Claims

1. Process for preparing a product mixture (2) by industrial hydroformylation of an isobutene-containing hydrocarbon stream (1) and for separating the resulting product mixture (2), comprising the following steps:

a) hydroformylating the isobutene-containing hydrocarbon stream (1) in a hydroformylation reactor (R) in the presence of a transition metal complex catalyst, so as to obtain a product mixture (2) comprising at least 3-methylbutanal, conversion products in the form of high boilers and 3-methylbutanoic acid, and the transition metal complex catalyst along with the free ligands thereof,

b) separating the product mixture (2) by means of a nanofiltration device (M) comprising one or more membrane separation stages, such that the transition metal complex catalyst and the free ligands thereof are enriched in the resulting retentate (3) of the nanofiltration device (M) with respect to 3-methylbutanal and 3-methylbutanoic acid, and such that 3-methylbutanal and 3-methylbutanoic acid are each enriched in the resulting permeate (4) of the nanofiltration device (M) with respect to the transition metal complex catalyst, the concentration of the 3-methylbutanoic acid being lower in the resulting retentate (3) than in the permeate (4),

c) separating the resulting permeate (4) of the nanofiltration device (M) by means of a thermal separating device (D) comprising one or more separation stages into at least one first fraction (5) and a second fraction (6), the first fraction (5) having a higher concentration of 3-methylbutanal than the second fraction (6) and a lower concentration of conversion products in the form of high boilers and 3-methylbutanoic acid than the second fraction (6),

d) recycling at least a substream of the resulting retentate (3) of the nanofiltration device (M) into the hydroformylation reactor (R).

2. Process according to Claim 1, characterized in that one or more process parameters in the step of hydroformylating the isobutenic hydrocarbon stream are set such that the total concentration of conversion products in the form of high boilers and 3-methylbutanoic acid, based on the weight of the overall product mixture, is 30% by weight or less,
the parameter(s) to be set preferably being selected from the group consisting of pressure, temperature, mean residence time, composition of the synthesis gas, concentration of the transition metal and transition metal-ligand ratio of the transition metal complex catalyst.

3. Process according to Claim 1 or 2, characterized in that the step of hydroformylating the isobutenic hydrocarbon stream is performed
- at a pressure in the range from 0.2 to 8 MPa,
and/or
- at a temperature in the range from 70 to 130°C
and/or
- with a mean residence time in the range from 1 to 4 h
and/or
- a synthesis gas composition $CO:H_2$ of 1:3 to 3:1
and/or
- a transition metal concentration in the range from 10 to 100 ppm
and/or
- a transition metal/ligand ratio in the range from 1:4 to 1:50.

4. Process according to any of the preceding claims, characterized in that
- the transition metal of the transition metal complex catalyst is rhodium
and/or
- the ligand(s) of the transition metal complex catalyst is/are selected from the group of the organophosphorus ligands.

5. Process according to any of the preceding claims, characterized in that a transition metal complex catalyst selected from the group consisting of rhodium and cobalt is used.

6. Process according to any of the preceding claims, characterized in that one or more nanofiltration membranes are used in the nanofiltration device, the or at least one of the nanofiltration membranes having a retention for 3-methylbutanoic acid of -1 or less, more preferably -5 or less and especially -10 or less.

7. Process according to any of the preceding claims, characterized in that one or more nanofiltration membranes which are used in the nanofiltration device comprise or consist of one or more polymers containing imide groups, the polymer(s) containing imide groups being selected such that the retention of the nanofiltration membrane for 3-methylbutanoic acid is -1 or less, more preferably -5 or less, and especially -10 or less.

8. Process according to any of the preceding claims, characterized in that one or more nanofiltration membranes are used in the nanofiltration device, the or at least one of the nanofiltration membranes having a separation limit in the range from 150 to 2000 g/mol, preferably 200 to 600 g/mol and most preferably 350 to 500 g/mol.

