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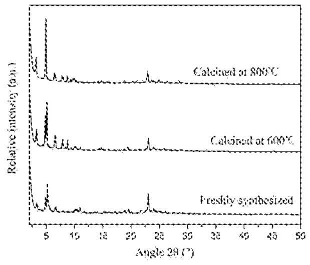
(54) NOVEL SILICATE ZEOLITE ZMQ-1 AND APPLICATION THEREOF.

(57) The present invention relates to zeolites, and in particular to a novel silicate zeolite ZMQ-1 and a preparation method and application thereof. A silicate zeolite precursor has an anhydrous chemical composition of $SiO_2 \cdot 1/xXO_{1.5} \cdot mMO_{0.5} \cdot qQ$, wherein X is a framework trivalent element, a molar ratio of Si/X is $x \geq 5$, M is a framework balanced cation, a molar ratio of M/Si is $0 \leq m \leq 1$, Q is a bis-quaternary ammonium or bis-quaternary phosphonium organic structure directing agent, and a molar ratio of Q/Si is $q \geq 0.01$. A novel silicate zeolite has an anhydrous chemical composition of $SiO_2 \cdot 1/xXO_{1.5} \cdot mMO_{0.5}$, wherein X is the framework trivalent element, the molar ratio of Si/X is $x \geq 5$, M is a framework balanced cation, and the molar ratio of M/Si is $0 \leq m \leq 1$. In view of the novel topological structure, unique pore system, and high thermal and hydrothermal stability, the zeolite of the present invention can be further applied in an energy storage, sensing, loading, adsorption, separation or catalytic process, and may exhibit unique properties. Fig. 1



NOVEL SILICATE ZEOLITE ZMQ-1 AND APPLICATION THEREOF

Technical Field

The present invention relates to zeolites, and in particular to a novel silicate zeolite ZMQ-1 and a preparation method and application thereof.

Background

Zeolites are a kind of very important inorganic microporous materials. For half a century, zeolites have been widely used in petroleum processing, petrochemical industry and environmental chemical industry as catalysis, adsorption&separation and ion exchange materials. According to T-atom numbers of the pore ring, zeolite material can be divided into small pore, medium pore, large pore and extraextra-large pore zeolites, corresponding to window ring numbers of 8-membered rings and less, 10-membered rings and less, 12-membered rings and less, and 13-membered rings and more, respectively. Up to now, 260 zeolite structures have been recognized and accepted by International Zeolite Association (official website https://asia.iza-structure.org/IZA-SC/ftc_table.php), with each structure represented by three capital English letters. However, less than 20 types of zeolites are actually used in industrial applications, such as zeolites A, X, Y, mordenite, ZSM-5, ZSM-11, MCM-22, L, β, erionite, RHO, CHA, AEL, TS-1, SAPO-34, SAPO-11 and SAPO-31. In view of this, developing zeolite materials with new structures and properties has always been an important direction in the field, which will effectively expand the application range of zeolites to meet the growing needs of relevant industrial processes.

At present, a trial and error method or an empirical method are still the main methods for synthesis and exploration of novel zeolites; such methods have the disadvantages of being time and labor consuming and being not easy to obtain desired products; in addition, a large amount of waste will be generated concomitantly, so the methods are non-green zeolite synthesis methods. In 1960s, researchers introduced organic species into a zeolite synthesis process, and a number of high-silica zeolites with new structures were synthesized. For example, zeolites β, ZSM-5 and ZSM-11 were successfully synthesized by using tetraethyl ammonium, tetrapropylammonium and tetrabutylammonium cations as organic additives, respectively. The organic species are often used in conjunction with alkali metal cations; at the same time, because of a filling effect thereof on zeolite pores, the organic species are initially called "template agents"; however, with the further study of zeolite synthesis mechanism, it is found that many organic species are not matched strictly with zeolite pore sizes or structures, and in many cases, a single organic species can be synthesized to obtain multiple zeolite structures. Therefore, the organic species are named as "Organic Structure Directing Agents (OSDAs)" by the researchers, which is also a generally accepted definition in the field of zeolite synthesis at present, and an effect produced by the organic species is called "structure directing effect" (M. E. Davis and R. F. Lobo, Chem. Mater., 1992, 4, 756-768). The proposal of the "structure directing effect" provides a methodological guidance for zeolite synthesis, and makes the synthesis process more purposeful.

According to different types of organic cations, organic structure directing agents can be divided into quaternary ammonium cations, imidazolyl cations, quaternary phosphonium cations, sulfonium cations, proton sponges, metal complexes and neutral amines, among which quaternary ammonium and quaternary phosphonium cations are most mature and most widely used. Compared with quaternary ammonium cations, quaternary phosphonium cations have the advantages that: (1) phosphorus atoms are larger than nitrogen atoms, and are therefore easier to connect with larger substituent groups; (2) quaternary phosphonium cations have a higher thermal stability and can exist stably in a 190℃ alkali system, whereas most quaternary ammonium cations will decompose and lose the structure directing effect. Therefore, in recent years, phosphorous-containing organic structure directing agents have been widely used and developed, and several zeolites with new structures (CN114538466A and CN115611293A) have been synthesized, showing a great potential in the development of zeolites with new structures, which is worth of further excavation and exploration.

In addition, the introduction of other inorganic species may also provide additional structure directing effect, and the inorganic species can jointly direct the formation of zeolites with specific structures; the zeolites are microporous materials formed by TO_4 tetrahedrons through alternate connection, and other elements, such as B, Al, Be, Mg, Ga, Ge, Zn and Co, can enter a zeolite framework by isomorphous replacement. The introduction of heteroatoms can simultaneously produce some unique chemical and physical properties, such as catalytic property, selective adsorption or magnetic property. In addition, the heteroatoms can significantly affect a formation process of a zeolite. For example, Ge, B, Ga, Zn and Be are conducive to the formation of specific structural units (such as 4-membered and 3-membered rings, double-4-membered and double-3-membered rings, and spiral 5-membered rings) because the appropriate T-O bond and T-O-T bond angles thereof can play a role in stabilizing the structural units.

In recent years, dozens of zeolites with new structures have been synthesized based on a strategy of designing organic structure directing agents and introducing heteroatoms. A number of novel zeolites named as ITQ-n have been developed by Avelino Corma's research group in Spain (Moliner et al, Angew. Chem. Int. Ed. 2013, 52, 13880-13889 and Li et al, Chem. Soc. Rev., 2015, 44, 7112-7127), and the pore size of the zeolites covers the range from small to extra-large pores, which greatly expands the structural types of zeolites. However, most of the novel zeolites contain germanium, and after the organic structure directing agents of the novel zeolites are removed and the novel zeolites are exposed to air, the framework germanium will undergo hydrolysis, eventually leading to the collapse of the framework, thus greatly limiting the practical application of the novel zeolites. It is found by Sukbong Hong's research team in South Korea that the weak interaction of organic cations and inorganic cations with aluminum species can also lead to the formation of zeolites with specific structures. The types of the organic cations are fixed and two inorganic cations (Na^+ and Cs^+) are introduced by Sukbong Hong's research team to form a charge density mismatch of a synthetic system, and a structurally stable large-pore SBT aluminosilicate zeolite is synthesized, which solves the problem of structural collapse after the organic structure directing agents in the zeolites are removed (Lee et al., 2021, Science 373, 104–107 and Lee et al., J. Am. Chem. Soc. 2022, 144, 18700−18709). However, relatively more modulation factors are involved in the method, and a target zeolite can only be obtained within a narrow gel ratio range, so the reproducibility of synthesis results will be affected to some extent. In addition, the zeolite synthesized has a relatively low silica-alumina ratio, that is, the content of aluminum is high, so the hydrothermal stability of the zeolite is reduced.

To sum up, although zeolites or zeolitic materials with various topological structures have been obtained, in the face of the increasing new demands during adsorption&separation and catalytic conversion, it is urgent to develop a novel zeolite material with diversified topological and porous structures and having thermal and hydrothermal stability in view of the problems of structural instability and type restriction of the existing zeolites.

Summary

The purpose of the present invention is to provide a novel silicate zeolite (ZMQ-1) and a preparation method and application thereof.

A silicate zeolite precursor has an anhydrous chemical composition of $SiO_2 \cdot 1/xXO_{1.5} \cdot mMO_{0.5} \cdot qQ$, wherein X is a framework trivalent element, a molar ratio of Si/X is $x \geqslant 5$, M is a framework balanced cation, a molar ratio of M/Si is $0 \leqslant m \leqslant 1$, Q is a bis-quaternary ammonium or bis-quaternary phosphonium organic structure directing agent, and a molar ratio of Q/Si is $q \geqslant 0.01$.

