Synthesis of mordenite from natural aluminosilicate clay minerals

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Abstract

This article reports the synthesis of mordenite from natural aluminosilicate clay minerals while the natural rectorite was activated via the SMS (sub-molten salt) activation method. Using the SMS activated rectorite and the SMS activated kaolinite as the raw material, respectively, a series of mordenite zeolites were synthesized, and the influences of different raw materials and synthesis conditions were investigated.

Keywords: Rectorite, Kaolinite, Sub-molten salt activation.

1. Introduction

Mordenite has been widely used as catalysts for hydroisomerization, alkylation and hydrocracking processes[1] due to its specific pore structure, high thermal stability and excellent acidity[2]. Nowadays, mordenite is synthesized from inorganic chemicals such as sodium silicate and aluminum sulfate, whose manufacture from natural aluminosilicate minerals is associated with huge waste discharge and extensive energy consumption.

Therefore, fabricating zeolites directly from natural aluminosilicate minerals attracts lots of attention. However, there exist two problems when using natural aluminosilicate minerals as raw materials for fabricating zeolites: on the one hand, the traditional thermal activation and alkali fusion activation methods need high temperature; on the other hand, some amount of chemicals should be added into the synthesis system to modulate the SiO_2/Al_2O_3 molar ratio, especially for synthesis zeolites with medium or high SiO_2/Al_2O_3 molar ratio[3], such as mordenite or ZSM-5. Therefore, our group has developed a sub-molten salt (SMS) activation method to decrease the energy consumption of activation and promote the utilization efficiency of silicon and aluminum species[4] in the natural minerals, and Y zeolite has been successfully synthesized using the resulted SMS activated kaolinite as the starting material and diatomite as the supplementary silica source[5].

2. Experimental

In a typical synthesis, rectorite powder (or kaolinite powder) activated via the SMS method at 250 ^{o}C was mixed with diatomite powder calcined at 600 ^{o}C, tetraethylammonium bromide (TEABr) and deionized water under gentle stirring; then, the resultant mixture was crystallized at 100 ^{o}C for 36 h and 170 ^{o}C for 48 h; finally, the solid product was recovered by filtration, washing and drying at 100 ^{o}C overnight.

3. Results and discussion

From the X-ray powder diffraction characterization result of the as-synthesized sample, it can be seen that the diffraction peaks of the as-synthesized sample are exclusively characteristic of the typical MOR structure, evidencing that the sample is pure-phase mordenite with a high relative crystallinity.

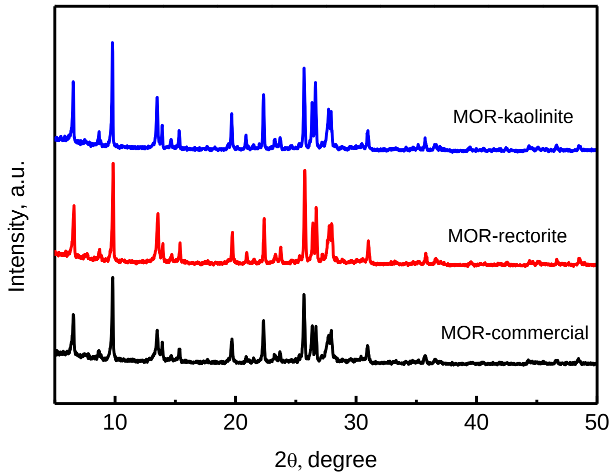

Figure 1. XRD patterns of the MOR samples.

From the chemical compositions of MOR-commercial and mordenite synthesized from natural minerals, it can be seen that the framework SiO_2/Al_2O_3 molar ratios of the mordenite products synthesized respectively from the SMS activated rectorite and kaolinite using or without using organic template are similar and both smaller than that of MOR-commercial synthesized from chemical reagent, thus they both possess more acid amount. Meanwhile the contents of metal atoms such as Na and Mg, especially Fe are larger than that of MOR-commercial because of the natural aluminosilicate clay minerals are rich with these metals.

Samples	Component (wt.%)							SiO_2/Al_2O_3 molar ratio
	Na_2O	MgO	Al_2O_3	SiO_2	CaO	TiO_2	Fe_2O_3	
MOR-kaolinite (using TEABr)	0.85	0.25	11.85	84.15	0.83	0.96	0.73	12.1
MOR-rectorite (using TEABr)	0.37	0.12	12.30	85.00	0.43	0.26	1.09	11.7
MOR-kaolinite (without TEABr)	1.39	0.30	16.46	79.73	0.26	0.26	1.00	8.2
MOR-commercial	0.05	--	11.58	87.89	--	0.37	0.07	12.9

Table 1. Chemical compositions of MOR-commercial and mordenite synthesized from natural minerals.

4. Conclusions

Mordenite was synthesized from the SMS activated rectorite and thermal activated diatomite. The results indicate that the relative crystallinity degree could be up to 82%. However, under the same conditions, relative crystallinity degree of mordenite synthesized from the SMS activated kaolinite could be up to 100%.

The framework SiO_2/Al_2O_3 molar ratios of the mordenite products synthesized respectively from the SMS activated rectorite and kaolinite using organic template are similar and both smaller than that of MOR-commercial synthesized from chemical reagent, thus they both possess more acid amount. However, the framework SiO_2/Al_2O_3 molar ratio of the mordenite product synthesized from the SMS activated kaolinite without using the organic template is less than that of the product synthesized using the organic template, thus it possesses higher acid amount.

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