9. Process according to any of the preceding claims, characterized in that one or more nanofiltration membranes which are used in the nanofiltration device comprise or consist of one or more polymers containing imide groups, the polymer(s) containing imide groups being selected such that the separation limit of the nanofiltration membrane is in the range from 150 to 2000 g/mol.

10. Process according to any of the preceding claims, characterized in that the step of separating the product mixture obtained by hydroformylating the isobutenic hydrocarbon stream in the nanofiltration device is performed

- at a temperature in the range from 10 to 150°C

and/or

- at a transmembrane pressure in the range from 0.5 to 6 MPa

and/or

- at a Reynolds number between 55 and 13 500, preferably between 100 and 3500 and most preferably between 170 and 900,

and/or

- in the presence of carbon monoxide and/or hydrogen, preferably at a partial carbon monoxide pressure of at least 200 kPa in the feed, in the retentate and in the permeate of each membrane separation stage.

11. Process according to any of the preceding claims, characterized in that the step of thermal separation comprises a distillation in which the first fraction is obtained as the top product and the second fraction as the bottom product.

12. Process according to any of the preceding claims, comprising the steps of:

- hydroformylating an isobutenic hydrocarbon stream in the presence of a rhodium complex catalyst having one or more organophosphorus ligands at a pressure in the range from 0.2 to 8 MPa and a temperature in the range from 70 to 130°C with a mean residence time in the range from 1 to 4 h, a synthesis gas composition $CO:H_2$ of 1:3 to 3:1, a rhodium concentration in the reactor in the range from 10 to 100 ppm and a rhodium/ligand ratio in the range from 1:4 to 1:50;
- separating the resulting product mixture of the hydroformylation by means of a nanofiltration device at a temperature in the range from 10 to 150°C, a transmembrane pressure in the range from 0.5 to 6 MPa, at a Reynolds number between 170 and 900, and a partial carbon monoxide pressure of greater than 0.2 MPa into feed, retentate and permeate of each membrane separation stage;
- thermally separating the resulting permeate of the nanofiltration device by means of distillation into a first fraction and a second fraction, the first fraction containing a higher concentration of 3-methylbutanal than the second fraction and a lower concentration of high boilers than the second fraction,
- recycling at least a substream of the resulting retentate of the nanofiltration device into the hydroformylation reactor.

13. Process according to any of the preceding claims, comprising the step of

- monitoring the concentration of 3-methylbutanoic acid in the resulting retentate of the nanofiltration device, and preferably also in the product mixture and/or in the resulting permeate of the nanofiltration device.

14. Process according to Claim 13, characterized in that, on exceedance of a fixed maximum concentration of the 3-methylbutanoic acid in the resulting retentate of the nanofiltration device, the recycling of the resulting retentate into the hydroformylation reactor is stopped.

15. Process according to Claim 13 or 14, characterized in that, on exceedance of a fixed maximum concentration of 3-methylbutanoic acid in the feed of the nanofiltration device, one or more process parameters in the step of hydroformylating the isobutenic hydrocarbon stream in the hydroformylation reactor are modified such that the concentration of 3-methylbutanoic acid in the feed of the nanofiltration device is lowered below the fixed maximum.

16. Apparatus (1) for performance of the process according to any of Claims 1 to 15, comprising

- a hydroformylation reactor (R) for hydroformylation of an isobutenic hydrocarbon stream (1) in the presence of a transition metal complex catalyst,
- a nanofiltration device (M) comprising one or more membrane separation stages for separation of the product mixture (2) formed in the hydroformylation reactor (R), such that the transition metal complex catalyst and the free ligands thereof are enriched in the resulting retentate (3) of the nanofiltration device (M) with respect to 3-methylbutanal and 3-methylbutanoic acid, and 3-methylbutanal and 3-methylbutanoic acid are enriched in the resulting permeate of the nanofiltration device (M) with respect to the transition metal complex catalyst, such that the concentration of 3-methylbutanoic acid in the resulting retentate (3) is lower than in the resulting permeate (4), the nanofiltration device (M) comprising one or more nanofiltration membranes, the or at least one of the nanofiltration membranes having a retention for 3-methylbutanoic acid of -1 or less, more preferably -5 or less, and especially -10 or less,
- at least one thermal separation device (D) for thermal separation of the resulting permeate (4) of the nanofiltration device (M) into a first fraction (5) and a second fraction (6), the first fraction (5) having a higher concentration of 3-methylbutanal than the second fraction (6) and a lower concentration of conversion products in the form of high boilers and 3-methylbutanoic acid than the second fraction (6),

