CLAIMS

1. A full-Si molecular sieve having a Q^4/Q^3 of (10-90):1, wherein Q^4 is the peak strength at the chemical shift of −112±2 ppm in the ^{29}Si NMR spectrum of the full-Si molecular sieve, expressed as the peak height relative to the base line; and

Q^3 is the peak strength at the chemical shift of −103±2 ppm in the ^{29}Si NMR spectrum of the full-Si molecular sieve, expressed as the peak height relative to the base line.

2. The full-Si molecular sieve of claim **1**, wherein the Q^4/Q^3 is in a range of (10-70):1.

3. The full-Si molecular sieve of claim **1**, wherein a particle of the full-Si molecular sieve is a single crystal grain or an aggregate of multiple crystal grains, wherein the crystal grain has a size of 200 nm to 2 μm.

4. The full-Si molecular sieve of claim **3**, wherein the crystal grain of the full-Si molecular sieve has a hollow structure, with a radial length of 5-300 nm for the cavity portion of the hollow grain, wherein an adsorption capacity of benzene of the molecular sieve measured under the conditions of 25° C., P/P0=0.10 and 1 hour of adsorption time is at least 70 mg/g.

5. The full-Si molecular sieve of claim **1**, wherein the full-Si molecular sieve is a S-1 molecular sieve, a S-2 molecular sieve, or a β molecular sieve.

6. The full-Si molecular sieve of claim **1**, wherein the full-Si molecular sieve has micropores with a pore diameter of less than 1 nm and mesopores with a pore diameter of 2-8 nm, a volume of mesopores having a pore diameter of 2-8

nm is 0.3-0.8 mL/g, and a volume of micropores having a pore diameter less than 1 nm is 0.12-0.19 mL/g.

7. The full-Si molecular sieve of claim **6**, wherein (the volume of mesopores with a pore diameter of 2-8 nm)/(the volume of mesopores with a pore diameter of 2-8 nm plus the volume of micropores with a pore diameter of less than 1 nm) is in a numerical range of 0.6 to 0.8.

8. The full-Si molecular sieve of claim **6**, wherein (the volume of mesopores with a pore diameter of 2-8 nm)/(the volume of mesopores with a pore diameter of 2-8 nm plus the volume of micropores with a pore diameter of less than 1 nm) is in a numerical range of 0.7 to 0.75.

9. A process for producing caprolactam by rearranging cyclohexanone oxime, comprising the step of contacting cyclohexanone oxime with the full-Si molecular sieve according to claim **1**.

10. The full-Si molecular sieve of claim **1**, wherein the Q^4/Q^3 is in the numerical range of (16-36):1.

11. A process for synthesizing a full-Si molecular sieve, comprising:

(1) mixing a template agent, an organic silicon source, water and an inorganic amine source to form a mixture, subjecting the mixture to hydrolyzation, and removing alcohols from the mixture;

(2) aging a product obtained in the step (1) 15-50° C.; and

(3) subjecting a mixture of the aged product obtained in the step (2) and a solid silicon source to crystallization to obtain a crystalline material.

12. The process of claim **11**, wherein the template agent is selected from the group consisting of an organic quaternary ammonium base, an organic amine, an organic quater-

nary ammonium salt, a long-chain alkyl ammonium compound, and mixtures thereof.

13. The process of claim **12**, wherein the organic quaternary ammonium base is selected from the group consisting of tetrapropylammonium hydroxide, tetrabutylammonium hydroxide, tetraethylammonium hydroxide, and mixtures thereof;

the organic amine is selected from the group consisting of aliphatic amine, aromatic amine, alcoholic amine, and mixtures thereof,

wherein the aliphatic amine has a general formula of $R^3(NH_2)_n$, wherein R^3 is C_{1-4}alkyl or C_{1-4}alkylene, n is 1 or 2,

the alcoholic amine can have a general formula of $(HOR^4)_mNH_{(3-m)}$, wherein R^4 is C_1-C_4alkyl, m is 1, 2, or 3, and

the aromatic amine is selected from the group consisting of aniline, amino toluene, p-phenylene diamine, and mixtures thereof;

the organic quaternary ammonium salt is selected from the group consisting of tetrapropylammonium bromide, tetrabutylammonium bromide, tetraethylammonium bromide, tetrapropylammonium chloride, tetrabutylammonium chloride, tetraethylammonium chloride, and mixtures thereof;

the long-chain alkyl ammonium compound has a formula of R^5NH_3X or $R^5N(R^6)_3X$, wherein R^5 is a C_{12}-C_{18} alkyl, R^6 is a C_1-C_6 (e.g. C_1-C_4)alkyl, wherein three R^6s in $R^5N(R^6)_3X$ are identical or not and X is a monovalent anion.

