkaline earth metal present in the epoxidation catalyst is deemed to the quantity insofar as it can be extracted from the epoxidation catalyst with 10 %w nitric acid in de-ionized water at 100 °C. The extraction method involves extracting a 10-gram sample of the catalyst by boiling it with a 100 ml portion of 10 %w nitric acid for 30 minutes (1 atm., i.e. 101.3 kPa) and determining in the combined extracts the relevant metals by using a known method, for example atomic absorption spectroscopy. Reference is made to US-A-5801259, which is incorporated herein by reference.

In some embodiments, the invention provides processes for the epoxidation of an olefin comprising reacting a feed comprising the olefin and oxygen in the presence an epoxidation catalyst, as described hereinbefore, contained in one or more process microchannels of a microchannel reactor.

5 The olefin for use in the present invention may be an aromatic olefin, for example styrene, or a di-olefin, whether conjugated or not, for example 1,9-decadiene or 1,3-butadiene. A mixture of olefins may be used. Typically, the olefin is a monoolefin, for example 2-butene or isobutene. Preferably, the olefin is a mono- α -olefin, for example 1-butene or propylene. The most preferred olefin is ethylene.

10 The feed for the epoxidation process of this invention comprises the olefin and oxygen. As used herein, the feed to a process is understood to represent the total of reactants and other components which is fed to the section of the process microchannels in which the process in question takes place. Some of the feed components may be fed to the epoxidation process through an opening in upstream
15 end 220 of process microchannels 210. Some of the feed components may be fed through first feed channel 260 and one or more first orifices 280. For example, an olefin rich feed component may be fed through the opening in the upstream end of the process microchannels and an oxygen rich feed component may be fed through the first feed channel and the one or more first orifices. Alternatively, the oxygen rich
20 feed component may be fed through the opening in the upstream end of the process microchannels and the olefin rich feed component may be fed through the first feed channel and the one or more first orifices. Certain feed components may be fed through the opening in the upstream end of the process microchannels and through the first feed channel and the one or more first orifices. For example, the olefin may be
25 fed partly through the opening in the upstream end of the process microchannels and partly through the first feed channel and the one or more first orifices. As another example, oxygen may be fed partly through the opening in the upstream end of the process microchannels and partly through the first feed channel and the one or more first orifices.

30 An organic halide may be present in the feed as a reaction modifier for increasing the selectivity, suppressing the undesirable oxidation of the olefin or the olefin oxide to carbon dioxide and water, relative to the desired formation of the olefin oxide. The organic halide may be fed as a liquid or as a vapor. The organic

halide may be fed separately or together with other feed components through an opening in upstream end 220 of the process microchannels 210 or through first feed channel 260 and one or more first orifices 280. An aspect of feeding the organic halide through a plurality first orifices is that there may be an increase in the level of the quantity of the organic halide along the length of the epoxidation catalyst, by which the activity and/or selectivity of the epoxidation catalyst can be manipulated in accordance with the teachings of EP-A-352850, which is incorporated herein by reference. For example, when using a rhenium containing epoxidation catalyst, the activity of the epoxidation catalyst can be enhanced along the length of the epoxidation catalyst. This could allow for better utilization of the epoxidation catalyst in regions where oxygen or the olefin is depleted relative to the regions where oxygen and the olefin are fed.

Organic halides are in particular organic bromides, and more in particular organic chlorides. Preferred organic halides are chlorohydrocarbons or bromohydrocarbons. More preferably they are selected from the group of methyl chloride, ethyl chloride, ethylene dichloride, ethylene dibromide, vinyl chloride or a mixture thereof. Most preferred are ethyl chloride and ethylene dichloride.

In addition to an organic halide, an organic or inorganic nitrogen compound may be employed as reaction modifier, but this is generally less preferred. It is considered that under the operating conditions of the epoxidation process the nitrogen containing reaction modifiers are precursors of nitrates or nitrites (cf. e.g. EP-A-3642 and US-A-4822900, which are incorporated herein by reference). Organic nitrogen compounds and inorganic nitrogen compounds may be employed. Suitable organic nitrogen compounds are nitro compounds, nitroso compounds, amines, nitrates and nitrites, for example nitromethane, 1-nitropropane or 2-nitropropane. Suitable inorganic nitrogen compounds are, for example, nitrogen oxides, hydrazine, hydroxylamine or ammonia. Suitable nitrogen oxides are of the general formula NO_x wherein x is in the range of from 1 to 2, and include for example NO, N_2O_3 and N_2O_4 .