- means for recycling at least a substream of the resulting retentate (3) of the nanofiltration device (M) into the hydroformylation reactor (R).

17. Apparatus according to Claim 16, additionally comprising

- an apparatus (P) for determination of the concentration of 3-methylbutanoic acid in the retentate (3) of the nanofiltration device (M), preferably additionally a device (P) for determination of the concentration of 3-methylbutanoic acid in the resulting product mixture (2) of the hydroformylation and/or in the feed of the nanofiltration device (M) and/or a device (P) for determination of the concentration of 3-methylbutanoic acid in the permeate (4) of the nanofiltration device (M),

or

- a device (P) for taking of samples from the retentate (3) of the nanofiltration device (M), preferably additionally a device (P) for taking of samples from the resulting product mixture (2) of the hydroformylation and/or from the feed of the nanofiltration device (M) and/or a device (P) for taking of samples from the permeate (4) of the nanofiltration device (M).

18. Process according to Claim 16 or 17, characterized in that the nanofiltration device (M) comprises one or more nanofiltration membranes, the or at least one of the nanofiltration membranes having a separation limit in the range from 150 to 2000 g/mol, preferably from 200 to 600 g/mol and most preferably from 350 to 500 g/mol.

19. Use of an apparatus according to any of Claims 16 to 18 for preparation of a product mixture (2) by industrial hydroformylation of an isobutene-containing hydrocarbon stream (1) and for separation of the resulting product mixture (2).

Claims

1. Process for preparing a product mixture (2) by industrial hydroformylation of an isobutene-containing hydrocarbon stream (1) and for separating the resulting product mixture (2), comprising the following steps:

a) hydroformylating the isobutene-containing hydrocarbon stream (1) in a hydroformylation reactor (R) in the presence of a transition metal complex catalyst, so as to obtain a product mixture (2) comprising at least 3-methylbutanal, conversion products in the form of high boilers and 3-methylbutanoic acid, and the transition metal complex catalyst along with the free ligands thereof,

b) separating the product mixture (2) by means of a nanofiltration device (M) comprising one or more membrane separation stages, whereby each separation stage comprises at least one membrane module, each membrane module comprising an individual or more than one nanofiltration membrane, such that the transition metal complex catalyst and the free ligands thereof are enriched in the resulting retentate (3) of the nanofiltration device (M) with respect to 3-methylbutanal and 3-methylbutanoic acid, and such that 3-methylbutanal and 3-methylbutanoic acid are each enriched in the resulting permeate (4) of the nanofiltration device (M) with respect to the transition metal complex catalyst, the concentration of the 3-methylbutanoic acid being lower in the resulting retentate (3) than in the permeate (4),

c) separating the resulting permeate (4) of the nanofiltration device (M) by means of a thermal separating device (D) comprising one or more separation stages into at least one first fraction (5) and a second fraction (6), the first fraction (5) having a higher concentration of 3-methylbutanal than the second fraction (6) and a lower concentration of conversion products in the form of high boilers and 3-methylbutanoic acid than the second fraction (6),

d) recycling at least a substream of the resulting retentate (3) of the nanofiltration device (M) into the hydroformylation reactor (R).