14. The method of claim **13**, wherein the aliphatic amine is selected from the group consisting of ethylamine, n-butylamine, butylene diamine, hexamethylene diamine, and mixtures thereof,

the alcoholic amine is selected from the group consisting of monoethanolamine, diethanolamine, triethanolamine, and mixtures thereof, and

the long-chain alkyl ammonium compound is selected from the group consisting of a long-chain alkyl trimethyl ammonium chloride, a long-chain alkyl trimethyl ammonium bromide, a long-chain alkyl trimethyl ammonium hydroxide, cetyltrimethylammonium bromide, cetyltrimethylammonium chloride, cetyltrimethylammonium hydroxide, myristyltrimethylammonium bromide, myristyltrimethylammonium chloride, myristyltrimethylammonium hydroxide, dodecyltrimethylammonium bromide, dodecyltrimethylammonium chloride, dodecyltrimethylammonium hydroxide, octadecyltrimethylammonium bromide, octadecyltrimethylammonium chloride, octadecyltrimethylammonium hydroxide, and mixtures thereof.

15. The process of claim **12**, wherein a molar ratio of the template agent to a total silicon source is selected from one or more of the following values:

a molar ratio of the organic quaternary ammonium base to the total silicon source is (0.05-0.36):1,

a molar ratio of the organic amine to the total silicon source is (0-0.45):1,

a molar ratio of the organic quaternary ammonium salt to the total silicon source is (0-0.55):1, and

a molar ratio of the long-chain alkyl ammonium compound to the total silicon source is (0.04-0.45):1,

wherein the total silicon source is a sum of the organic silicon source and the solid silicate source.

16. The process of claim **11**, wherein

a molar ratio of the template agent to the total silicon source is (0.04-0.60):1;

a molar ratio of water to the total silicon source is (3-100):1;

a molar ratio of the inorganic amine source (as NH_4^+) to the total silicon source is (0.01-0.1):1; and

a weight ratio of the aged product to the solid silicon source, calculated based on the weight of SiO_2, is 1:(0.1-10), and

wherein the total silicon source is a sum of the organic silicon source and the solid silicate source.

17. The process of claim **11**, wherein the aging in the step (2) lasts for 1-60 hours.

18. The process of claim **11**, wherein in the step (3), the crystallization is carried out at 110° C. to 200° C. under an autogenous pressure for 2 hours to 20 days.

19. The process of claim **11**, wherein in the step (1), the template agent comprises an organic quaternary ammonium base, optionally an organic amine, and optionally an organic quaternary ammonium salt,

wherein a molar ratio of the organic quaternary ammonium base to a total silicon source is (0.05-0.36):1, a molar ratio of the organic amine to the total silicon source is (0-0.45):1 and a molar ratio of the organic quaternary ammonium salt to the total silicon source is (0-0.45):1,

wherein the total silicon source is a sum of the organic silicon source and the solid silicate source.

20. The process of claim **11**, wherein in the step (1), the template agent is a mixture of an organic quaternary ammonium salt and an organic amine, wherein a molar ratio of the organic quaternary ammonium salt to a total silicon source is (0.04-0.55):1, and a molar ratio of the organic amine to the total silicon source is (0.04-0.45):1, wherein the total silicon source is a sum of the organic silicon source and the solid silicate source.

21. The process of claim **11**, wherein in the step (1), the template agent is a mixture of an organic quaternary ammonium salt and an organic amine, wherein a molar ratio of the organic quaternary ammonium salt to a total silicon source is (0.05-0.30):1, and a molar ratio of the organic amine to the total silicon source is (0.05-0.35):1, wherein the total silicon source is a sum of the organic silicon source and the solid silicate source.

22. The process of claim **11**, wherein in the step (1), the template agent is a mixture of a quaternary ammonium source, an long-chain alkyl ammonium compound, and an optional organic amine,

wherein the quaternary ammonium source is an organic quaternary ammonium base, an organic quaternary ammonium salt, or a mixture thereof,

wherein a molar ratio of the quaternary ammonium source to a total silicon source is (0.04-0.45):1, a molar ratio of the long-chain alkyl ammonium compound to the total silicon source is (0.04-0.45):1, and a molar ratio of the organic amine to the total silicon source is (0-0.40):1, and

wherein the total silicon source is a sum of the organic silicon source and the solid silicate source.

23. The process of claim **22**, wherein in the step (1), the molar ratio of the quaternary ammonium source to the total silicon source is (0.05-0.30):1, and the molar ratio of the long-chain alkyl ammonium compound to the total silicon source is (0.05-0.30): 1.

24. The process of claim **23**, wherein in the step (1), the molar ratio of the quaternary ammonium source to the total silicon source is (0.05-0.20):1, the molar ratio of the organic amine to the total silicon source is (0.05-0.25):1, and the molar ratio of the long-chain alkyl ammonium compound to the total silicon source is (0.05-0.25):1.

25. The process of claim **11**, wherein the full-Si molecular sieve is a S-1 molecular sieve, and the template agent is selected from the group consisting of tetrapropylammonium hydroxide, tetrapropylammonium chloride, tetrapropylammonium bromide, an organic amine, a long-chain alkyl ammonium compound, and mixtures thereof; or

the full-Si molecular sieve is a S-2 molecular sieve, and the template agent is selected from the group consisting of tetrabutylammonium hydroxide, tetrabutylammonium chloride, tetrabutylammonium bromide, an organic amine, a long-chain alkyl ammonium compound, and mixtures thereof; or

the full-Si molecular sieve is a β molecular sieve, and the template agent is selected from the group consisting of tetraethylammonium hydroxide, tetraethylammonium chloride, tetrapropylammonium bromide, an organic amine, a long-chain alkyl ammonium compound, and mixtures thereof.