The organic halides and the organic or inorganic nitrogen compounds are generally effective as reaction modifier when used in low total concentration, for example up to 0.01 mole-%, relative to the total feed. It is preferred that the organic halide is present at a concentration of at most 50×10^{-4} mole-%, in particular at most 20×10^{-4} mole-%, more in particular at most 15×10^{-4} mole-%, relative to the total feed,

and preferably at least 0.2×10^{-4} mole-%, in particular at least 0.5×10^{-4} mole-%, more in particular at least 1×10^{-4} mole-%, relative to the total feed.

In addition to the olefin, oxygen and the organic halide, the feed may additionally comprise one or more further components, for example saturated hydrocarbons, as ballast gas, inert gases and carbon dioxide. The one or more further components may be fed separately or together with other feed components through an opening in upstream end 220 of the process microchannels 210 or through first feed channel 260 and one or more first orifices 280.

The olefin concentration in the feed may be selected within a wide range. Typically, the olefin concentration in the feed will be at most 80 mole-%, relative to the total feed. Preferably, it will be in the range of from 0.5 to 70 mole-%, in particular from 1 to 60 mole-%, on the same basis.

The oxygen concentration in the feed may be selected within a wide range. Typically, the concentration of oxygen applied will be within the range of from 1 to 15 mole-%, more typically from 2 to 12 mole-% of the total feed.

The saturated hydrocarbons comprise, for example, methane and ethane. Unless stated herein otherwise, saturated hydrocarbons may be present in a quantity of up to 80 mole-%, in particular up to 75 mole-%, relative to the total feed, and frequently they are present in a quantity of at least 30 mole-%, more frequently at least 40 mole-%, on the same basis.

Carbon dioxide may be present in the feed as it is formed as a result of undesirable oxidation of the olefin and/or the olefin oxide, and it may accordingly be present in feed components present in a recycle stream. Carbon dioxide generally has an adverse effect on the catalyst activity. Advantageously, the quantity of carbon dioxide is, for example, below 2 mole-%, preferably below 1 mole-%, or in the range of from 0.2 to 1 mole-%, relative to the total feed.

The inert gases include, for example nitrogen or argon. Unless stated herein otherwise, the inert gases may be present in the feed in a concentration of from 30 to 90 mole-%, typically from 40 to 80 mole-%.

The epoxidation process of this invention may be air-based or oxygen-based, see "Kirk-Othmer Encyclopedia of Chemical Technology", 3rd edition, Volume 9, 1980, pp. 445-447. In the air-based process air or air enriched with oxygen is employed as the source of the oxidizing agent while in the oxygen-based processes

high-purity (at least 95 mole-%) oxygen is employed as the source of the oxidizing agent. Presently most epoxidation plants are oxygen-based and this is preferred in the practice of certain embodiment of this invention. It is an advantage of other embodiments of this invention that air may be fed to the process as the source of the oxidizing agent.

The epoxidation process may be carried out using reaction temperatures selected from a wide range. Preferably the reaction temperature is in the range of from 150 to 340 °C, more preferably in the range of from 180 to 325 °C. Typically, the heat transfer liquid present in the first heat exchange channels may have a temperature which is typically 0.5 to 10 °C lower than the reaction temperature.

The epoxidation process is preferably carried out at a pressure, as measured at upstream 220 end of the process microchannels 210, in the range of from 1000 to 3500 kPa.

The olefin oxide leaving the section of the process microchannels containing the epoxidation catalyst is comprised in a reaction mixture which may further comprise unreacted olefin, unreacted oxygen, and other reaction products such as carbon dioxide. Typically, the content of olefin oxide in the reaction product is in general in the range of from 1 to 25 mole-%, more typically from 2 to 20 mole-%, in particular from 2 to 5 mole-%.

The epoxidation process may comprise reacting a feed comprising the olefin and oxygen in a total quantity of at least 50 mole-%, relative to the total feed. In this embodiment, the olefin and oxygen may be present in the feed in a total quantity of at least 80 mole-%, in particular at least 90 mole-%, more in particular at least 95 mole-%, relative to the total feed, and typically up to 99.5 mole-%, in particular up to 99 mole-%, relative to the total feed. The molar ratio of olefin to oxygen may be in the range of from 3 to 100, in particular from 4 to 50, more in particular from 5 to 20. The saturated hydrocarbons and the inert gases may be substantially absent. As used herein, in this context “substantially absent” means that the quantity of saturated hydrocarbons in the feed is at most 10 mole-%, in particular at most 5 mole-%, more in particular at most 2 mole-%, relative to the total feed, and that the quantity of inert gases in the feed is at most 10 mole-%, in particular at most 5 mole-%, more in particular at most 2 mole-%, relative to the total feed. In this particular embodiment, process conditions may be applied such that the quantity of olefin oxide in the epoxidation reaction mixture is in the range of from 4 to 15 mole-%, in particular from 5 to 12 mole-%, for example from 6 to

10 mole-%. Preferably, the epoxidation reaction mixture, including the olefin oxide, is quenched, as described herein.