Powder X-ray diffraction characteristics of the silicate zeolite precursor are as follows:

Diffraction angle 2θ (°)	Interplanar spacing (Å)	Relative intensity
3.27	27.00	mw
4.78	18.47	s
5.12	17.25	vs
5.42	16.29	m
6.58	13.42	m
7.92	11.15	w
8.37	10.55	w
8.75	10.10	w
9.34	9.46	w
9.58	9.23	w
9.96	8.87	w
10.15	8.71	mw
10.35	8.54	mw
10.55	8.38	w
10.85	8.15	m
11.36	7.78	w
11.74	7.53	w
12.84	6.89	w
13.56	6.52	w
13.97	6.33	w
14.41	6.14	w
14.66	6.04	w
15.10	5.86	w
15.53	5.70	w
15.94	5.56	w
16.53	5.36	w
16.78	5.28	w
17.17	5.16	w
17.83	4.97	w
18.20	4.87	w
18.48	4.80	w
18.81	4.71	mw
19.33	4.59	mw
19.53	4.54	mw
20.01	4.43	w
21.20	4.19	w
21.70	4.09	mw
21.88	4.06	w
22.60	3.93	w
22.98	3.87	vs
23.61	3.77	w
23.96	3.71	mw
24.42	3.64	w
24.72	3.60	w
25.00	3.56	w
25.18	3.53	w
25.63	3.47	w
25.99	3.43	w

26.66	**3.34**	**w**
27.67	**3.22**	**w**
28.01	**3.18**	**w**
28.55	**3.12**	**w**
29.12	**3.06**	**w**
29.82	**2.99**	**w**
30.18	**2.96**	**w**

For the data in the table of the present application, the relative intensity of a diffraction peak is defined in the following table.

$100I/I_0$	**Relative intensity**
<10	w (Weak peak)
10-20	mw (Medium weak peak)
20-40	m (Medium peak)
40-70	s (Strong peak)
>70	vs (Very strong peak)

A novel silicate zeolite has an anhydrous chemical composition of $SiO_2 \cdot 1/xXO_{1.5} \cdot mMO_{0.5}$, wherein X is the framework trivalent element, the molar ratio of Si/X is $x \geqslant 5$, M is a framework balanced cation, and the molar ratio of M/Si is $0 \leqslant m \leqslant 1$.

The novel silicate zeolite contains a three-dimensional pore system composed of 28×10×10-membered rings, wherein the 28-membered ring has a size of 23.11×12.39 Å, and the 10-membered ring has a size of 5.88 Å.

The above silicate zeolite is an extra-large pore zeolite with a novel topological structure, and the largest pore thereof is composed of the 28-membered ring with a size of 23.11 Å, which exceeds the micropore range (less than 2 nm) defined by International Union of Pure and Applied Chemistry (IUPAC). More importantly, the zeolite ZMQ-1 can be modulated in a composition range of low silicon, high silicon or pure silicon.

Powder X-ray diffraction characteristics of the novel silicate zeolite are shown in the following table,

Diffraction angle 2θ (°)	Interplanar spacing (Å)	Relative intensity
3.15	**28.00**	**m**
4.89	**18.04**	**vs**
6.42	**13.76**	**mw**
6.64	**13.30**	**w**
7.75	**11.40**	**w**
7.94	**11.13**	**w**
8.65	**10.21**	**w**
9.33	**9.48**	**w**
9.66	**9.15**	**w**
9.93	**8.90**	**w**
10.27	**8.60**	**w**
10.75	**8.22**	**w**
12.15	**7.28**	**w**
13.54	**6.53**	**w**
14.12	**6.27**	**w**
14.66	**6.04**	**w**
14.94	**5.92**	**w**
15.55	**5.69**	**w**
15.88	**5.58**	**w**
16.05	**5.52**	**w**
17.42	**5.09**	**w**
17.83	**4.97**	**w**
18.10	**4.90**	**w**
18.75	**4.73**	**w**
19.05	**4.65**	**w**
19.76	**4.49**	**w**
20.12	**4.41**	**w**
20.79	**4.27**	**w**
21.42	**4.15**	**w**
22.80	**3.90**	**mw**
23.57	**3.77**	**w**
23.96	**3.71**	**w**
24.76	**3.59**	**w**
25.15	**3.54**	**w**
26.00	**3.42**	**w**
26.25	**3.39**	**w**
26.70	**3.34**	**w**
27.31	**3.26**	**w**
27.97	**3.19**	**w**
28.53	**3.13**	**w**
28.99	**3.08**	**w**
29.77	**3.00**	**w**
30.89	**2.89**	**w**

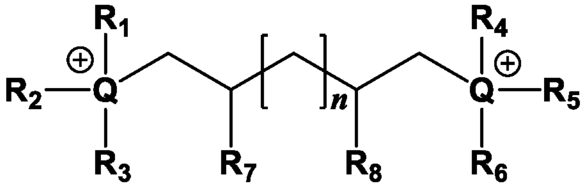
The novel silicate zeolite is obtained after the novel silicate zeolite precursor is calcined.

The framework trivalent element X is selected from at least one of boron, aluminum, gallium, indium, iron and chromium; the framework balanced cation M is selected from at least one of hydrogen ion, ammonium ion, lithium ion, sodium ion, potassium ion, rubidium ion and cesium ion; silicon in the zeolite contains a non-silicon tetravalent framework element (Y) with an amount of greater than or equal to 0 and less than or equal to 10w% of the mass of the silicon, wherein Y is selected from at least one of germanium, tin, titanium, zirconium and hafnium.

A preparation method of the novel silicate zeolite is that:

A framework balanced cation compound, a silicon source, a framework trivalent element X source, an organic structure directing agent and water are mixed to obtain a synthetic gel; then the synthetic gel is aged and hydrothermally crystallized, and the crystallized product is calcined to remove the organic structure directing agent; among which, the synthetic gel after aging has a molar ratio of SiO_2:mM_2O:xX_2O_3:$qQ(OH)_2$:hH_2O, wherein M is the framework balanced cation, X is the framework trivalent element, Q is a cationic group of the organic structure directing agent, and value ranges of m, x, q and h are: m=0-1, x=0-0.5, q=0.1-1, and h=3-100;

The organic structure directing agent has the following configuration:



wherein Q can be identical or different and selected from nitrogen or phosphorus, R_1, R_2, R_3, R_4, R_5 and R_6 can be identical or different and selected from cyclohexyl or phenyl, R7 and R8 can be identical or different and selected from H or methyl, and n is 0-8, preferably 3-5.

Further, the preparation method is that:

(1) The framework balanced cation compound, the silicon source, the framework trivalent element X source, the organic structure directing agent and deionized water are weighed according to stoichiometry, respectively, added to

an open container in an order of first liquid phase and then solid phase, and mixed well under magnetic stirring to obtain an initial synthetic gel.

(2) The synthetic gel is stirred at room temperature to age for 12-24 hours, and then placed in an oven at 60-100℃ to age for 2-6 hours and remove excess solvent and water at the same time. The synthetic gel finally obtained has a molar ratio of SiO_2:$m$$M_2O$:$x$$X_2O_3$:$qQ(OH)_2$:$h$$H_2O$, wherein M is the framework balanced cation, X is the framework trivalent element, Q is a cationic group of the organic structure directing agent, and value ranges of m, x, q and h are: m=0-1, x=0-0.5, q=0.1-1, and h=3-100. The value ranges are preferably: m=0-0.15, x=0-0.1, q=0.1-0.3, and h=5-50.

(3) The synthetic gel is transferred into a Teflon liner, sealed in a stainless steel autoclave, and finally placed in an oven for crystallization. Temperature for crystallization is 50-250℃, preferably 100-200℃; time for crystallization is 1-30 days, preferably 5-15 days; mode for crystallization is static or/and rotational crystallization.

(4) After being washed and dried, crystallization reaction products are calcined in air or/and inert atmosphere at 500-1000℃ for 3-24 hours to remove the organic structure directing agent and obtain an aluminosilicate zeolite. The temperature for calcination is preferably 550-750℃, and the time for calcination is preferably 6-12 hours.

In addition, a crystal seed can be further added in the gel synthesis process of the zeolite synthesis process, and the crystal seed is the zeolite synthesized by the above step (3), which is in the form of a zeolite before or after calcination.