26. The process of claim **11**, wherein said process further comprises:

(4): recrystallizing the crystalline material obtain in the step (3) in an aqueous solution containing an organic base at a temperature range of 100° C. to 200° C. for a duration of 0.1 day to 10 days, and

wherein a molar ratio of the crystalline material obtained in the step (3) to the organic base is 1: (0.02-0.5) and a molar ratio of the crystalline material obtained in the step (3) to water in the aqueous solution is 1: (2-50).

27. The process of claim **11**, wherein the solid silicon source is carbon white having a specific surface area of 50-400 m^2/g.

28. The process of claim **11**, wherein a molar ratio of the template agent to a total silicon source (as SiO_2) is in a numerical range of (0.08-0.60):1.

29. The process of claim **11**, wherein a molar ratio of the inorganic amine source (as NH_4^+) to the total silicon source (as SiO_2) is in a numerical range of (0.01-0.1):1.

30. The process of claim **11**, wherein the molar ratio of water to the total silicon source (as SiO_2) is in a numerical range of (3-100):1.

31. The process of claim **11**, wherein the weight ratio of the aged product (as SiO_2) to the solid silicon source (as SiO_2) is 1:(1-9).

32. The process of claim **11**, wherein in the step (1), the time for hydrolyzation and removing alcohols is at least 10 minutes.

33. The process of claim **11**, wherein, in the step (1),

hydrolyzation is carry out in a temperature range of 0° C. to 150° C.

34. The process of claim **11**, wherein, in the mixture obtained in the step (1), the weight content of alcohols produced by hydrolyzation is not higher than 10 ppm.

35. The process of claim **11**, wherein, in the step (2), the aging time is in a duration of 1 hour to 60 hours.

36. The process of claim **11**, wherein, in the step (2), the aging temperature is in a temperature range of room temperature to 50° C.

37. The process of claim **11**, wherein, in the step (3), the crystallization temperature is in a temperature range of 110° C. to 200° C.

38. The process of claim **11**, wherein, in the step (3), the crystallization time is in a duration of 2 hours to 20 days.

39. The process of claim **11**, wherein, in the step (3), the crystallization is carried out under an autogenous pressure.

40. The process of claim **11**, wherein, in the step (3), the crystallization is carried out at 100° C. to 130° C. for 0.5 day to 1.5 days, and then at 160° C. to 180° C. for 1 day to 3 days, under an autogenous pressure.

41. The process of claim **11**, wherein the organic silicate is selected from the group consisting of tetramethyl orthosilicate, tetraethyl orthosilicate, tetrabutyl orthosilicate, dimethyl diethyl orthosilicateone, and mixtures thereof.

42. The process of claim **11**, wherein the organic silicon source is an organic silicate having general formula of $Si(OR^1)_4$, and R^1 is a linear or a branched C_1-C_6 alkyl,

the solid silicon source is a high purity silica particle or powder, on a dry basis and by weight, the solid silicon source has a SiO_2 content of more than 99.99 wt %, a

total content by atom of Fe, Al and Na of less than 10 ppm, and

the inorganic amine source is an inorganic ammonium salt, an aqueous ammonia, or a combination thereof.

43. The process of claim **11**, wherein said process further comprises:

(4): recrystallizing the crystalline material obtain in the step (3) in an aqueous solution containing an organic base at a temperature range of 120° C. to 200° C. for a duration of 1 day to 6 days, and

wherein a molar ratio of the crystalline material obtained in the step (3) to the organic base is 1: (0.02-0.5) and a molar ratio of the crystalline material obtained in the step (3) to water in the aqueous solution is 1: (2-50).

44. The process of claim **11**, wherein a molar ratio of the template agent to a total silicon source (as SiO_2) is in the numerical range of (0.10-0.55):1.

45. The process of claim **11**, wherein a molar ratio of the inorganic amine source (as NH_4^+) to the total silicon source (as SiO_2) is in a numerical range of (0.01-0.07): 1.

46. The process of claim **11**, wherein a molar ratio of water to the total silicon source (as SiO_2) is in a numerical range of (6-50):1.

47. The process of claim **11**, wherein, in the step (1), hydrolyzation is carried out in a temperature range of 50° C. to 95° C.

48. The process of claim **11**, wherein, in the step (2), the aging time is in a duration of 2 hour to 50 hours.

49. The process of claim **11**, wherein, in the step (2), the aging temperature is in a temperature range of 26° C. to 30° C.

50. The process of claim **11**, wherein, in the step (3), the crystallization temperature is in a temperature range of 140° C. to 180° C.

51. The process of claim **11**, wherein, in the step (3), the crystallization time is in a duration of 0.5 day to 6 days.