The epoxidation process may comprise applying conditions such that the conversion of the olefin or the conversion of oxygen is at least 90 mole-%. The conversion of the olefin may be at least 90 mole-% and the conversion of oxygen may be at least 90 mole-%. In particular, in this embodiment, the feed may comprise the olefin and oxygen in a quantity of at most 50 mole-%, relative to the total feed, and the feed may additionally comprise saturated hydrocarbons, as ballast gas, and inert gas. Typically, process conditions are applied such that the conversion of the olefin or the conversion of oxygen is at least 95 mole-%, in particular at least 98 mole-%, more in particular at least 99 mole-%. As used herein, the conversion is the quantity of a reactant converted relative to the quantity of the reactant in the feed, expressed in mole-%. Preferably, the conversion of the olefin is at least 95 mole-%, in particular at least 98 mole-%, more in particular at least 99 mole-% and oxygen may be at least partly replenished. The presence of an excess of oxygen in the feed, relative to the olefin, assists in achieving a high conversion of the olefin. For example, the molar ratio of oxygen over the olefin in the feed may be at least 1.01, typically at least 1.05, in particular at least 1.1, more in particular at least 1.2; and for example at most 5, in particular at most 3, more in particular at most 2. In this embodiment, a relatively high selectivity in the conversion of the olefin into the olefin oxide is achieved. As used herein, the selectivity is the quantity of olefin oxide formed, relative to the quantity of olefin converted, expressed in mole-%. Moreover, such high conversion of the olefin enables that the process may be carried out economically in a once-through mode, which means that no recycle of unconverted reactants is applied, and that air may be fed to the epoxidation process, which means effectively that the need of an air separation unit is eliminated.

In the practice of this invention, the reaction product, including the olefin oxide, may be quenched by heat exchange with a heat exchange fluid. The quenching may be conducted in second section 340 of process microchannels 210 by heat exchange with heat exchange fluid present in one or more second heat exchange channels 350. Typically, the temperature of the reaction product, including the olefin oxide, may be decreased to a temperature of at most 250 °C, more typically at most 225 °C, preferably in the range of from 20 to 200 °C, more preferably 50 to 190 °C, in particular from 80 to 180 °C. The quenching may result in a reduction in temperature in the range of from 50 to 200 °C, in particular from 70 to 160 °C. Quenching enables

increasing the total quantity of the olefin oxide and oxygen in the feed of the epoxidation process, and eliminating the ballast gas or reducing the quantity of ballast gas in the feed of the epoxidation process. Also, a result of quenching is that the olefin oxide produced is a cleaner product, comprising less aldehyde and carboxylic acid impurities.

In some embodiments, the epoxidation process may comprise

- reacting a feed comprising an olefin and oxygen in the presence of an epoxidation catalyst contained in a first section 240 of one or more process microchannels 210 of a microchannel reactor to thereby form a first mixture comprising the olefin oxide and carbon dioxide, as described hereinbefore,
- quenching the first mixture in intermediate section 440 of the one or more process microchannels 210 positioned downstream of first section 240 by heat exchange with a heat exchange fluid, in a same manner as described hereinbefore, and
- converting in second section 340 of the one or more process microchannels 210 positioned downstream of intermediate section 440 the quenched first mixture to form a second mixture comprising the olefin oxide and a 1,2-carbonate.

The conversion of the quenched first mixture comprising the olefin oxide and carbon dioxide to form the second mixture comprising the olefin oxide and a 1,2-carbonate typically involves reacting at least a portion of the olefin oxide present in the first mixture with at least a portion of the carbon dioxide present in the first mixture to form the 1,2-carbonate. Typically, carbon dioxide present in the first mixture is carbon dioxide co-formed in the epoxidation reaction. The molar quantity of carbon dioxide present in the first mixture may be in the range of from 0.01 to 1 mole, in particular 0.02 to 0.8 mole, more in particular 0.05 to 0.6 mole-%, per mole of the olefin oxide present in the first mixture. Suitable catalysts for the conversion of the olefin oxide with carbon dioxide may be, for example, resins which comprising quaternary phosphonium halide groups or quaternary ammonium halide groups on a styrene/divinylbenzene copolymer matrix, wherein the halide may be in particular chloride or bromide. Such catalysts for this conversion are known from T. Nishikubo, A. Kameyama, J. Yamashita and M. Tomoi, Journal of Polymer Science, Pt. A. Polymer Chemist, 31, 939 - 947 (1993), which is incorporated herein by reference. Other suitable catalysts for the conversion of the olefin oxide with carbon dioxide are, for example, quaternary phosphonium halides, quaternary ammonium halides, and certain metal halides. An example is methyltributylphosphonium iodide. The temperature may be in the range of from 30 to

200 °C, in particular from 50 to 150 °C. The pressure may be in the range of from 500 to 3500 kPa, as measured at the second feed channel, described hereinbefore. Typically, at least 50 mole-%, in particular at least 80 mole-%, more in particular at least 90 mole-% of the carbon dioxide is converted, for example at least 98 mole-%, and in the practice of this invention, frequently at most 99.9 mole-% is converted.