2. Process according to Claim 1, characterized in that one or more process parameters in the step of hydroformylating the isobutenic hydrocarbon stream are set such that the total concentration of conversion products in the form of high boilers and 3-methylbutanoic acid, based on the weight of the overall product mixture, is 30% by weight or less, whereby

the parameter(s) to be set being selected from the group consisting of pressure, temperature, mean residence time, composition of the synthesis gas, concentration of the transition metal and transition metal-ligand ratio of the transition metal complex catalyst, whereby

the step of hydroformylating the isobutenic hydrocarbon stream is performed

- at a pressure in the range from 0.2 to 8 MPa,

and

- at a temperature in the range from 70 to 130°C

and

- with a mean residence time in the range from 1 to 4 h

and

- a synthesis gas composition $CO:H_2$ of 1:3 to 3:1

and

- a transition metal concentration in the range from 10 to 100 ppm

and

- a transition metal/ligand ratio in the range from 1:4 to 1:50.

3. Process according to any of the preceding claims, characterized in that

- the transition metal of the transition metal complex catalyst is rhodium

and/or

- the ligand(s) of the transition metal complex catalyst is/are selected from the group of the organophosphorus ligands.

4. Process according to any of the preceding claims, characterized in that one or more nanofiltration membranes are used in the nanofiltration device, the or at least one of the nanofiltration membranes having a retention for 3-methylbutanoic acid of -1 or less, more preferably -5 or less and especially -10 or less.

5. Process according to any of the preceding claims, characterized in that one or more nanofiltration membranes which are used in the nanofiltration device comprise or consist of one or more polymers containing imide groups, the polymer(s) containing imide groups being selected such that the retention of the nanofiltration membrane for 3-methylbutanoic acid is -1 or less, more preferably -5 or less, and especially -10 or less.

6. Process according to any of the preceding claims, characterized in that one or more nanofiltration membranes are used in the nanofiltration device, the or at least one of the nanofiltration membranes having a separation limit in the range from 150 to 2000 g/mol, preferably 200 to 600 g/mol and most preferably 350 to 500 g/mol.

7. Process according to any of the preceding claims, characterized in that one or more nanofiltration membranes which are used in the nanofiltration device comprise or consist of one or more polymers containing imide groups, the polymer(s) containing imide groups being selected such that the separation limit of the nanofiltration membrane is in the range from 150 to 2000 g/mol.

8. Process according to any of the preceding claims, characterized in that the step of separating the product mixture obtained by hydroformylating the isobutenic hydrocarbon stream in the nanofiltration device is performed

- at a temperature in the range from 10 to 150°C

and/or

- at a transmembrane pressure in the range from 0.5 to 6 MPa

and/or

- at a Reynolds number between 55 and 13 500, preferably between 100 and 3500 and most preferably between 170 and 900,

and/or

- in the presence of carbon monoxide and/or hydrogen, preferably at a partial carbon monoxide pressure of at least 200 kPa in the feed, in the retentate and in the permeate of each membrane separation stage.

9. Process according to any of the preceding claims, characterized in that the step of thermal separation comprises a distillation in which the first fraction is obtained as the top product and the second fraction as the bottom product.

10. Process according to any of the preceding claims, comprising the steps of:
- hydroformylating an isobutenic hydrocarbon stream in the presence of a rhodium complex catalyst having one or more organophosphorus ligands at a pressure in the range from 0.2 to 8 MPa and a temperature in the range from 70 to 130°C with a mean residence time in the range from 1 to 4 h, a synthesis gas composition $CO:H_2$ of 1:3 to 3:1, a rhodium concentration in the reactor in the range from 10 to 100 ppm and a rhodium/ligand ratio in the range from 1:4 to 1:50;
- separating the resulting product mixture of the hydroformylation by means of a nanofiltration device at a temperature in the range from 10 to 150°C, a transmembrane pressure in the range from 0.5 to 6 MPa, at a Reynolds number between 170 and 900, and a partial carbon monoxide pressure of greater than 0.2 MPa into feed, retentate and permeate of each membrane separation stage;
- thermally separating the resulting permeate of the nanofiltration device by means of distillation into a first fraction and a second fraction, the first fraction containing a higher concentration of 3-methylbutanal than the second fraction and a lower concentration of high boilers than the second fraction,
- recycling at least a substream of the resulting retentate of the nanofiltration device into the hydroformylation reactor.

