Claims

~~New claims (clean copy)~~

1. A process for the preparation of a zeolitic material having an MFI-type framework structure, comprising the steps of:

(1) providing a mixture comprising one or more sources for YO_2 and one or more alkenyltrialkylammonium cation $R^1R^2R^3R^4N^+$-containing compounds as structure directing agent; and

(2) crystallizing the mixture obtained in step (1) to obtain a zeolitic material;

wherein Y is a tetravalent element, and

wherein R^1, R^2, and R^3 independently from one another stand for n-propyl; and R^4 stands for 2-propen-1-yl or 1-propen-1-yl.

2. The process of claim 1, wherein the structure directing agent provided in step (1) comprises *N*-(2-propen-1-yl)-tri-n-propylammonium hydroxide and/or *N*-(1-propen-1-yl)-tri-n-propylammonium hydroxide.

3. The process of claim 1 or 2, wherein the mixture provided in step (1) comprises two or more $R^1R^2R^3R^4N^+$-containing compounds, wherein R^4 of the two or more compounds are different from one another and stand for 2-propen-1-yl and 1-propen-1-yl.

4. The process of claim 3, wherein the mixture provided in step (1) comprises two $R^1R^2R^3R^4N^+$-containing compounds, wherein R^4 of the first compound (A) contains an end-chain $-CH=CH_2$ moiety, and R^4 of the second compound (B) contains an end-chain moiety $-CH_3$, and

wherein the molar ratio A : B in the mixture provided in step (1) is preferably comprised in the range of from 25 : 75 to 99 : 1, more preferably of from 50 : 50 to 98 : 2, more preferably of from 75 : 25 to 97 : 3, more preferably of from 85 : 15 to 96 : 4, and even more preferably of from 90 : 10 to 95 : 5.

5. The process of claim 4, wherein the compounds A and B are constitutional isomers with respect to the position of the double bond in R^4, and wherein said constitutional isomers are *N*-(2-propen-1-yl)-tri-n-propylammonium hydroxide and *N*-(1-propen-1-yl)-tri-n-propylammonium hydroxide.

6. The process of any of claims 1 to 5, wherein Y is selected from the group consisting of Si, Sn, Ti, Zr, Ge, and mixtures of two or more thereof, Y preferably being Si.

7. The process of any of claims 1 or 6, wherein the one or more sources for YO_2 comprises one or more compounds selected from the group consisting of fumed silica, silica hydrosols, reactive amorphous solid silicas, silica gel, silicic acid, water glass, sodium metasilicate hydrate, sesquisilicate, disilicate, colloidal silica, pyrogenic silica, silicic acid esters, and mixtures of two or more thereof, preferably from the group consisting of fumed silica, silica hydrosols, reactive amorphous solid silicas, silica gel, colloidal silica, pyrogenic silica, tetraalkoxysilanes, and mixtures of two or more thereof, more preferably from the group consisting of fumed silica, reactive amorphous solid silicas, silica gel, pyrogenic silica, $(C_1\text{-}C_3)$tetraalkoxysilanes, and mixtures of two or more thereof, more preferably from the group consisting of fumed silica, $(C_1\text{-}C_2)$tetraalkoxysilanes, and mixtures of two or more thereof, wherein even more preferably the one or more sources for YO_2 comprises fumed silica and/or tetraethoxysilane.

8. The process of any of claims 1 to 7, wherein the mixture provided in step (1) further comprises one or more sources for X_2O_3, wherein X is a trivalent element.

9. The process of claim 8, wherein X is selected from the group consisting of Al, B, In, Ga, and mixtures of two or more thereof, X preferably being Al and/or B, and more preferably being Al.

10. The process of claim 8 or 9, wherein the one or more sources for X_2O_3 comprises one or more compounds selected from the group consisting of aluminum, aluminum alkoxides, alumina and/or one or more aluminates, preferably one or more aluminates, more preferably one or more aluminate salts selected from the group consisting of alkali metal aluminates, aluminum hydroxide, and mixtures of two or more thereof, wherein more preferably the one or more sources for X_2O_3 comprises aluminum hydroxide and/or one or more alkali metal aluminates, the alkali metal preferably being sodium and/or potassium, more preferably sodium.

11. The process of any of claims 8 to 10, wherein the $YO_2 : X_2O_3$ molar ratio of the mixture according to step (1) ranges from 0.5 to 500, preferably from 1 to 400, more preferably from 5 to 300, more preferably from 20 to 200, more preferably from 30 to 150, more preferably from 35 to 120, and even more preferably from 40 to 100.