In this embodiment, in cases that the second mixture is formed as a gaseous phase, the process may additionally comprise condensing at least a portion of the second mixture comprising the olefin oxide and the 1,2-carbonate in a third section of the one or more process microchannels, which third section is positioned downstream of the second section. Typically, condensing at least a portion of the second mixture involves removal of heat by heat exchange with a heat exchange fluid. Such heat exchange fluid may be present in a fourth heat exchange channel, as described hereinbefore. Typically, at least 50 mole-%, in particular at least 80 mole-%, more in particular at least 90 mole-% of the total of the olefin oxide and the 1,2-carbonate present in the second mixture is condensed, for example at least 98 mole-%, and in the practice of this invention, frequently at most 99.9 mole-% is condensed. Preferably, in cases that the second mixture comprises water at least partly as a gaseous phase, the process may additionally comprise condensing at least a portion of such water present in the second mixture in the third section. Typically, water present in the second mixture, if any, is water co-formed in the epoxidation reaction. The molar quantity of water present in the second mixture may be in the range of from 0.01 to 1 mole, in particular 0.02 to 0.8 mole, more in particular 0.05 to 0.6 mole-%, per mole of the total quantities of the olefin oxide and the 1,2-carbonate present in the second mixture. Typically, at least 50 mole-%, in particular at least 80 mole-%, more in particular at least 90 mole-% of the total of the water present in the second mixture is condensed, for example at least 98 mole-%, and in the practice of this invention, frequently at most 99.9 mole-% is condensed.

The epoxidation reaction mixture, including the olefin oxide, may be withdrawn from the process microchannel and the microchannel reactor and be processed in the conventional manner, using conventional methods and conventional equipment. A separation system may provide for the separation of the olefin oxide from any unconverted olefin, any unconverted oxygen, any ballast gas and carbon dioxide. An aqueous extraction fluid such as water may be used to separate these components. The enriched extraction fluid containing the olefin oxide may be further processed for recovery of the olefin oxide. The olefin oxide produced may be

recovered from the enriched extraction fluid, for example by distillation or extraction. A mixture which comprises any unconverted olefin, any unconverted oxygen, any ballast gas and carbon dioxide and which is lean in olefin oxide may be extracted to at least partly remove carbon dioxide. The resulting carbon dioxide lean mixture may be
5 recompressed, dried and recycled as a feed component to the epoxidation process of this invention.

The olefin oxide produced in the epoxidation process of the invention may be converted by conventional methods into a 1,2-diol, a 1,2-diol ether, a 1,2-carbonate or an alkanol amine.

10 The conversion into the 1,2-diol or the 1,2-diol ether may comprise, for example, reacting the ethylene oxide with water, in a thermal process or by using a catalyst, which may be an acidic catalyst or a basic catalyst. For example, for making predominantly the 1,2-diol and less 1,2-diol ether, the olefin oxide may be reacted with a ten fold molar excess of water, in a liquid phase reaction in presence of an acid
15 catalyst, e.g. 0.5-1.0 %w sulfuric acid, based on the total reaction mixture, at 50-70 °C at 100 kPa absolute, or in a gas phase reaction at 130-240 °C and 2000-4000 kPa absolute, preferably in the absence of a catalyst. The presence of such a large quantity of water may favor the selective formation of 1,2-diol and may function as a sink for the reaction exotherm, helping controlling the reaction temperature. If the
20 proportion of water is lowered the proportion of 1,2-diol ethers in the reaction mixture is increased. The 1,2-diol ethers thus produced may be a di-ether, tri-ether, tetra-ether or a subsequent ether. Alternative 1,2-diol ethers may be prepared by converting the olefin oxide with an alcohol, in particular a primary alcohol, such as methanol or ethanol, by replacing at least a portion of the water by the alcohol.

25 The olefin oxide may be converted into the corresponding 1,2-carbonate by reacting it with carbon dioxide. If desired, a 1,2-diol may be prepared by subsequently reacting the 1,2-carbonate with water or an alcohol to form the 1,2-diol. For applicable methods, reference is made to US-A-6080897, which is incorporated herein by reference.