The framework trivalent element X source in the synthesis process is selected from at least one of a boron source, an aluminum source, a gallium source, an indium source, an iron source and a chromium source; the boron source is selected from at least one of boric acid, sodium tetraborate, amorphous boron oxide, potassium borate, sodium metaborate, ammonium tetraborate and organoborate; the aluminum source is selected from at least one of aluminum sulfate, sodium aluminate, aluminum nitrate, aluminum chloride, pseudo boehmite, aluminum oxide, aluminum hydroxide, aluminum silicate zeolite, aluminum carbonate, elemental aluminum, aluminum isopropoxide and aluminum acetate; the gallium source, the indium source and the chromium source can be selected from at least one of gallium oxide, gallium nitrate, indium oxide, indium nitrate, chromium chloride and chromium nitrate in turn. the iron source is selected from at least one of ferric sulfate, ferric nitrate, ferric halide, ferrocene and ferric citrate; the framework balanced cation compound is selected from at least one of hydroxides (such as lithium hydroxide and sodium hydroxide), halides (such as lithium chloride and sodium chloride); the silicon source is selected from one or more of tetraethyl orthosilicate, silica sol, fumed silica, silicic acid and sodium silicate; at the same time, a non-silicon tetravalent framework element (Y) source can also be added to the silicon source, with an addition amount of greater than or equal to 0 and less than or equal to 10w% of the mass of the silicon source, wherein Y is selected from at least one of germanium, tin, titanium, zirconium and hafnium.

A zeolite composition contains the silicate zeolite precursor and/or the silicate zeolite.

An application of the novel silicate zeolite comprises an application of the silicate zeolite or the zeolite composition as an energy storage material, a chemical sensing material, a carrier, an adsorbent, a separating agent or a catalyst.

Further, the application of the silicate zeolite or the zeolite composition as an energy storage material, a chemical sensing material, a carrier, an adsorption&separation agent or a catalyst is provided.

In the application as an energy storage material, the silicate zeolite or the zeolite composition can be used as a storage material for hydrogen and hydrogen mixed media (such as hydrogen-methane, hydrogen-ethane and hydrogen-propane systems).

In the application as a chemical sensing material, the silicate zeolite or the zeolite composition can be used to monitor light hydrocarbons, alkaline molecules, carbon dioxide, sulfide gases, humidity, etc.

In the application as a carrier, the silicate zeolite or the zeolite composition can be used to carry metals, non-metals and biological enzymes for preparing catalysts, and to carry drugs for imaging and treatment.

In the application as an adsorption&separation agent, the silicate zeolite or the zeolite composition can be used as a desiccant to remove water, adsorb macromolecules of volatile organic compounds (VOCs), treat wastewater containing dye macromolecules and separate micromolecules of carbohydrate and proteins. More specifically, in view of regular extra-large micropores thereof, the silicate zeolite or the zeolite composition can be used as a filler of a high-performance liquid chromatographic column.

In the application as a catalyst, the silicate zeolite or the zeolite composition can be used to produce gasoline, diesel and other chemicals by vacuum gas oil (VGO) hydrogenation or direct catalytic cracking, produce oil, olefins and other chemicals by vacuum residue hydrogenation or direct catalytic cracking, and produce high-value-added chemicals by conversion of other fossil energy and renewable energy, and used in isomerization, disproportionation, Friedel-Crafts alkylation and other reactions with the involvement of organic macromolecular substrates. More specifically, in view of the unique 10-membered rings and medium micropores thereof, the application of the zeolite in the VGO and vacuum residue cracking processes may be beneficial to the production of light olefins, especially ethylene and propylene, which is of great significance for the upgraded utilization of fossil energy in China.

The present invention has the following advantages:

In the present invention, a novel aluminosilicate zeolite with a three-dimensional open framework structure is synthesized by designing bis-tricyclohexyl/phenyl quaternary phosphonium cations as an organic structure directing agent; the zeolite has a unique powder X-ray diffraction peak; in addition, the zeolite has a three-dimensional pore system composed of one type of extra-large micropores and two types of medium micropores.

In view of the novel topological structure, unique pore system, and high thermal and hydrothermal stability, the zeolite of the present invention can be further applied in an adsorption, separation or catalytic process with the involvement of macromolecules, and may exhibit unique properties.

Description of Drawings

Fig. 1 is a powder X-ray diffraction spectrum of a freshly synthesized zeolite precursor and a zeolite ZMQ-1 calcined at 600℃ for 6 hours and at 800℃ for 1 hour provided in embodiments 3-5 of the present invention.

Fig. 2 is a powder X-ray diffraction spectrum of ZMQ-1 for the zeolite calcined at 600℃ and treated with 50% water vapor at 600℃, 700℃ and 800℃ for 3 hours provided in embodiments 3-5 of the present invention.

Fig. 3 shows an argon adsorption isotherm and a pore size distribution curve of a zeolite ZMQ-1 calcined at 600℃ for 6 hours provided in embodiment 3 of the present invention.

Fig. 4 a scanning electron microscope (SEM) photograph of a zeolite ZMQ-1 calcined at 600℃ for 6 hours provided in embodiment 3 of the present invention.

Fig. 5 a transmission electron microscope (TEM) photograph of a zeolite ZMQ-1 calcined at 600℃ for 6 hours provided in embodiment 3 of the present invention.

Fig. 6 a topological structure diagram of a zeolite ZMQ-1 calcined at 600℃ for 6 hours provided in embodiment 3 of the present invention.

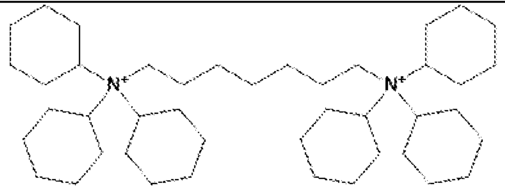
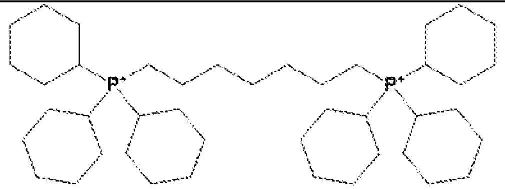
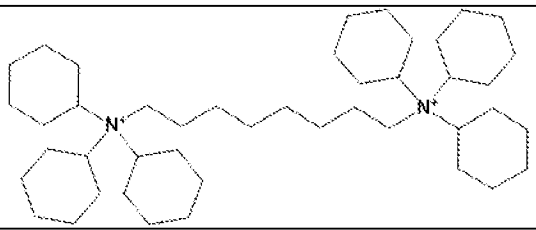
Detailed Description

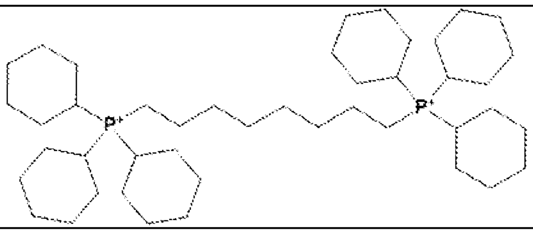
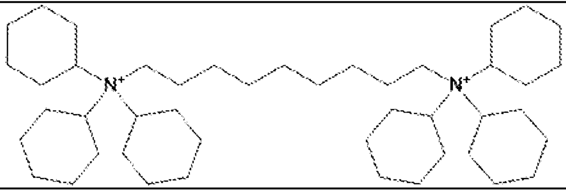
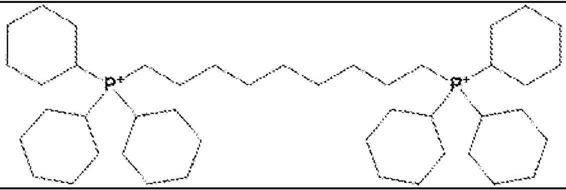
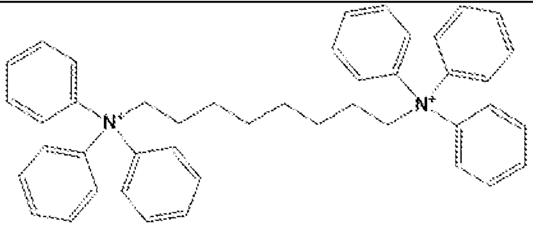
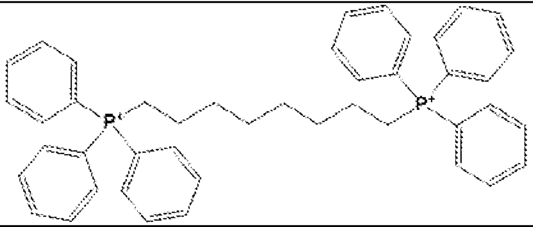
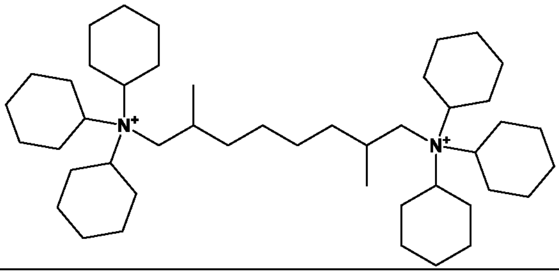
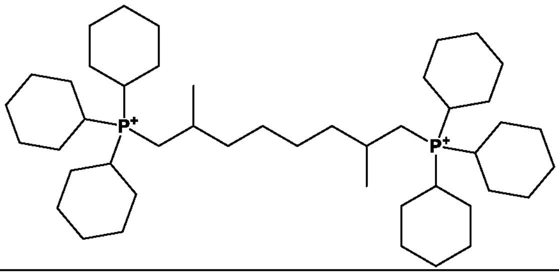
Detailed description of the present invention is further illustrated below in combination with examples. It shall be noted that the detailed description described herein is only used to illustrate and explain the present invention, not limited to the present invention.

