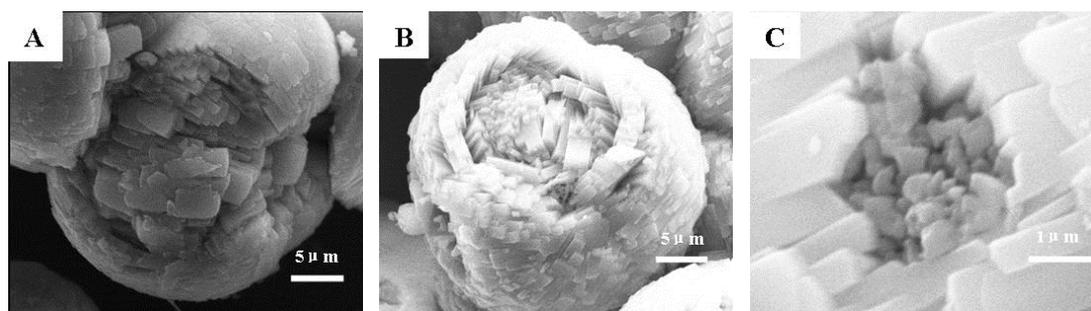


## Supporting Information

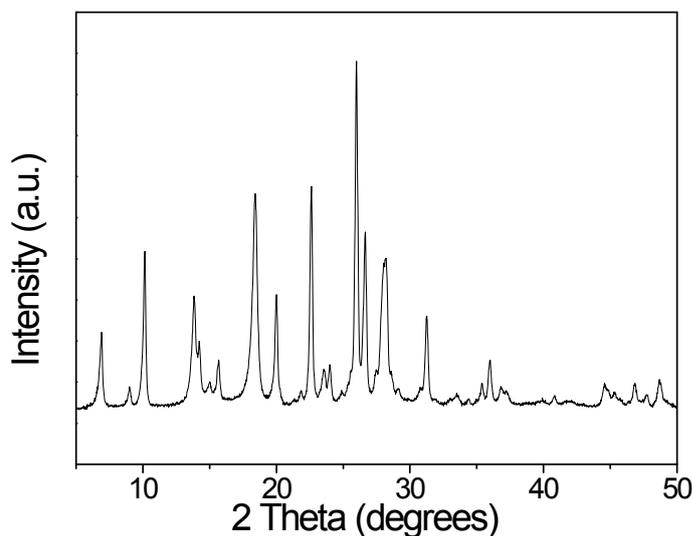
### Morphology controlled synthesis of large mordenite crystals

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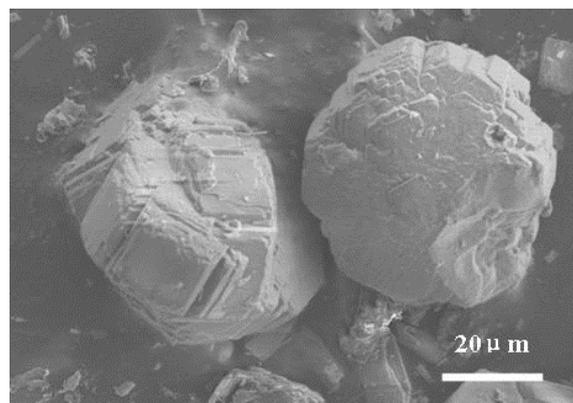
*State Key Laboratory of Materials-Oriented Chemical Engineering, College of Chemistry and Chemical Engineering, Nanjing Tech University, Nanjing, 210009, Jiangsu, China. \*Corresponding author, Tel: +86-25-83172264, Fax: +86-25-83172261, E-mail: junwang@njtech.edu.cn*