12. The process of any of claims 1 to 7, wherein the mixture provided in step (1) does not contain a source for X_2O_3, wherein X is a trivalent element.

13. The process of any of claims 1 to 12, wherein the mixture according to step (1) further comprises one or more solvents, wherein said one or more solvents preferably comprises water, more preferably distilled water.

14. The process of claim 13, wherein the $H_2O : YO_2$ molar ratio of the mixture according to step (1) ranges from 3 to 100, more preferably from 5 to 50, more preferably from 6 to 30, more preferably from 7 to 20, more preferably from 8 to 15, and even more preferably from 9 to 12.

15. The process of any of claims 1 to 14, wherein the molar ratio of the one or more alkenyltrialkylammonium cations $R^1R^2R^3R^4N^+ : YO_2$ in the mixture provide according to step (1) ranges from 0.01 to 5, preferably from 0.03 to 2, more preferably from 0.05 to 1, more preferably from 0.1 to 0.5, more preferably from 0.13 to 0.3, and even more preferably from 0.15 to 0.2.

16. The process of any of claims 1 to 15, wherein the mixture according to step (1) further comprises one or more sources of one or more elements suitable for isomorphous substitution of at least a portion of the Y atoms and/or of the X atoms in the zeolite framework structure, wherein the one or more elements are preferably selected from the group consisting of B, Fe, Ti, Sn, Ga, Ge, Zr, V, Nb, Cu, Zn, Li, Be, and mixtures of two or more thereof, more preferably wherein the one or more elements are selected from the group consisting of B, Fe, Ti, Sn, Zr, Cu, and mixtures of two or more thereof, wherein even more preferably the one or more element is Ti and/or B, preferably Ti.

17. The process of claim 16, wherein the one or more sources for isomorphous substitution preferably comprises one or more titania precursor compounds, preferably one or more compounds selected from the group consisting of titanic

acids, titanic acid esters, and mixtures of two or more thereof, wherein more preferably the one or more sources for isomorphous substitution comprises one or more titanic acid esters, preferably one or more tetraalkoxytitanates, more preferably one or more (C_1-C_3)tetraalkoxytitanates, more preferably one or more (C_1-C_2)tetraalkoxytitanates, wherein even more preferably the one or more sources for isomorphous substitution comprises tetraethoxytitanate.

18. The process of claim 16 or 17, wherein the molar ratio of YO_2 to the one or more elements suitable for isomorphous substitution of at least a portion of the Y atoms and/or of the X atoms in the zeolite framework structure ranges from 1 to 300, more preferably from 5 to 150, more preferably from 10 to 100, more preferably from 20 to 70, more preferably from 25 to 50, and even more preferably from 30 to 40.

19. The process of any of claims 1 to 18, wherein the crystallization in step (2) involves heating of the mixture, preferably at a temperature ranging from 90 to 210°C, preferably from 110 to 200°C, more preferably from 130 to 190°C, more preferably from 145 to 180°C, and even more preferably from 155 to 170°C.

20. The process of claim 19, wherein the crystallization in step (2) is conducted under solvothermal conditions, preferably under hydrothermal conditions.

21. The process of claim 19 or 20, wherein the crystallization in step (2) involves heating of the mixture for a period ranging from 5 to 120 h, more preferably from 8 to 80 h, more preferably 10 to 50 h, more preferably from 13 to 35 h, and even more preferably from 15 to 25 h.

22. The process of any of claims 1 to 21, wherein the crystallization in step (2) involves agitating the mixture, preferably by stirring.

23. The process of any of claims 1 to 22 further comprising one or more of the following steps of

(3) isolating the zeolitic material, preferably by filtration,

and/or

(4) washing the zeolitic material,

and/or

(5) drying the zeolitic material,

and/or

(6) subjecting the zeolitic material to an ion-exchange procedure,
wherein the steps (3) and/or (4) and/or (5) and/or (6) can be conducted in any order, and
wherein one or more of said steps is preferably repeated one or more times.