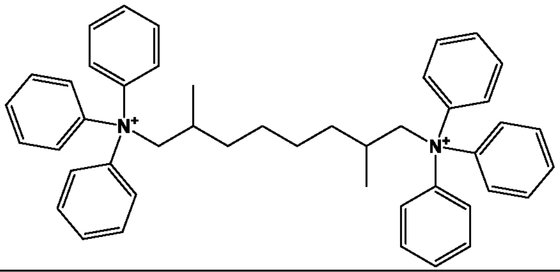
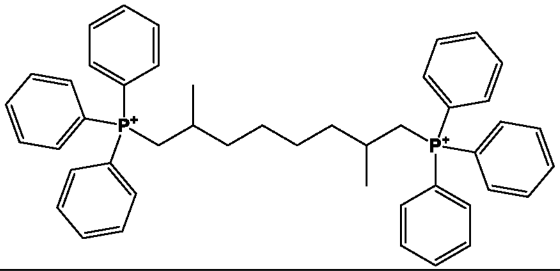
In embodiments 3-10, powder X-ray diffraction data analysis of a sample is performed by an X-ray diffractometer from Rigaku Corporation, with a model number of LabView, an X-ray source of CuKα and a wavelength of 1.5418 Å. Argon desorption analysis of the sample is performed by an adsorption instrument from Quantachrome Instruments, with a model number of Autosorb iQ. Elemental analysis of the sample is performed by an inductively coupled plasma emission spectrometer from Agilent Technologies Inc. (formerly Varian), with a model number of ICP-730ES. Scanning electron microscope (SEM) and transmission electron microscope (TEM) photographs of the sample are obtained by a cold field emission scanning electron microscope (S-4800) from Hitachi Limited and a high-resolution transmission electron microscope (JEM-F200) from JEOL Ltd., respectively.

In the following embodiments, the zeolite is prepared by hydrothermal crystallization, the organic structure directing agent used is bis-quaternary ammonium/phosphonium cations with different lengths, and the organic structure directing agent is selected from any one or more of the agents in the following table.

Table 1

No.	**Structure**
1	
2	
3	

4	
5	
6	
7	
8	
9	
10	

11	
12	

Among the above organic structure directing agents, any one or more of the organic structure directing agents 3, 4, 9 and 10 are preferred.

Embodiment 1: synthesis of organic structure directing agent

The synthesis of organic structure directing agent 4 is taken as an example. 36.51 g of tricyclohexylphosphane is weighed and put into a 500 mL three-necked round-bottomed flask, added with 200 mL of chloroform, dissolved under magnetic stirring, and transferred into an ice bath. 13.88 g of 1,8-dibromooctane is slowly dripped into the flask under magnetic stirring, and stirring is continued for the mixture solution obtained for 1 hour. Subsequently, the mixture solution is refluxed and heated at 75℃ for 3 days. After the mixture is cooled to room temperature, an excessive amount of ethyl acetate is added to make a product precipitated, filtered by suction, washed by ethyl acetate, and treated by rotary evaporation to remove residual solvent. A final product of 39.16 g is obtained, with a yield rate of 94%. The final product is determined to be a target compound by liquid-phase NMR and CHN elemental analysis.

Powder of the organic structure directing agent is dissolved in 200 mL of deionized water in a plastic beaker, and then poured into pre-activated strongly basic anion exchange resin ZXUR-90 from Zhengzhou Xidian Electric Co., Ltd.; magnetic stirring is performed for 12 hours. A concentrated organic structure directing agent solution is obtained after sucking filtration, washing, filtrate recovering and rotary evaporation. A small amount of solution is taken and diluted to 50 mL with water, 1.00 g of 0.1 mol/L hydrochloric acid standard solution is taken and added with phenol as an indicator to titrate the diluted organic structure directing agent solution, and a final exchange degree is determined to be 95%.

Embodiment 2: synthesis of organic structure directing agent

The synthesis of organic structure directing agent 10 is taken as an example. 36.51 g of tricyclohexylphosphane is weighed and put into a 500 mL three-necked round-bottomed flask, added with 200 mL of chloroform, dissolved under magnetic stirring, and transferred into an ice bath. 15.31 g of 1,8-dibromo-2,7-dimethyl octane is slowly dripped into the flask under magnetic stirring, and stirring is continued for the mixture solution obtained for 1 hour. Subsequently, the mixture solution is refluxed and heated at 75℃ for 3 days. After the mixture is cooled to room temperature, an excessive amount of ethyl acetate is added to make a product precipitated, filtered by suction, washed by ethyl acetate, and treated by rotary evaporation to remove residual solvent. A final product of 40.99 g is obtained, with a yield rate of 95%. The final product is confirmed to be a target compound by liquid-phase NMR and CHN elemental analysis.

Powder of the organic structure directing agent is dissolved in 200 mL of deionized water in a plastic beaker, and then poured into pre-activated strongly basic anion exchange resin ZXUR-90 from Zhengzhou Xidian Electric Co., Ltd.; magnetic stirring is performed for 12 hours. A concentrated organic structure directing agent solution is obtained after sucking filtration, washing, filtrate recovering and rotary evaporation. A small amount of solution is taken and diluted to 50 mL with water, 1.00 g of 0.1 mol/L hydrochloric acid standard solution is taken and added with phenol as an indicator to titrate the diluted organic structure directing agent solution, and a final exchange degree is determined to be 97%.

At the same time, the other organic structure directing agents described in

Table 1 can be obtained by replacing the phosphonium cations with nitrogen cations or substituting the substituent groups or methylene chain lengths attached to the phosphonium/nitrogen cations according to the preparation process recorded in embodiment 1 or 2 above.

Embodiment 3: synthesis of zeolite

A gel is prepared according to a molar ratio of SiO_2:0.02Al_2O_3:0.25$Q(OH)_2$:10H_2O, and the specific steps are as follows: weighing 6 mmol of the organic structure directing agent solution described in embodiment 1, adding 0.21 g of aluminum isopropoxide, performing magnetic stirring for 1 hour, adding 5.21 g of tetraethyl orthosilicate subsequently, stirring obtained mixture at room temperature for 12 hours, placing obtained transparent gel in a vacuum oven, and heating at 100℃ for 3 hours to remove solvent and excess water. Transferring a final synthetic gel to a 25 mL stainless steel autoclave with a Teflon liner, and crystallizing the final synthetic gel at 190℃ for 10 days. Washing a product by sucking filtration with 200 mL of water, 200 mL of ethanol and 100 mL of acetone, and drying overnight to obtain a freshly synthesized zeolite precursor. Subsequently, calcination at 600℃ for 6 hours in a muffle furnace in a flowing air atmosphere to remove the organic structure guide agent. The phase of the product is determined to be ZMQ-1 by powder X-ray diffraction, and an atomic ratio of Si to Al is determined to be 15.9 by ICP-OES elemental analysis.

Embodiment 4: calcination of zeolite

The freshly synthesized zeolite precursor prepared in embodiment 3 is placed in the muffle furnace and calcined at 800℃ for 1 hour in a flowing air atmosphere to remove the organic structure guide agent, and the phase of the product is determined to be ZMQ-1 by powder X-ray diffraction, as shown in Fig. 1.

Embodiment 5: hydrothermal treatment of zeolite

A sample calcined at 600℃ in embodiment 3 is divided into three parts and placed in a fixed-bed reactor, respectively, deionized water is fed into the reactor through a peristaltic pump in a nitrogen atmosphere, a relative humidity of 50% is kept, and the sample is treated at 600℃, 700℃, and 800℃ for 3 hours, respectively. After being naturally cooled to room temperature, the treated zeolite is taken out, and the phase of the treated zeolite is determined to be ZMQ-1 by powder X-ray diffraction, as shown in Fig. 2.