30 The conversion into the alkanol amine may comprise reacting the olefin oxide with an amine, such as ammonia, an alkyl amine or a dialkyl amine. Anhydrous or aqueous ammonia may be used. Anhydrous ammonia is typically used to favor the production of mono alkanol amine. For methods applicable in the conversion of the

olefin oxide into the alkanol amine, reference may be made to, for example US-A-4845296, which is incorporated herein by reference.

The 1,2-diols and 1,2 diol ethers, for example ethylene glycol, 1,2-propylene glycol and ethylene glycol ethers may be used in a large variety of industrial applications, for example in the fields of food, beverages, tobacco, cosmetics, thermoplastic polymers, curable resin systems, detergents, heat transfer systems, etc. The 1,2-carbonates, for example ethylene carbonate, may be used as a diluent, in particular as a solvent. Ethanol amines may be used, for example, in the treating (“sweetening”) of natural gas.

Unless specified otherwise, the organic compounds mentioned herein, for example the olefins, alcohols, 1,2-diols, 1,2-diol ethers, 1,2-carbonates, ethanol amines and organic halides, have typically at most 40 carbon atoms, more typically at most 20 carbon atoms, in particular at most 10 carbon atoms, more in particular at most 6 carbon atoms. Typically, the organic compounds have at least one carbon atom. As defined herein, ranges for numbers of carbon atoms (i.e. carbon number) include the numbers specified for the limits of the ranges.

The following example is intended to illustrate the advantages of the present invention and is not intended to unduly limit the scope of the invention.

Example

This prophetic example describes how an embodiment of this invention may be practiced.

A microchannel reactor will comprise process microchannels, first heat exchange microchannels, second heat exchange microchannels and first feed channels. The process microchannels will comprise an upstream end, a first section and a second section.

The first section will be adapted to exchange heat with a heat exchange fluid flowing in the first heat exchange microchannels. The second heat exchange microchannels will comprise two sets of second heat exchange microchannels adapted to exchange heat with the second section, such that in the downstream portion of the second section a lower temperature will be achieved than in the upstream portion of the second section. A feed microchannel will end in the first section of the process microchannel through orifices. The orifices will be positioned at approximately equal distances into the downstream direction of the first section from the upstream end of the microchannel till two thirds of the length of the first section, and in the

perpendicular direction the orifices will be positioned at approximately equal distances approximately across the entire width of the process microchannel.

The first section will comprise an epoxidation catalyst comprising silver, rhenium, tungsten, cesium and lithium deposited on a particulate carrier material. The particulate carrier material will be an α -alumina having a surface area of $1.5 \text{ m}^2/\text{g}$, a total pore volume of 0.4 ml/g , and a pore size distribution such that that pores with diameters in the range of from 0.2 to $10 \text{ }\mu\text{m}$ represent 95 % of the total pore volume, and that pores with diameters in the range of from 0.3 to $10 \text{ }\mu\text{m}$ represent more than 92 %, of the pore volume contained in the pores with diameters in the range of from 0.2 to $10 \text{ }\mu\text{m}$.

The microchannel reactor will be assembled in accordance with methods known from WO-A-2004/099113, and references cited therein. The carrier material will be deposited on the walls of the first section of the process microchannels by wash coating. Thereafter, the process microchannels will be assembled, and after assembly silver, rhenium, tungsten, cesium and lithium will be deposited on the carrier material by using methods, which are known per se from US-A-5380697. The quantity of silver is 350 kg/m^3 reactor volume, reactor volume being the total volume defined by the cross sectional area and the total length of the portions of the microchannels occupied by the epoxidation catalyst.

The first section will be heated at $220 \text{ }^\circ\text{C}$ by heat exchange with the heat exchange fluid flowing in the first heat exchange microchannel, while ethylene is fed through an opening positioned at the upstream end of the process microchannels. A mixture of oxygen and ethyl chloride (3 parts by million by volume) will be fed through the feed channels. The molar ratio of oxygen to ethylene will be 1:1. The mixture exiting the first section and entering the second section of the process microchannels will be quenched in the second section in two steps, initially to a temperature of $150 \text{ }^\circ\text{C}$ and subsequently to a temperature of $80 \text{ }^\circ\text{C}$. The temperature and the feed rate of the ethylene and oxygen will be adjusted such that the conversion of ethylene is 97 mole-%. Then, the quantity of ethyl chloride in the mixture of oxygen and ethyl chloride will be adjusted so as to optimize the selectivity to ethylene oxide.

The ethylene oxide rich product may be purified by removing carbon dioxide and unconverted oxygen and ethylene. The purified ethylene oxide may be converted with water to yield ethylene glycol.