24. The process of claim 23, wherein in the at least one step (6) one or more ionic non-framework elements contained in the zeolite framework is ion-exchanged, preferably against one or more cations and/or cationic elements, wherein the one or more cation and/or cationic elements are preferably selected from the group consisting of H^+, NH_4^+, Mg, Sr, Zr, Cr, Mo, Fe, Co, Ni, Cu, Zn, Ru, Rh, Pd, Ag, Os, Ir, Pt, Au, and mixtures of two or more thereof, more preferably from the group consisting of H^+, NH_4^+, Mg, Sr, Cr, Mo, Fe, Co, Ni, Cu, Zn, Ag, and mixtures of two or more thereof, more preferably from the group consisting of H^+, NH_4^+, Mg, Cr, Mo, Fe, Ni, Cu, Zn, Ag, and mixtures of two or more thereof, and even more preferably from the group consisting of Mg, Mo, Fe, Ni, Cu, Zn, Ag, and mixtures of two or more thereof,
wherein the one or more ionic non-framework elements preferably comprise H^+ and/or an alkali metal, the alkali metal preferably being selected from the group consisting of Li, Na, K, Cs, and combinations of two or more thereof, more preferably from the group consisting of Li, Na, K, and combinations of two or more thereof, wherein more preferably the alkali metal is Na and/or K, even more preferably Na.

25. A synthetic zeolitic material having an MFI-type framework structure obtainable and/or obtained according to any one of claims 1 to 24.

26. The synthetic zeolitic material of claim 25 comprising YO_2 and optionally comprising X_2O_3,
wherein Y is a tetravalent element, and X is a trivalent element,
said material having an X-ray diffraction pattern comprising at least the following reflections:

Intensity (%)	Diffraction angle 2θ/° [Cu K(alpha 1)]
15 - 55	7.88 - 8.16
11 - 35	8.83 - 9.13
100	23.04 - 23.46
27 - 40	23.68 - 23.89

21 - 66	23.90 - 24.23
22 - 44	24.29 - 24.71

wherein 100% relates to the intensity of the maximum peak in the X-ray powder diffraction pattern.

27. The zeolitic material of claim 26, wherein the ^{29}Si MAS NMR of the zeolitic material comprises:

a first peak (P1) in the range of from -110.4 to -114.0, preferably of from -110.8 to -113.4 ppm, and even more preferably of from -111.2 to -112.8 ppm; and

a second peak (P2) in the range of from -101.4 to -106.8 ppm, preferably of from -101.6 to -106.5 ppm, and even more preferably of from -101.8 to -106.2 ppm.

28. The zeolitic material of claim 26 or 27, wherein the ^{27}Al MAS NMR of the zeolitic material comprises:

a first peak (P'1) in the range of from 50.00 to 53.50 ppm, preferably of from 50.50 to 52.80 ppm, more preferably of from 50.70 to 52.50 ppm, and even more preferably of from 50.85 - 52.30 ppm; and

a second peak (P'2) in the range of from -0.50 to -2.00 ppm, preferably of from -0.80 to -1.70 ppm, more preferably of from -1.00 to -1.50 ppm, and even more preferably of from -1.10 to -1.35 ppm;

wherein the integration of the first and second peaks in the ^{27}Al MAS NMR of the zeolitic material preferably offers a ratio of the integration values P'1 : P'2 of 1 : (0.5 – 1.2), preferably of 1 : (0.1 – 0.9), more preferably of 1 : (0.2 – 0.7), more preferably of 1 : (0.3 – 0.6), more preferably of 1 : (0.35 – 0.55), and even more preferably of 1 : (0.4 - 0.5).

29. The synthetic zeolitic material of claim 25 comprising YO_2 and optionally comprising X_2O_3,

wherein Y is a tetravalent element, and X is a trivalent element,

said material having an X-ray diffraction pattern comprising at least the following reflections:

Intensity (%)	Diffraction angle 2θ/° [Cu K(alpha 1)]
55 – 100	7.66 – 8.20
40 – 75	8.58 – 9.05

92 – 100	22.81 – 23.34
49 – 58	23.64 – 24.18
16 – 24	29.64 – 30.21
14 – 25	44.80 – 45.25
16 - 24	45.26 – 45.67

wherein 100% relates to the intensity of the maximum peak in the X-ray powder diffraction pattern.

30. The zeolitic material of claim 29, wherein the ^{29}Si MAS NMR of the zeolitic material comprises:

a first peak (P"1) in the range of from -110.4 to -114.0 ppm, preferably of from -110.8 to -113.4 ppm, and even more preferably of from -111.2 to -112.8 ppm; and

a second peak (P"2) in the range of from -100.2 to -104.2 ppm, preferably of from -100.8 to -103.6 ppm, and even more preferably of from -101.4 to -103.0 ppm.