Embodiment 6: synthesis of zeolite

A gel is prepared according to a molar ratio of SiO_2:0.01Al_2O_3:0.25Q$(OH)_2$:10H_2O, and the specific steps are as follows: weighing 6 mmol of the organic structure directing agent solution described in embodiment 1, adding 0.10 g of aluminum isopropoxide, performing magnetic stirring for 1 hour, adding 5.21 g of tetraethyl orthosilicate subsequently, stirring obtained mixture at room temperature for 12 hours, placing obtained transparent gel in a vacuum oven, and heating at 100℃ for 3 hours to remove solvent and excess water. Transferring a final synthetic gel to a 25 mL stainless steel autoclave with a Teflon liner, and crystallizing the final synthetic gel at 190℃ for 10 days. Washing a product by sucking filtration with 200 mL of water, 200 mL of ethanol and 100 mL of acetone, and drying overnight to obtain a freshly synthesized zeolite precursor. Subsequently, calcination at 600℃ for 6 hours in a muffle furnace in a flowing air atmosphere to remove the organic structure guide agent. The phase of the product is determined to be ZMQ-1 by powder X-ray diffraction, and an atomic ratio of Si to Al is determined to be 35.2 by ICP-OES elemental analysis.

Embodiment 7: synthesis of zeolite

A gel is prepared according to a molar ratio of SiO_2:0.005Al_2O_3:0.25Q$(OH)_2$:10H_2O, and the specific steps are as follows: weighing 6 mmol of the organic structure directing agent solution described in embodiment 1, adding 0.05 g of aluminum isopropoxide, performing magnetic stirring for 1 hour, adding 5.21 g of tetraethyl orthosilicate subsequently, stirring obtained mixture at room temperature for 12 hours, placing obtained transparent gel in a vacuum oven, and heating at 100℃ for 3 hours to remove solvent and excess water. Transferring a final synthetic gel to a 25 mL stainless steel autoclave with a Teflon liner, and crystallizing the final synthetic gel at 190℃ for 10 days. Washing a product by sucking filtration with 200 mL of water, 200 mL of ethanol and 100 mL of acetone, and drying overnight to obtain a freshly synthesized zeolite precursor. Subsequently, calcination at 600℃ for 6 hours in a muffle furnace in a flowing air atmosphere to remove the organic structure guide agent. The phase of the product is determined to be ZMQ-1 by powder X-ray diffraction, and an atomic ratio of Si to Al is determined to be 67.5 by ICP-OES elemental analysis.

Embodiment 8: synthesis of zeolite

A gel is prepared according to a molar ratio of SiO_2:0.02Al_2O_3:0.25Q$(OH)_2$:10H_2O, and the specific steps are as follows: weighing 6 mmol of the organic structure directing agent solution described in embodiment 2, adding 0.21 g of aluminum isopropoxide, performing magnetic stirring for 1 hour, adding 5.21 g of tetraethyl orthosilicate subsequently, stirring obtained mixture at room temperature for 12 hours, placing obtained transparent gel in a vacuum oven, and heating at 100℃ for 3 hours to remove solvent and excess water. Transferring a final synthetic gel to a 25 mL stainless steel autoclave with a Teflon liner, and crystallizing the final synthetic gel at 190℃ for 15 days. Washing a product by sucking filtration with 200 mL of water, 200 mL of ethanol and 100 mL of acetone, and drying overnight to obtain a freshly synthesized zeolite precursor. Subsequently, calcination at 600℃ for 6 hours in a muffle furnace in a flowing air atmosphere to remove the organic structure guide agent. The phase of the product is determined to be ZMQ-1 by powder X-ray diffraction, and an atomic ratio of Si to Al is determined to be 15.3 by ICP-OES elemental analysis.

Embodiment 9: synthesis of zeolite

A gel is prepared according to a molar ratio of SiO_2:0.02Na_2O:0.02Al_2O_3:0.25Q$(OH)_2$:10H_2O, and the specific steps are as follows: weighing an appropriate amount of the organic structure directing agent solution described in embodiment 1, adding 0.04 g of sodium hydroxide, stirring until the sodium hydroxide is dissolved, adding 0.21 g of aluminum isopropoxide, performing magnetic stirring for 1 hour, adding 5.21 g of tetraethyl orthosilicate subsequently, stirring obtained mixture at room temperature for 12 hours, placing obtained transparent gel in a vacuum oven, and heating at 100℃ for 3 hours to remove solvent and excess water. Transferring a final synthetic gel to a 25 mL stainless steel autoclave with a Teflon liner, and crystallizing the final synthetic gel at 190℃ for 5 days. Washing a product by sucking filtration with 200 mL of water, 200 mL of ethanol and 100 mL of acetone, and drying overnight to obtain a freshly synthesized zeolite. Subsequently, calcination at 600℃ for 6 hours in a muffle furnace in a flowing air atmosphere to remove the organic structure guide agent. The phase of the product is determined to be ZMQ-1 by powder X-ray diffraction, and an atomic ratio of Si to Al is determined to be 15.9 by ICP-OES elemental analysis.

Embodiment 10: synthesis of zeolite

A gel is prepared according to a molar ratio of SiO_2:$0.02Na_2O$:$0.02Al_2O_3$:$0.25Q(OH)_2$:$10H_2O$, and the specific steps are as follows: weighing an appropriate amount of the organic structure directing agent solution described in embodiment 2, adding 0.04 g of sodium hydroxide, stirring until the sodium hydroxide is dissolved, adding 0.21 g of aluminum isopropoxide, performing magnetic stirring for 1 hour, adding 5.21 g of tetraethyl orthosilicate subsequently, stirring obtained mixture at room temperature for 12 hours, placing obtained transparent gel in a vacuum oven, and heating at 100℃ for 3 hours to remove solvent and excess water. Transferring a final synthetic gel to a 25 mL stainless steel autoclave with a Teflon liner, and crystallizing the final synthetic gel at 190℃ for 5 days. Washing a product by sucking filtration with 200 mL of water, 200 mL of ethanol and 100 mL of acetone, and drying overnight to obtain a freshly synthesized zeolite. Subsequently, calcination at 600℃ for 6 hours in a muffle furnace in a flowing air atmosphere to remove the organic structure guide agent. The phase of the product is determined to be ZMQ-1 by powder X-ray diffraction, and an atomic ratio of Si to Al is determined to be 16.5 by ICP-OES elemental analysis.

In the above embodiments for obtaining different zeolites ZMQ-1, three-dimensional diffraction data is obtained by continuous rotation electron diffraction (cRED), and structural analysis is performed. Results show that the structures of the zeolites ZMQ-1 have orthogonal symmetry and belong to a *Cmmm* space group, and cell parameters obtained are:

a=19.58800, b=56.51200, c=21.05700, V=23309.193497 Å^3。

In addition, a powder X-ray diffraction spectrum of the zeolites synthesized in embodiments 3-5 is shown in Fig. 1, both the freshly synthesized and the calcined products show clear and unique characteristic diffraction peaks, and the phases of the sample calcined at 600℃ and 800℃ are determined to be zeolites ZMQ-1 according to the diffraction peaks, proving that the zeolites have a high thermal stability. Fig. 2 shows X-ray diffraction peaks of the products after 50% water vapor treatment, which are all zeolites ZMQ-1, proving that the aluminosilicate zeolite has a high hydrothermal stability. Argon physical adsorption data of the zeolites after calcination proves that the zeolites have high micropore adsorption capacities and contain medium and extra-large micropore structures, as shown in Fig. 3. Fig. 4 and Fig. 5 are SEM and TEM photographs of the sample calcined at 600℃ in embodiment 3, respectively, with grains having a morphology of quadrangular prisms. Fig. 6 shows a topological structure of the zeolite calcined at 600℃ in embodiment 3, from which pores with 28-membered rings along a c-axis and pores with 10-membered rings along an a-axis and a b-axis can be observed.

Claims

1. A silicate zeolite precursor, having an anhydrous chemical composition of $SiO_2 \cdot 1/x XO_{1.5} \cdot m MO_{0.5} \cdot q Q$, wherein X is a framework trivalent element, a molar ratio of Si/X is $x \geqslant 5$, M is a framework balanced cation, a molar ratio of M/Si is $0 \leqslant m \leqslant 1$, Q is a bis-quaternary ammonium or bis-quaternary phosphonium organic structure directing agent, and a molar ratio of Q/Si is $q \geqslant 0.01$.

