

Claims

1. A continuous process for the production of propylene oxide comprising reacting propene with hydrogen peroxide in methanolic solution in the presence of a titanium silicalite-1 catalyst to obtain propylene oxide, wherein a reaction feed comprising propene, methanol and hydrogen peroxide is introduced into a reactor, said reaction feed containing potassium cations (K^+) in an amount of from 110 to 190 micromol, relative to 1 mol hydrogen peroxide contained in the reaction feed, and further containing phosphorus (P) in the form of anions of at least one phosphorus oxyacid.
2. The process of claim 1, wherein the reaction feed contains K^+ in an amount of from 120 to 175 micromol, preferably of from 130 to 160 micromol, relative to 1 mol hydrogen peroxide contained the reaction feed.
3. The process of claim 1 or 2, wherein in the reaction feed, the molar ratio of K^+ relative to P is in the range of from 0.75 to 1.0, preferably of from 0.8 to 0.95.
4. The process of any of claims 1 to 3, wherein the reaction feed additionally contains sodium ions (Na^+).
5. The process of claim 4, wherein in the reaction feed, the molar ratio of K^+ relative to Na^+ is greater than or equal to 0.1.
6. The process of claim 4 or 5, wherein in the reaction feed, the molar ratio of K^+ relative to Na^+ is in the range of from 1.0 to 3.0, preferably from 1.2 to 1.75, more preferably of from 1.3 to 1.6.
7. The process of any of claims 4 to 6, wherein in the reaction feed, the molar ratio of K^+ plus Na^+ relative to P is in the range of from 1.43 to 1.53, preferably from 1.44 to 1.52, more preferably of from 1.45 to 1.51.
8. The process of any of claims 1 to 7, wherein the total amount of (a) alkali metal ions other than K^+ and Na^+ , (b) alkaline earth metal ions, and (c) other metal ions contained in the reaction feed is 5 micromol at most, preferably of 3 micromol at most, more preferably of 1 micromol at most, relative to 1 mol hydrogen peroxide contained the reaction feed.
9. The process of any of claims 1 to 8, wherein the reaction feed is obtained from a hydrogen peroxide feed, a methanol feed, and a propene feed.

10. The process of claim 9, wherein the hydrogen peroxide feed contains K^+ in an amount of less than 110 micromol, preferably less than 70 micromol, more preferably less than 30 micromol, in particular less than 5 micromol, relative to 1 mol hydrogen peroxide contained in the hydrogen peroxide feed.
- 5 11. The process of claim 10, wherein at least one solution containing K^+ and P in the form of anions of at least one phosphorus oxyacid is added to the hydrogen peroxide feed or to the propene feed or to the methanol feed or a mixed feed of two or three thereof, in such an amount that the reaction feed contains K^+ , Na^+ , and P in the form of anions of at least one phosphorus oxyacid in amounts as defined in
- 10 12. The process of claim 11, wherein the at least one solution is an aqueous solution of dipotassium hydrogen phosphate.
- 15 13. The process of any of claims 9 to 12, wherein the hydrogen peroxide feed is an aqueous or a methanolic or an aqueous/methanolic, preferably an aqueous hydrogen peroxide feed, containing hydrogen peroxide preferably in an amount of from 25 to 75 wt.-%, more preferably of from 30 to 50 wt.-%.
- 20 14. The process of any of claims 9 to 13, wherein the propene feed additionally contains propane wherein the volume ratio of propene to propane is preferably in the range of from 99.99:0.01 to 95:5.
- 25 15. The process of any of claims 1 to 14, wherein the reaction feed when introduced into the reactor consists of one liquid phase.
- 30 16. The process of any of claims 1 to 15, wherein the pressure under which the reaction of propene with hydrogen peroxide in methanolic solution in the presence of the titanium silicalite-1 catalyst is carried out in the reactor is at least 10 bar, preferably at least 15 bar, more preferably at least 20 bar and in particular in the range of from 20 to 40 bar.
- 35 17. The process of any of claims 1 to 16, wherein the reaction mixture in the reactor is externally and/or internally cooled such that the maximum temperature of the reaction mixture in the reactor is in the range of from 30 to 70 °C.
- 40 18. The process of any of claims 1 to 17, wherein the titanium silicalite-1 catalyst contains titanium silicalite-1 as catalytically active material embedded in a porous matrix, preferably in a mesoporous matrix, more preferably in a mesoporous silica matrix.