C L A I M S

1. A method of installing an epoxidation catalyst in one or more process microchannels of a microchannel reactor, which method comprises depositing a Group 11 metal or a cationic Group 11 metal component on at least a portion of the walls of the said process microchannels, depositing one or more promoter components on at least a portion of the same walls prior to, together with or subsequent to the deposition of the Group 11 metal or the cationic Group 11 metal component, and, if a cationic Group 11 metal component is deposited, reducing at least a portion of the cationic Group 11 metal component.

2. A method as claimed in claim 1, wherein the Group 11 metal is deposited as a cationic Group 11 metal component.

3. A method as claimed in claim 2, wherein the Group 11 metal is deposited by contacting the walls with a liquid mixture comprising the cationic Group 11 metal component, removing a liquid component of the liquid mixture, and applying a reducing agent prior to, together with or after the deposition of cationic Group 11 metal component.

4. A method as claimed in any of claims 1-3, wherein the method additionally comprises covering the walls of the process microchannels at least partly with a carrier material and then depositing the Group 11 metal or cationic Group 11 metal component on or in the carrier material.

5. A method as claimed in claim 4, wherein the carrier material is a particulate material having a d_{50} in the range of from 0.1 to 100 μm .

6. A method as claimed in claim 5, wherein the particulate material has a d_{50} in the range of from 0.5 to 50 μm .

7. A method as claimed in claim 4, wherein the carrier material is a particulate material capable of passing an ASTM sieve with openings sized at at most 50 % of the smallest dimension of the process microchannel.

8. A method as claimed in claim 7, wherein the carrier material is a particulate material capable of passing an ASTM sieve with openings sized at at most 30 % of the smallest dimension of the process microchannel.

9. A method as claimed in claim 1, wherein the Group 11 metal is deposited by contacting the walls with a liquid containing dispersed Group 11 metal, and removing the liquid, while leaving Group 11 metal on the wall.

10. A method as claimed in claim 1, wherein the Group 11 metal is deposited by a vapour deposition technique.

11. A method as claimed in any of claims 1-10, wherein the Group 11 metal or the cationic Group 11 metal component is deposited on at least a portion of the walls by depositing Group 11 metal or cationic Group 11 metal component on at least a portion of one or more sheets, and manufacturing a microchannel reactor by assembling the sheets so as to form process microchannels having Group 11 metal or cationic Group 11 metal component deposited on at least a portion of the walls.

12. A method as claimed in any of claims 1-11, wherein the quantity of Group 11 metal deposited is in the range of from 10 to 500 kg/m³ reactor volume, reactor volume being the total volume defined by the cross sectional area and the total length of the portions of the microchannels which is occupied by the epoxidation catalyst.

13. A method as claimed in claims 12, wherein the quantity of Group 11 metal deposited is in the range of from 50 to 400 kg/m³ reactor volume, reactor volume being the total volume defined by the cross sectional area and the total length of the portions of the microchannels which is occupied by the epoxidation catalyst.

14. A method as claimed in any of claims 4-13, wherein the method comprises depositing the Group 11 metal or cationic Group 11 metal component or the carrier material on at least partly roughened or corrugated portions of the walls of the process microchannels of which portions the roughened or corrugated wall surface is effectively enlarged by a factor of from 0.5 to 10, relative to the surface area of the roughened or corrugated wall surface as defined by its outer dimensions.

15. A method as claimed in any of claims 1-14, wherein the catalyst comprises silver as the Group 11 metal.

16. A method as claimed in claim 15, wherein the catalyst additionally comprises a promoter component comprising one or more elements selected from rhenium,

tungsten, molybdenum, chromium, and mixtures thereof, and additionally comprises an alkali metal selected from lithium, potassium, cesium, and mixtures thereof.

17. A process for the epoxidation of an olefin comprising

- installing an epoxidation catalyst in one or more process microchannels of a microchannel reactor by a method as claimed in any of claims 1-16, and
- reacting a feed comprising the olefin and oxygen in the presence of the epoxidation catalyst installed in the one or more process microchannels.

18. A process as claimed in claim 17, wherein the feed comprises the olefin and oxygen in a total quantity of at least 50 mole-%, relative to the total feed.

19. A process as claimed in claim 17 or 18, wherein the process comprises reacting a feed comprising the olefin and oxygen and applying conditions such that the conversion of the olefin or the conversion of oxygen is at least 90 mole-%.

20. A process as claimed in any of claims 17-19, wherein the process additionally comprises quenching the reaction product in a downstream section of the process microchannels.

21. A process as claimed in claim 20, wherein the process additionally comprises converting in the one or more process microchannels the quenched reaction product to form a mixture comprising the olefin oxide and a 1,2-carbonate.