2. The silicate zeolite precursor according to claim 1, wherein powder X-ray diffraction characteristics of the silicate zeolite precursor are as follows:

Diffraction angle 2θ (°)	Interplanar spacing (Å)	Relative intensity
3.27	27.00	mw
4.78	18.47	s
5.12	17.25	vs
5.42	16.29	m
6.58	13.42	m
7.92	11.15	w
8.37	10.55	w
8.75	10.10	w
9.34	9.46	w
9.58	9.23	w
9.96	8.87	w
10.15	8.71	mw
10.35	8.54	mw
10.55	8.38	w
10.85	8.15	m
11.36	7.78	w
11.74	7.53	w
12.84	6.89	w
13.56	6.52	w
13.97	6.33	w
14.41	6.14	w
14.66	6.04	w
15.10	5.86	w
15.53	5.70	w
15.94	5.56	w
16.53	5.36	w
16.78	5.28	w
17.17	5.16	w
17.83	4.97	w
18.20	4.87	w
18.48	4.80	w
18.81	4.71	mw
19.33	4.59	mw

19.53	**4.54**	**mw**
20.01	**4.43**	**w**
21.20	**4.19**	**w**
21.70	**4.09**	**mw**
21.88	**4.06**	**w**
22.60	**3.93**	**w**
22.98	**3.87**	**vs**
23.61	**3.77**	**w**
23.96	**3.71**	**mw**
24.42	**3.64**	**w**
24.72	**3.60**	**w**
25.00	**3.56**	**w**
25.18	**3.53**	**w**
25.63	**3.47**	**w**
25.99	**3.43**	**w**
26.66	**3.34**	**w**
27.67	**3.22**	**w**
28.01	**3.18**	**w**
28.55	**3.12**	**w**
29.12	**3.06**	**w**
29.82	**2.99**	**w**
30.18	**2.96**	**w**

3. A novel silicate zeolite, having an anhydrous chemical composition of $SiO_2 \cdot 1/x XO_{1.5} \cdot m MO_{0.5}$, wherein X is the framework trivalent element, the molar ratio of Si/X is $x \geqslant 5$, M is a framework balanced cation, and the molar ratio of M/Si is $0 \leqslant m \leqslant 1$.

4. The novel silicate zeolite according to claim 3, wherein the novel silicate zeolite contains a three-dimensional pore system composed of 28×10×10-membered rings, wherein the 28-membered ring has a size of 23.11×12.39 Å, and the 10-membered ring has a size of 5.88 Å.

5. The novel silicate zeolite according to claim 3, wherein powder X-ray diffraction characteristics of the novel silicate zeolite are shown in the following table,

Diffraction angle 2θ (°)	**Interplanar spacing (Å)**	**Relative intensity**
3.15	**28.00**	**m**
4.89	**18.04**	**vs**
6.42	**13.76**	**mw**
6.64	**13.30**	**w**
7.75	**11.40**	**w**
7.94	**11.13**	**w**

8.65	10.21	w
9.33	9.48	w
9.66	9.15	w
9.93	8.90	w
10.27	8.60	w
10.75	8.22	w
12.15	7.28	w
13.54	6.53	w
14.12	6.27	w
14.66	6.04	w
14.94	5.92	w
15.55	5.69	w
15.88	5.58	w
16.05	5.52	w
17.42	5.09	w
17.83	4.97	w
18.10	4.90	w
18.75	4.73	w
19.05	4.65	w
19.76	4.49	w
20.12	4.41	w
20.79	4.27	w
21.42	4.15	w
22.80	3.90	mw
23.57	3.77	w
23.96	3.71	w
24.76	3.59	w
25.15	3.54	w
26.00	3.42	w
26.25	3.39	w
26.70	3.34	w
27.31	3.26	w
27.97	3.19	w
28.53	3.13	w
28.99	3.08	w
29.77	3.00	w
30.89	2.89	w

6. The novel silicate zeolite according to any one of claims 3-5, wherein the novel silicate zeolite is obtained after the novel silicate zeolite precursor according to claim 1 is calcined.

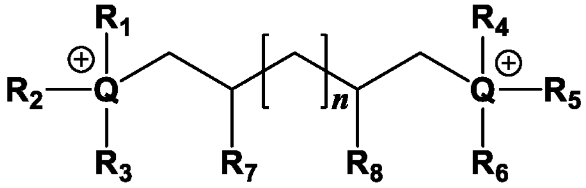
7. The zeolite according to claim 1 or 3, wherein the framework trivalent element X is selected from at least one of boron, aluminum, gallium, indium, iron and chromium; the framework balanced cation M is selected from at least one of hydrogen ion, ammonium ion, lithium ion, sodium ion, potassium ion,

rubidium ion and cesium ion; silicon in the zeolite contains a non-silicon tetravalent framework element (Y) with an amount of greater than or equal to 0 and less than or equal to 10w% of the mass of the silicon, wherein Y is selected from at least one of germanium, tin, titanium, zirconium and hafnium.

8. A preparation method of the novel silicate zeolite according to claim 3, wherein:

a framework balanced cation compound, a silicon source, a framework trivalent element X source, an organic structure directing agent and water are mixed to obtain a synthetic gel; then the synthetic gel is aged and hydrothermally crystallized, and the crystallized product is calcined to remove the organic structure directing agent; among which, the synthetic gel after aging has a molar ratio of $SiO_2:mM_2O:xX_2O_3:qQ(OH)_2:hH_2O$, wherein M is the framework balanced cation, X is the framework trivalent element, Q is a cationic group of the organic structure directing agent, and value ranges of m, x, q and h are: m=0-1, x=0-0.5, q=0.1-1, and h=3-100;

the organic structure directing agent has the following configuration:



wherein Q can be identical or different and selected from nitrogen or phosphorus, R_1, R_2, R_3, R_4, R_5 and R_6 can be identical or different and selected from cyclohexyl or phenyl, R7 and R8 are H or methyl, and n is 0-8.

9. A zeolite composition, containing the silicate zeolite precursor according to claim 1 and/or the silicate zeolite according to claim 3.

10. An application of the novel silicate zeolite according to claim 3, comprising an application of the silicate zeolite according to claim 3 or the zeolite composition according to claim 9 as an energy storage material, a chemical sensing material, a carrier, an adsorbent, a separating agent or a catalyst.

REVENDICATIONS

1. Un précurseur de tamis moléculaire de zéolite silicatée, caractérisé en ce que: la composition chimique anhydre du précurseur de tamis moléculaire de zéolite silicatée est en $SiO_2 \cdot 1/x XO_{1,5} \cdot m MO_{0,5} \cdot q Q$, où X représente des éléments trivalents de squelette, le rapport molaire Si/X $x \geq 5$, M représente des cations d'équilibre du squelette, le rapport molaire M/Si $0 \leq m \leq 1$, Q représente un agent directeur de la structure organique de l'ammonium bi-quaternaire ou du phosphore bi-quaternaire, le rapport molaire Q/Si $q \geq 0,01$.

2. Précurseur de tamis moléculaire de zéolite silicatée selon la revendication 1, caractérisé en ce que : la diffraction de rayons X sur poudre de précurseur de tamis moléculaire de zéolite silicatée présente les caractéristiques suivantes :

Angle de diffraction 2θ (°)	**Espacement interplanaire (Å)**	**Intensité relative**
3,27	**27,00**	**mw**
4,78	**18,47**	**s**
5,12	**17,25**	**vs**
5,42	**16,29**	**m**
6,58	**13,42**	**m**
7,92	**11,15**	**w**
8,37	**10,55**	**w**
8,75	**10,10**	**w**
9,34	**9,46**	**w**
9,58	**9,23**	**w**
9,96	**8,87**	**w**
10,15	**8,71**	**mw**
10,35	**8,54**	**mw**
10,55	**8,38**	**w**
10,85	**8,15**	**m**
11,36	**7,78**	**w**
11,74	**7,53**	**w**
12,84	**6,89**	**w**
13,56	**6,52**	**w**
13,97	**6,33**	**w**
14,41	**6,14**	**w**
14,66	**6,04**	**w**
15,10	**5,86**	**w**
15,53	**5,70**	**w**
15,94	**5,56**	**w**
16,53	**5,36**	**w**
16,78	**5,28**	**w**
17,17	**5,16**	**w**
17,83	**4,97**	**w**

18,20	**4,87**	**w**
18,48	**4,80**	**w**
18,81	**4,71**	**mw**
19,33	**4,59**	**mw**
19,53	**4,54**	**mw**
20,01	**4,43**	**w**
21,20	**4,19**	**w**
21,70	**4,09**	**mw**
21,88	**4,06**	**w**
22,60	**3,93**	**w**
22,98	**3,87**	**vs**
23,61	**3,77**	**w**
23,96	**3,71**	**mw**
24,42	**3,64**	**w**
24,72	**3,60**	**w**
25,00	**3,56**	**w**
25,18	**3,53**	**w**
25,63	**3,47**	**w**
25,99	**3,43**	**w**
26,66	**3,34**	**w**
27,67	**3,22**	**w**
28,01	**3,18**	**w**
28,55	**3,12**	**w**
29,12	**3,06**	**w**
29,82	**2,99**	**w**
30,18	**2,96**	**w**

3. Un nouveau type de tamis moléculaire de zéolite silicatée, caractérisé en ce que : la composition chimique anhydre du tamis moléculaire de zéolite silicatée est en $SiO_2\cdot 1/x XO_{1,5}\cdot m MO_{0,5}$, où X représente des éléments trivalents de squelette, le rapport molaire Si/X $x \geq 5$, M représente des cations d'équilibre du squelette, le rapport molaire M/Si $0 \leq m \leq 1$.