11. Process according to any of the preceding claims, comprising the step of
- monitoring the concentration of 3-methylbutanoic acid in the resulting retentate of the nanofiltration device, and preferably also in the product mixture and/or in the resulting permeate of the nanofiltration device.

12. Process according to Claim 11, characterized in that, on exceedance of a fixed maximum concentration of the 3-methylbutanoic acid in the resulting retentate of the nanofiltration device, the recycling of the resulting retentate into the hydroformylation reactor is stopped.

13. Process according to Claim 11 or 12, characterized in that, on exceedance of a fixed maximum concentration of 3-methylbutanoic acid in a feed of the nanofiltration device,

one or more process parameters in the step of hydroformylating the isobutenic hydrocarbon stream in the hydroformylation reactor are modified such that the concentration of 3-methylbutanoic acid in the feed of the nanofiltration device is lowered below the fixed maximum.

14. Apparatus (1) for performance of the process according to any of Claims 1 to 13, comprising

- a hydroformylation reactor (R) for hydroformylation of an isobutenic hydrocarbon stream (1) in the presence of a transition metal complex catalyst,
- a nanofiltration device (M) comprising one or more membrane separation stages for separation of the product mixture (2) formed in the hydroformylation reactor (R), such that the transition metal complex catalyst and the free ligands thereof are enriched in the resulting retentate (3) of the nanofiltration device (M) with respect to 3-methylbutanal and 3-methylbutanoic acid, and 3-methylbutanal and 3-methylbutanoic acid are enriched in the resulting permeate of the nanofiltration device (M) with respect to the transition metal complex catalyst, such that the concentration of 3-methylbutanoic acid in the resulting retentate (3) is lower than in the resulting permeate (4), the nanofiltration device (M) comprising one or more nanofiltration membranes, the or at least one of the nanofiltration membranes having a retention for 3-methylbutanoic acid of -1 or less, more preferably -5 or less, and especially -10 or less,
- at least one thermal separation device (D) for thermal separation of the resulting permeate (4) of the nanofiltration device (M) into a first fraction (5) and a second fraction (6), the first fraction (5) having a higher concentration of 3-methylbutanal than the second fraction (6) and a lower concentration of conversion products in the form of high boilers and 3-methylbutanoic acid than the second fraction (6),
- means for recycling at least a substream of the resulting retentate (3) of the nanofiltration device (M) into the hydroformylation reactor (R).

15. Apparatus according to Claim 14, additionally comprising

- an apparatus (P) for determination of the concentration of 3-methylbutanoic acid in the retentate (3) of the nanofiltration device (M), preferably additionally a device (P) for determination of the concentration of 3-methylbutanoic acid in the resulting product mixture (2) of the hydroformylation and/or in the feed of the nanofiltration

device (M) and/or a device (P) for determination of the concentration of 3-methylbutanoic acid in the permeate (4) of the nanofiltration device (M),

or

- a device (P) for taking of samples from the retentate (3) of the nanofiltration device (M), preferably additionally a device (P) for taking of samples from the resulting product mixture (2) of the hydroformylation and/or from the feed of the nanofiltration device (M) and/or a device (P) for taking of samples from the permeate (4) of the nanofiltration device (M).

16. Apparatus according to Claim 14 or 15, characterized in that the nanofiltration device (M) comprises one or more nanofiltration membranes, the or at least one of the nanofiltration membranes having a separation limit in the range from 150 to 2000 g/mol, preferably from 200 to 600 g/mol and most preferably from 350 to 500 g/mol.

17. Use of an apparatus according to any of Claims 14 to 16 for preparation of a product mixture (2) by industrial hydroformylation of an isobutene-containing hydrocarbon stream (1) and for separation of the resulting product mixture (2).