22. A process for the preparation of a 1,2-diol, a 1,2-diol ether, a 1,2-carbonate or an alkanol amine, which process comprises

- installing an epoxidation catalyst in one or more process microchannels of a microchannel reactor by a method as claimed in any of claims 1-16,
- reacting a feed comprising the olefin and oxygen in the presence of the epoxidation catalyst installed in the one or more process microchannels to produce an olefin oxide, and
- converting the olefin oxide with water, an alcohol, carbon dioxide or an amine to form the 1,2-diol, 1,2-diol ether, 1,2-carbonate or alkanol amine.

23. A process as claimed in any of claim 17-22, wherein the olefin comprises ethylene.

24. A reactor suitable for the epoxidation of an olefin, which reactor is a microchannel reactor comprising one or more process microchannels having installed therein an epoxidation catalyst which comprises a Group 11 metal, wherein the quantity of the Group 11 metal is in the range of from 10 to 500 kg/m³ reactor volume, reactor

volume being the total volume defined by the cross sectional area and the total length of the portions of the microchannels occupied by the epoxidation catalyst.

25. A reactor as claimed in claim 24, wherein the quantity of Group 11 metal deposited is in the range of from 50 to 400 kg/m³ reactor volume, reactor volume being the total volume defined by the cross sectional area and the total length of the portions of the microchannels which is occupied by the epoxidation catalyst.

26. A reactor as claimed in claim 24 or 25, wherein the catalyst comprises silver as the Group 11 metal.

27. A reactor as claimed in claim 26, wherein the catalyst additionally comprises a promoter component comprising one or more elements selected from rhenium, tungsten, molybdenum, chromium, and mixtures thereof, and additionally comprises an alkali metal selected from lithium, potassium, cesium, and mixtures thereof.