4. Un nouveau type de tamis moléculaire de zéolite silicatée selon la revendication 3, caractérisé en ce que ledit nouveau type de tamis moléculaire de zéolite silicatée contient un système de pores tridimensionnel composé de cycles à 28×10×10 chaînons, parmi lesquels le cycle à 28 chaînons a une taille de 23,11×12,39Å, et le cycle à 10 chaînons a une taille de 5,88Å.

5. Un nouveau type de tamis moléculaire de zéolite silicatée selon la revendication 3, caractérisé en ce que : ledit nouveau type de tamis moléculaire de zéolite silicatée présente les caractéristiques de diffraction de rayons X sur poudre indiquées dans le tableau ci-dessous,

Angle de diffraction 2θ (°)	Espacement interplanaire (Å)	Intensité relative
3,15	28,00	m
4,89	18,04	vs
6,42	13,76	mw
6,64	13,30	w
7,75	11,40	w
7,94	11,13	w
8,65	10,21	w
9,33	9,48	w
9,66	9,15	w
9,93	8,90	w
10,27	8,60	w
10,75	8,22	w
12,15	7,28	w
13,54	6,53	w
14,12	6,27	w
14,66	6,04	w
14,94	5,92	w
15,55	5,69	w
15,88	5,58	w
16,05	5,52	w
17,42	5,09	w
17,83	4,97	w
18,10	4,90	w
18,75	4,73	w
19,05	4,65	w
19,76	4,49	w
20,12	4,41	w
20,79	4,27	w
21,42	4,15	w
22,80	3,90	mw
23,57	3,77	w
23,96	3,71	w
24,76	3,59	w
25,15	3,54	w
26,00	3,42	w
26,25	3,39	w
26,70	3,34	w
27,31	3,26	w
27,97	3,19	w
28,53	3,13	w
28,99	3,08	w
29,77	3,00	w
30,89	2,89	w

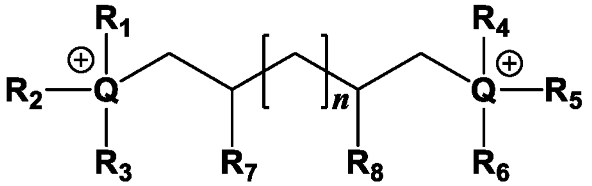
6. Un nouveau type de tamis moléculaire de zéolite silicatée selon l'une quelconque des revendications 3 à 5, caractérisé en ce que ledit nouveau type de tamis moléculaire de zéolite silicatée est obtenu par grillage dudit

précurseur d'un nouveau type de tamis moléculaire de zéolite silicatée selon la revendication 1.

7. Un tamis moléculaire selon la revendication 1 ou 3, caractérisé en ce que ledit (lesdits) élément(s) trivalent(s) de squelette est (sont) au moins un élément choisi parmi le bore, l'aluminium, le gallium, l'indium, le fer et le chrome ; ledit(lesdits) cations d'équilibre du squelette est (sont) choisi(s) parmi les ions hydrogène, les ions ammonium, les ions lithium, les ions sodium, les ions potassium, les ions rubidium et les ions césium ; le silicium dans le tamis moléculaire contient des éléments tétravalents de squelette hors silicium (Y) dont la masse est supérieure ou égale à 0 et inférieure ou égale à 10*w*% en poids, Y étant au moins un élément choisi parmi le germanium, l'étain, le titane, le zirconium et l'hafnium.

8. Procédé de préparation d'un nouveau type de tamis moléculaire de zéolite silicatée selon la revendication 3, caractérisé en ce que :
du composé cationique d'équilibre du squelette, la source de silicium, la source d'élément (s) trivalent(s) de squelette, de l'agent directeur de structure organique sont mélangés avec de l'eau pour obtenir un gel synthétique; ensuite, le gel synthétique fait l'objet d'un vieillissement et d'une cristallisation hydrothermale, et le produit issu de la cristallisation est grillé pour éliminer l'agent directeur de structure organique; le rapport molaire du gel synthétisé après vieillissement est $SiO_2{:}mM_2O{:}xX_2O_3{:}qQ(OH)_2{:}hH_2O$, où M représente le(s) cation(s) d'équilibre du squelette, X représente l'élément (les éléments) trivalent (s) de squelette et Q représente le groupe cationique d'agent directeur de structure organique, les valeurs des *m*, *x*, *q* et *h* étant retenues dans les plages suivantes : *m*=0-1, *x*=0 à 0,5, *q*=0,1 à 1, *h*=3 à 100 ;

Ledit agent directeur de structure organique a une configuration suivante :



Q pouvant être choisi parmi l'azote et le phosphore, pris seul ou en mélange, R_1, R_2, R_3, R_4, R_5, R_6 pouvant être choisis parmi le cyclohexyle ou le phényle, pris seuls ou en mélange, R_7 et R_8 étant choisis parmi H ou le méthyle, n étant compris entre 0 et 8.

9. Un composition de tamis moléculaire, caractérisée en ce que la composition contient le précurseur de tamis moléculaire de zéolite silicatée selon la revendication 1 et/ou le tamis moléculaire de zéolite silicatée selon la revendication 3.

10. Applications d’un nouveau type de tamis moléculaire de zéolite silicaté selon la revendication 3, caractérisée en ce que le tamis moléculaire de zéolite silicatée selon la revendication 3 ou la composition de tamis moléculaire selon la revendication 9 est utilisé comme matériau de stockage d'énergie ou matériau capteur chimique, support, adsorbant, agent de séparation ou catalyseur.

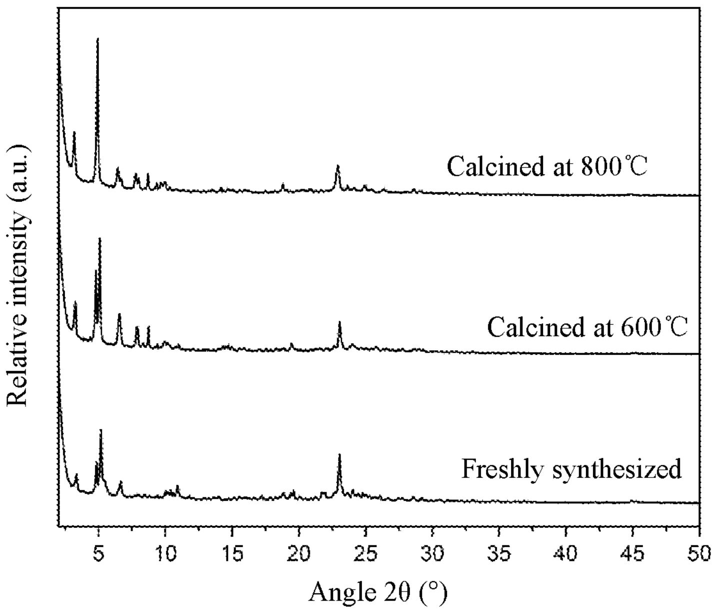

Relative intensity (a.u.)
Calcined at 800℃
Calcined at 600℃
Freshly synthesized
5
10
15
20
25
30
35
40
45
50
Angle 2θ (°)

Fig. 1

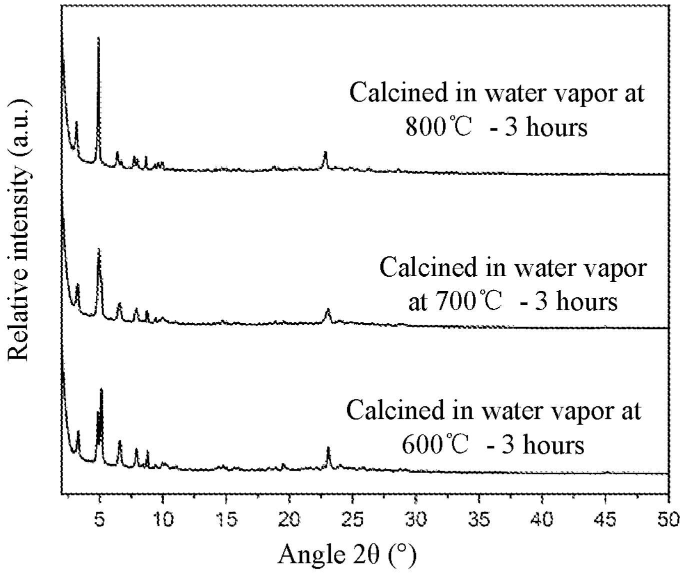

Calcined in water vapor at
800℃ - 3 hours
Calcined in water vapor
at 700℃ - 3 hours
Calcined in water vapor at
600℃ - 3 hours
Relative intensity (a.u.)
5
10
15
20
25
30
35
40
45
50
Angle 2θ (°)