19. The process of any of claims 1 to 18, wherein the reaction of propene with hydrogen peroxide in methanolic solution in the presence of the titanium silicalite-1 catalyst is carried out by a method comprising
- 5 (i) reacting propene with hydrogen peroxide in methanolic solution in the presence of the titanium silicalite-1 catalyst in at least one reactor R1 which is preferably operated in isothermal mode, wherein a reaction feed comprising propene, methanol and hydrogen peroxide is introduced into R1, said reaction feed containing potassium cations (K^+) in an amount of from 110 to 190 micromol, relative to 1 mol hydrogen peroxide contained in the reaction
- 10 feed, and further containing phosphorus (P) in the form of anions of at least one phosphorus oxyacid;
- (ii) separating a stream containing non-reacted hydrogen peroxide from the reaction mixture obtained from (i) and removed from R1, said separating preferably being carried out by distillation in at least 1, preferably 1 distillation column K1;
- 15 (iii) mixing the stream containing non-reacted hydrogen peroxide with a propene stream, passing the mixed stream into at least 1, preferably 1 reactor R2 containing the titanium silicalite-1 catalyst and preferably being operated
- 20 in adiabatic mode, and reacting propene with hydrogen peroxide in R2, wherein the hydrogen peroxide conversion in R1 is preferably in the range of from 85 to 95 %, more preferably in the range of from 87 to 93 %.
20. The process of claim 19, further comprising
- 25 (iv) distilling a combined stream consisting of the reaction mixture obtained from (iii) and removed from R2, and the top stream obtained from K1, in a distillation column K2 to obtain a top stream having a propylene oxide content of 100 vol.-ppm at most and a bottoms stream having a propene content of 100 wt.-ppm at most, said distilling preferably being carried out using a methanol stream as external reflux;
- 30 (v) distilling the bottoms stream obtained from (iv) in a distillation column K3 to obtain a top stream mixture containing at least 98 wt.-% of propylene oxide and a bottoms stream having a propylene oxide content of 100 wt.-ppm at most.
- 35 21. The process of claim 20, further comprising
- (vi) distilling the bottoms stream obtained from (v) in at least one distillation column K4 to obtain a bottoms stream having a methanol content of 0.5 wt.-% at most and a top stream having a methanol content of at least 98.5 wt.-% and a water content of 1 wt.-% at most;
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(vii) recycling the top stream obtained from (vi) as starting material of the continuous process according to any of claims 1 to 20 and/or as external reflux into (iv) according to claim 20.

- 5 22. A process for the preparation of hydrogen peroxide, comprising
- preparing a reaction mixture via an anthrachinone process;
 - preparing a crude hydrogen peroxide solution by subjecting the reaction
 - 10 mixture to an extraction stage using water, preferably essentially pure water as extracting agent, said crude hydrogen peroxide solution having a K^+ content of less than 110 micromol;
 - optionally subjecting the crude hydrogen peroxide solution to a further extraction stage to obtain a crude washed hydrogen peroxide solution;
 - adding a solution containing K^+ , preferably containing K^+ and P in the form of anions of at least one phosphorus oxyacid, to the crude hydrogen peroxide
 - 15 solution or to the crude washed hydrogen peroxide solution.
23. A mixture containing at least 99.99 wt.-% of propylene oxide, comprising less than 3 wt.-ppm acetaldehyde, preferably being free of acetaldehyde.
- 20 24. The mixture of claim 23, obtainable or obtained by a process according to claim 20 as top stream mixture in (v).

Claims

1. A continuous process for the production of propylene oxide comprising reacting propene with hydrogen peroxide in methanolic solution in the presence of a titanium silicalite-1 catalyst to obtain propylene oxide, wherein a reaction feed comprising propene, methanol and hydrogen peroxide is introduced into a reactor, said reaction feed containing potassium cations (K^+) in an amount of from 110 to 190 micromol, relative to 1 mol hydrogen peroxide contained in the reaction feed, and further containing phosphorus (P) in the form of anions of at least one phosphorus oxyacid.
2. The process of claim 1, wherein the reaction feed contains K^+ in an amount of from 120 to 175 micromol, preferably of from 130 to 160 micromol, relative to 1 mol hydrogen peroxide contained the reaction feed.
3. The process of claim 1 or 2, wherein in the reaction feed, the molar ratio of K^+ relative to P is in the range of from 0.75 to 1.0, preferably of from 0.8 to 0.95.
4. The process of any of claims 1 to 3, wherein the reaction feed additionally contains sodium ions (Na^+).
5. The process of claim 4, wherein in the reaction feed, the molar ratio of K^+ relative to Na^+ is greater than or equal to 0.1.
6. The process of claim 4 or 5, wherein in the reaction feed, the molar ratio of K^+ relative to Na^+ is in the range of from 1.0 to 3.0, preferably from 1.2 to 1.75, more preferably of from 1.3 to 1.6.
7. The process of any of claims 4 to 6, wherein in the reaction feed, the molar ratio of K^+ plus Na^+ relative to P is in the range of from 1.43 to 1.53, preferably from 1.44 to 1.52, more preferably of from 1.45 to 1.51.
8. The process of any of claims 1 to 7, wherein the total amount of (a) alkali metal ions other than K^+ and Na^+ , (b) alkaline earth metal ions, and (c) other metal ions contained in the reaction feed is 5 micromol at most, preferably of 3 micromol at most, more preferably of 1 micromol at most, relative to 1 mol hydrogen peroxide contained the reaction feed.
9. The process of any of claims 1 to 8, wherein the reaction feed is obtained from a hydrogen peroxide feed, a methanol feed, and a propene feed.
10. The process of claim 9, wherein the hydrogen peroxide feed contains K^+ in an amount of less than 110 micromol, preferably less than 70 micromol, more preferably less than

30 micromol, in particular less than 5 micromol, relative to 1 mol hydrogen peroxide contained in the hydrogen peroxide feed.