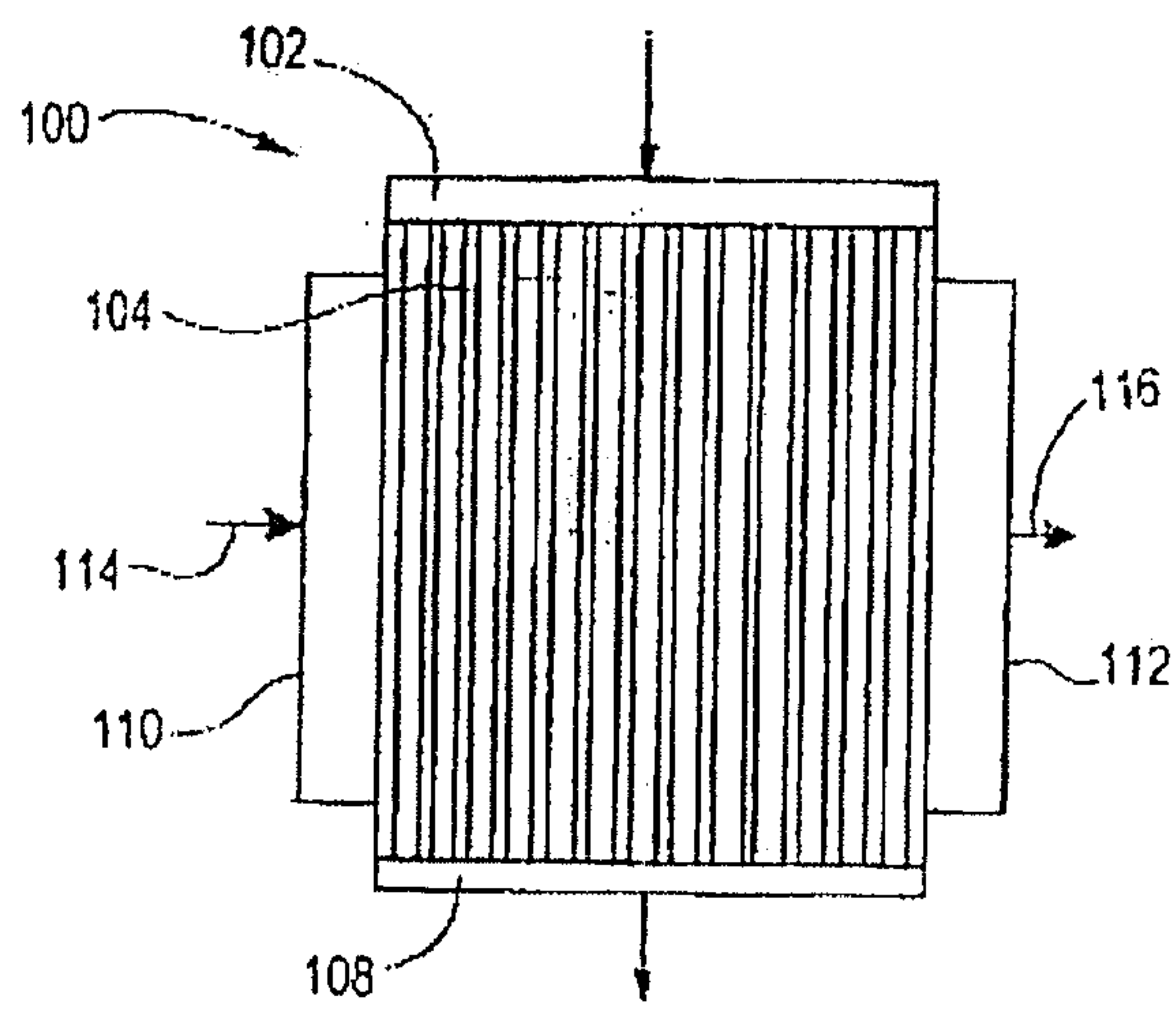


FIG. 1

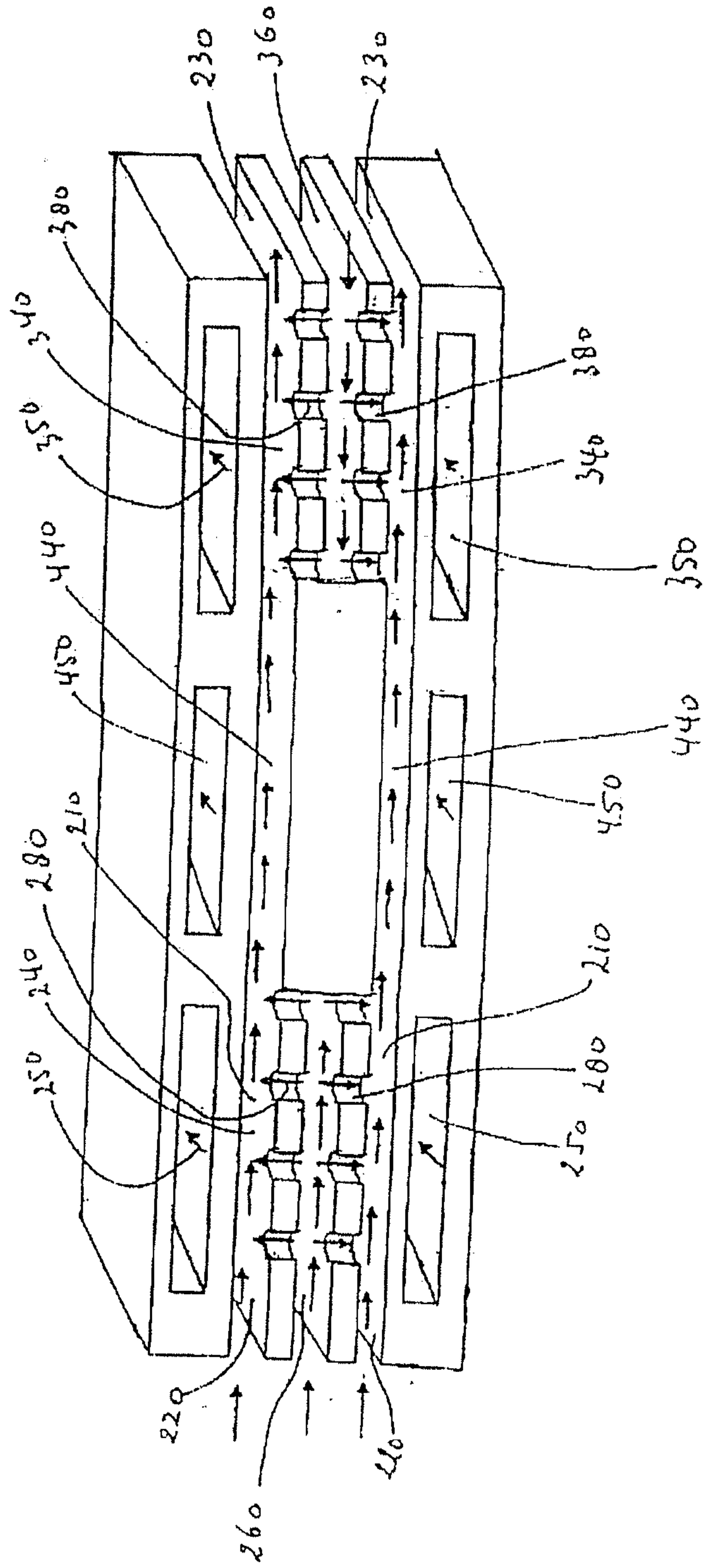


FIG. 2