Fig. 2

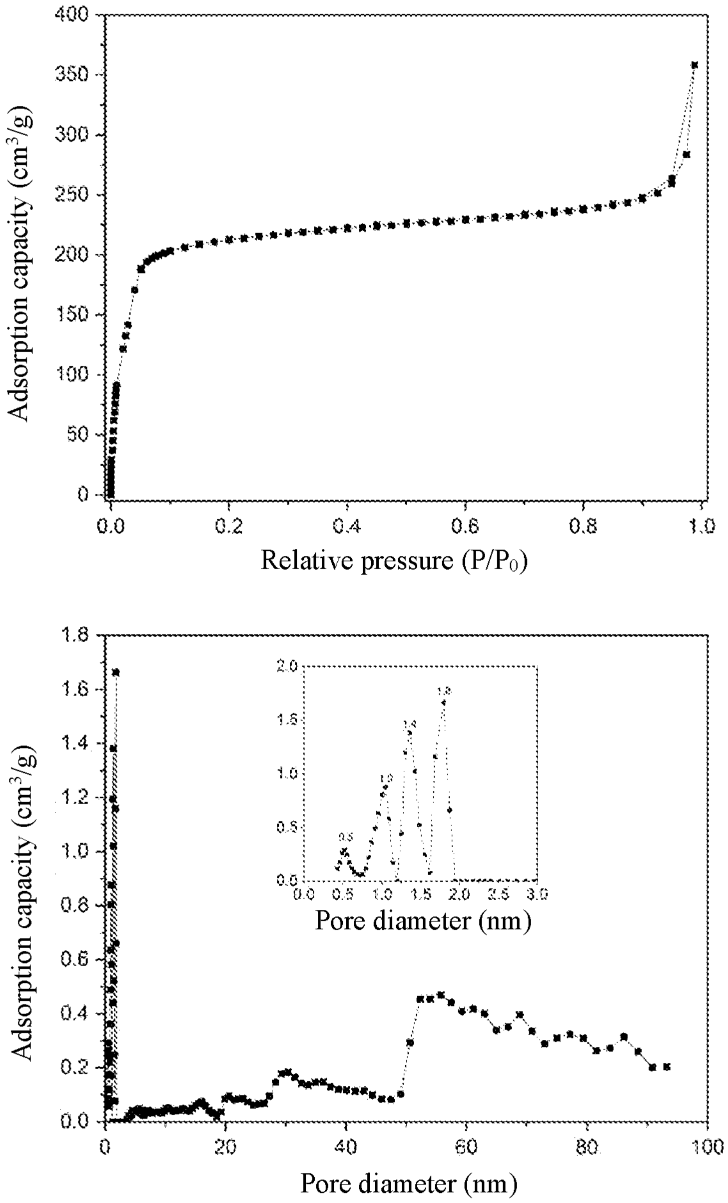

Adsorption capacity (cm³/g)
400
350
300
250
200
150
100
50
0
0.0
0.2
0.4
0.6
0.8
1.0
Relative pressure (P/P₀)
Adsorption capacity (cm³/g)
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
0
20
40
60
80
100
Pore diameter (nm)
2.0
1.5
1.0
0.5
0.0
0.0
0.5
1.0
1.5
2.0
2.5
3.0
0.5
1.0
1.4
1.8
Pore diameter (nm)

Fig. 3

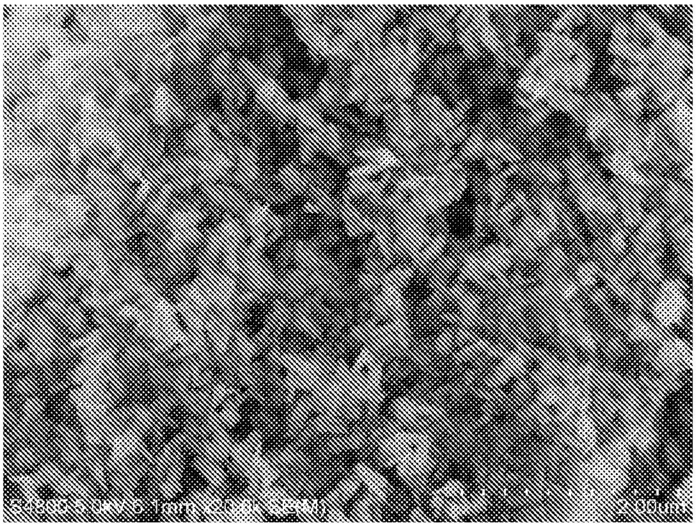


Fig. 4

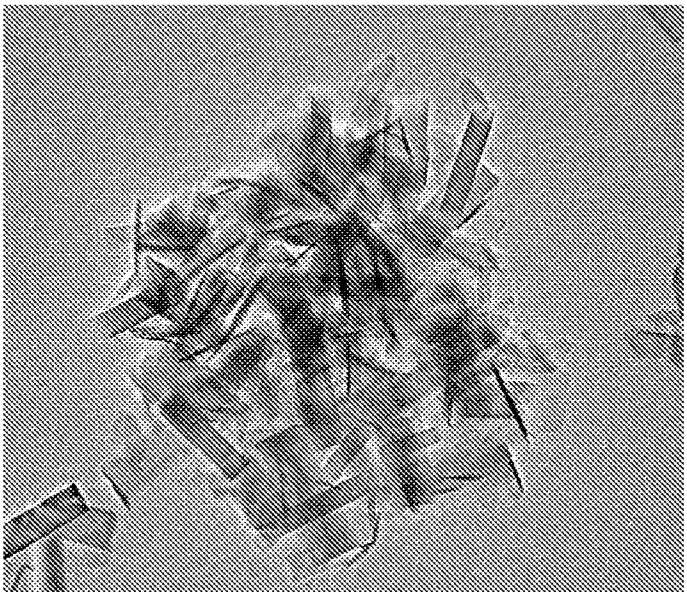


Fig. 5

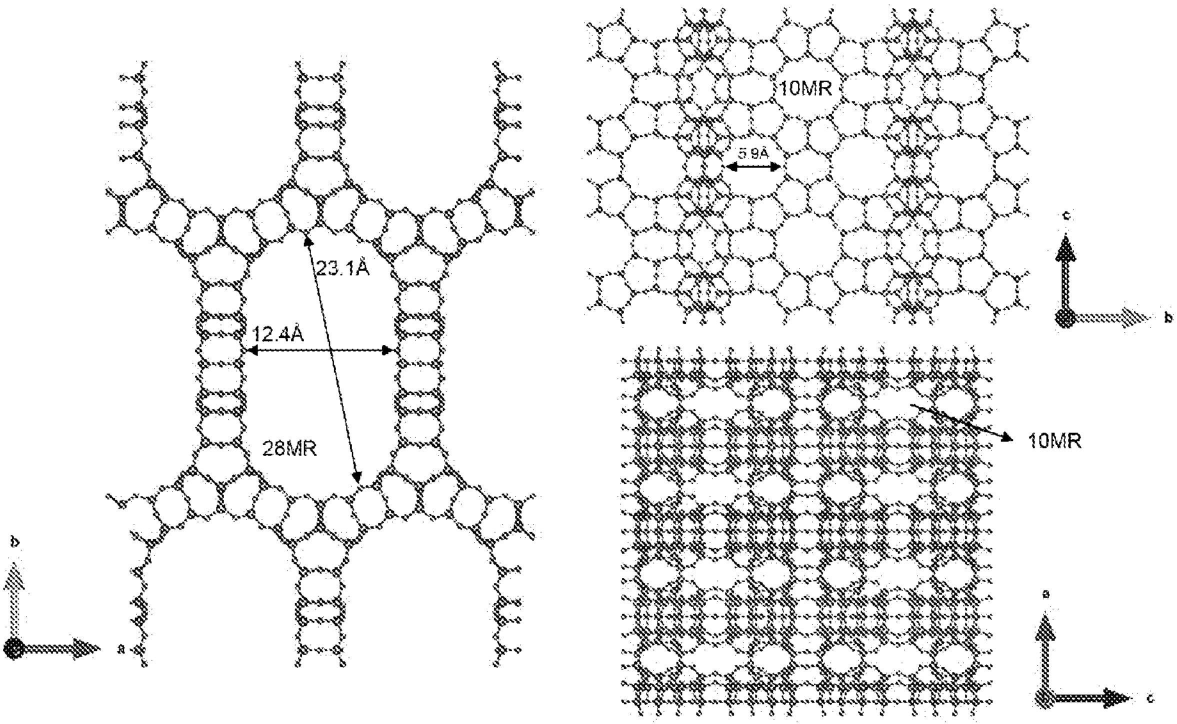


Fig. 6