31. The zeolitic material of claim 27 or 30, wherein the deconvoluted ^{29}Si MAS NMR spectrum comprises one additional peak comprised in the range of from -113.2 to -115.2 ppm, more preferably of from -113.5 to -114.9 ppm, and even more preferably of from -113.8 to -114.7 ppm.

32. The zeolitic material of any of claims 27 to 31, wherein at least a portion of the Y atoms and/or of the X atoms in the MFI-type framework structure is isomorphously substituted by one or more elements, wherein the one or more elements are preferably selected from the group consisting of B, Fe, Ti, Sn, Ga, Ge, Zr, V, Nb, Cu, Zn, Li, Be, and mixtures of two or more thereof, more preferably wherein the one or more elements are selected from the group consisting of B, Fe, Ti, Sn, Zr, Cu, and mixtures of two or more thereof, wherein even more preferably the one or more element is Ti and/or B, preferably Ti.

33. The zeolitic material of claim 32, wherein the molar ratio of YO_2 to the one or more element ranges from 5 to 100, preferably from 10 to 80, more preferably from 20 to 70, more preferably from 25 to 65, more preferably from 30 to 50, more preferably from 35 to 45, and even more preferably from 38 to 40.

34. The zeolitic material of any of claims 26 to 33, wherein the $YO_2 : X_2O_3$ molar ratio ranges from 2 to 200, preferably from 3 to 160, more preferably from 5 to 140, more preferably from 8 to 120, more preferably from 10 to 100, more preferably from 15 to 80, and even more preferably from 20 to 60.

35. The zeolitic material of any of claims 26 to 34, wherein the MFI-type framework structure of the zeolitic material does not contain X_2O_3.

36. The zeolitic material of any of claims 26 to 35, wherein Y is selected from the group consisting of Si, Sn, Ti, Zr, Ge, and mixtures of two or more thereof, Y preferably being Si.

37. The zeolitic material of any of claims 26 to 34 and 36, wherein X is selected from the group consisting of Al, B, In, Ga, and mixtures of two or more thereof, X preferably being Al and/or B, and more preferably being Al.

38. The zeolitic material of any of claims 26 to 37, wherein the zeolitic material comprises one or more cation and/or cationic elements as ionic non-framework elements, said one or more cation and/or cationic elements preferably comprising one or more selected from the group consisting of H^+, NH_4^+, Mg, Sr, Zr, Cr, Mo, Fe, Co, Ni, Cu, Zn, Ru, Rh, Pd, Ag, Os, Ir, Pt, Au, and mixtures of two or more thereof, more preferably from the group consisting of H^+, NH_4^+, Mg, Sr, Cr, Mo, Fe, Co, Ni, Cu, Zn, Ag, and mixtures of two or more thereof, more preferably from the group consisting of H^+, NH_4^+, Mg, Cr, Mo, Fe, Ni, Cu, Zn, Ag, and mixtures of two or more thereof, and even more preferably from the group consisting of Mg, Mo, Fe, Ni, Cu, Zn, Ag, and mixtures of two or more thereof.

39. The zeolitic material of any of claims 26 to 38, wherein the BET surface area of the zeolitic material determined according to DIN 66135 ranges from 50 to 700 m^2/g, preferably from 100 to 650 m^2/g, more preferably from 200 to 600 m^2/g, more preferably from 300 to 550 m^2/g, more preferably from 350 to 500 m^2/g, more preferably from 390 to 470 m^2/g, more preferably from 420 to 440 m^2/g.

40. Use of an alkenyltrialkylammonium cation $R^1R^2R^3R^4N^+$-containing compound as structure directing agent in the synthesis of zeolitic materials having an MFI-type framework structure, wherein

R^1, R^2, and R^3 independently from one another stand for n-propyl; and
R^4 stands for 2-propen-1-yl or 1-propen-1-yl, and preferably for 2-propen-1-yl.

41. Use of a zeolitic material according to any of claims 25 to 39 as a molecular sieve, as an adsorbent, for ion-exchange, as a catalyst and/or as a catalyst support, preferably as a catalyst for an acid-catalyzed and/or oxidation reaction, more preferably as a catalyst for one or more of an isomerization reaction, preferably the isomerization of one or more hydrocarbons, an alkylation reaction, an acylation reaction, an epoxidation reaction, an ammoxidation reaction, and combinations thereof, wherein even more preferably the catalyst is used as an epoxidation catalyst.