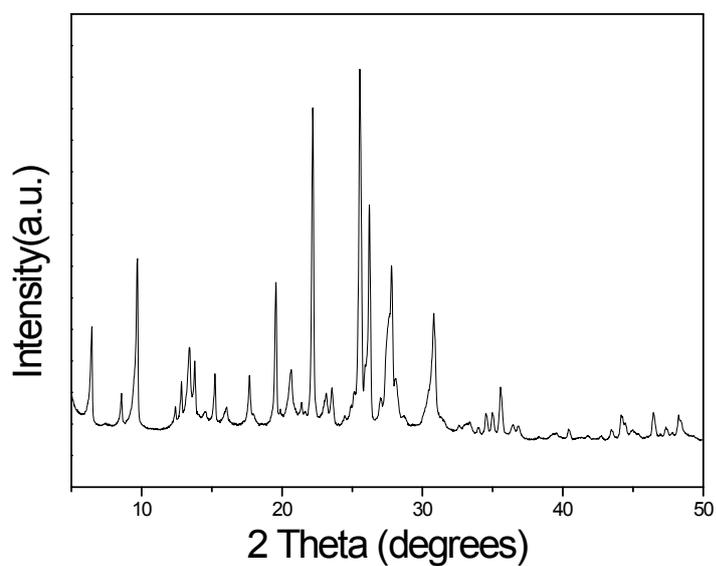
**Figure S1** SEM images of the internal structure of the MOR microsphere aggregates obtained with the crystallization time of 4 d and crystallization temperature of 443 K by using the gel composition of 15SiO<sub>2</sub>/8Na<sub>2</sub>O/900H<sub>2</sub>O/3.5TEAOH/1Al<sub>2</sub>O<sub>3</sub>.



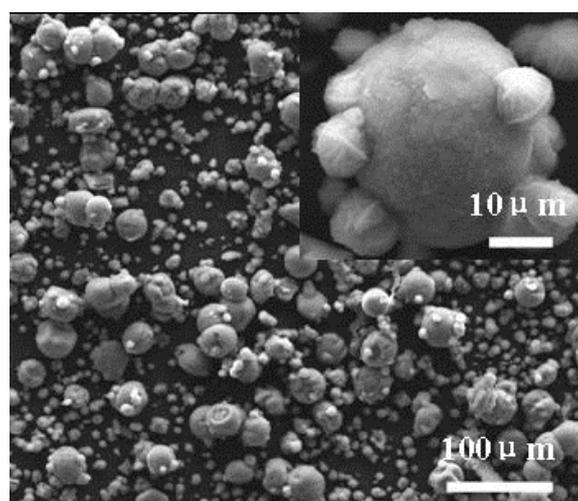
**Figure S2** XRD pattern of as-calcined MOR sample obtained by base-hydrolysis of TEOS.



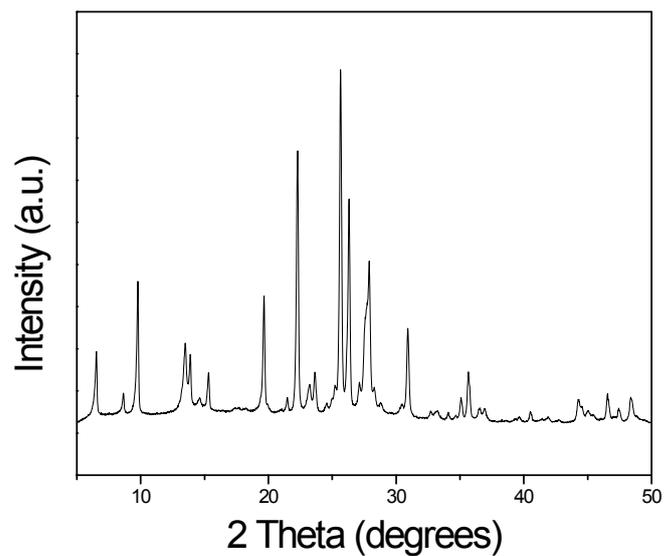
**Figure S3** SEM image of the as-calcined MOR sample obtained by base-hydrolysis of TEOS.