11. The process of claim 10, wherein at least one solution containing K^+ and P in the form of anions of at least one phosphorus oxyacid is added to the hydrogen peroxide feed or to the propene feed or to the methanol feed or a mixed feed of two or three thereof, in such an amount that the reaction feed contains K^+ , Na^+ , and P in the form of anions of at least one phosphorus oxyacid in amounts as defined in any of claims 1 to 7.
12. The process of claim 11, wherein the at least one solution is an aqueous solution of dipotassium hydrogen phosphate.
13. The process of any of claims 9 to 12, wherein the hydrogen peroxide feed is an aqueous or a methanolic or an aqueous/methanolic, preferably an aqueous hydrogen peroxide feed, containing hydrogen peroxide preferably in an amount of from 25 to 75 wt.-%, more preferably of from 30 to 50 wt.-%.
14. The process of any of claims 9 to 13, wherein the propene feed additionally contains propane wherein the volume ratio of propene to propane is preferably in the range of from 99.99:0.01 to 95:5.
15. The process of any of claims 1 to 14, wherein the reaction feed when introduced into the reactor consists of one liquid phase.
16. The process of any of claims 1 to 15, wherein the pressure under which the reaction of propene with hydrogen peroxide in methanolic solution in the presence of the titanium silicalite-1 catalyst is carried out in the reactor is at least 10 bar, preferably at least 15 bar, more preferably at least 20 bar and in particular in the range of from 20 to 40 bar.
17. The process of any of claims 1 to 16, wherein the reaction mixture in the reactor is externally and/or internally cooled such that the maximum temperature of the reaction mixture in the reactor is in the range of from 30 to 70°C.
18. The process of any of claims 1 to 17, wherein the titanium silicalite-1 catalyst contains titanium silicalite-1 as catalytically active material embedded in a porous matrix, preferably in a mesoporous matrix, more preferably in a mesoporous silica matrix.
19. The process of any of claims 1 to 18, wherein the reaction of propene with hydrogen peroxide in methanolic solution in the presence of the titanium silicalite-1 catalyst is carried out by a method comprising
 - (i) reacting propene with hydrogen peroxide in methanolic solution in the presence of the titanium silicalite-1 catalyst in at least one reactor R1 which is preferably

operated in isothermal mode, wherein a reaction feed comprising propene, methanol and hydrogen peroxide is introduced into R1, said reaction feed containing potassium cations (K^+) in an amount of from 110 to 190 micromol, relative to 1 mol hydrogen peroxide contained in the reaction feed, and further containing phosphorus (P) in the form of anions of at least one phosphorus oxyacid;

- (ii) separating a stream containing non-reacted hydrogen peroxide from the reaction mixture obtained from (i) and removed from R1, said separating preferably being carried out by distillation in at least 1, preferably 1 distillation column K1;
- (iii) mixing the stream containing non-reacted hydrogen peroxide with a propene stream, passing the mixed stream into at least 1, preferably 1 reactor R2 containing the titanium silicalite-1 catalyst and preferably being operated in adiabatic mode, and reacting propene with hydrogen peroxide in R2,

wherein the hydrogen peroxide conversion in R1 is preferably in the range of from 85 to 95 %, more preferably in the range of from 87 to 93 %.

20. The process of claim 19, further comprising

- (iv) distilling a combined stream consisting of the reaction mixture obtained from (iii) and removed from R2, and the top stream obtained from K1, in a distillation column K2 to obtain a top stream having a propylene oxide content of 100 vol.-ppm at most and a bottoms stream having a propene content of 100 wt.-ppm at most, said distilling preferably being carried out using a methanol stream as external reflux;
- (v) distilling the bottoms stream obtained from (iv) in a distillation column K3 to obtain a top stream mixture containing at least 98 wt.-% of propylene oxide and a bottoms stream having a propylene oxide content of 100 wt.-ppm at most.

21. The process of claim 20, further comprising

- (vi) distilling the bottoms stream obtained from (v) in at least one distillation column K4 to obtain a bottoms stream having a methanol content of 0.5 wt.-% at most and a top stream having a methanol content of at least 98.5 wt.-% and a water content of 1 wt.-% at most;
- (vii) recycling the top stream obtained from (vi) as starting material of the continuous process according to any of claims 1 to 20 and/or as external reflux into (iv) according to claim 20.