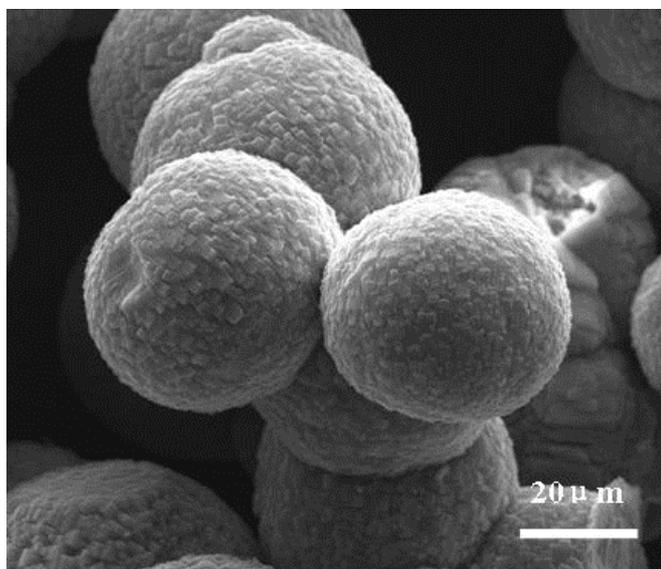
**Figure S4** XRD pattern of as-calcined sample obtained with the crystallization time of 4 d and crystallization temperature of 443 K by using the gel composition of  $15\text{SiO}_2/9\text{Na}_2\text{O}/900\text{H}_2\text{O}/3.5\text{TEAOH}/1\text{Al}_2\text{O}_3$ .



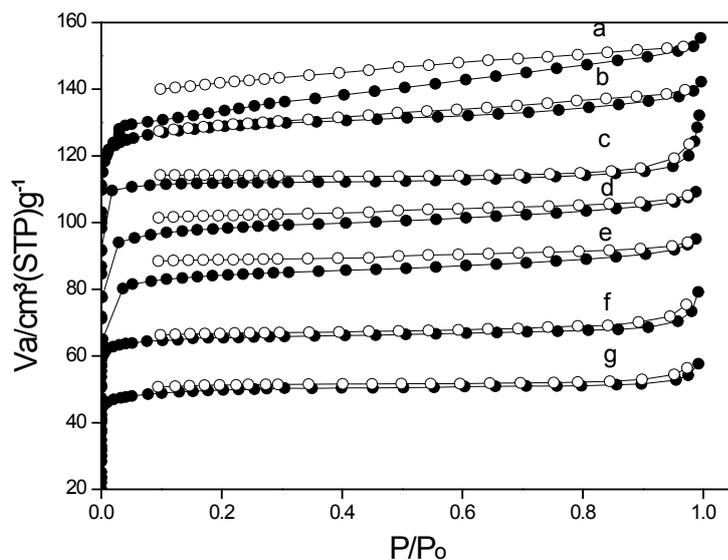
**Figure S5** SEM image of as-calcined sample obtained with the crystallization time of 4 d and crystallization temperature of 443 K by using the gel composition of  $15\text{SiO}_2/9\text{Na}_2\text{O}/900\text{H}_2\text{O}/3.5\text{TEAOH}/1\text{Al}_2\text{O}_3$ .



**Figure S6** XRD pattern of the as-calcined sample obtained with the crystallization time of 9 d and crystallization temperature of 443 K with the gel composition of 15SiO<sub>2</sub>/8Na<sub>2</sub>O/900H<sub>2</sub>O/3.5TEAOH/1Al<sub>2</sub>O<sub>3</sub>.



**Figure S7** SEM image of the as-calcined sample obtained with the crystallization time of 9 d and crystallization temperature of 443 K with the gel composition of 15SiO<sub>2</sub>/8.0Na<sub>2</sub>O/900H<sub>2</sub>O/3.5TEAOH/1Al<sub>2</sub>O<sub>3</sub>.



**Figure S8** N<sub>2</sub> sorption isotherms of selected as-calcined samples synthesized with TEAOH (a: Entry 1 and b: Entry 8) and all the as-calcined samples synthesized without using TEAOH (c: Entry 12; d: Entry 13; e: Entry 14; f: Entry 15; g: Entry 16). The adsorption isotherms for samples a, b, c, d, e, f and g are shifted by 18, 10, 29, 11, 0, -10 and -21 cm<sup>3</sup> g<sup>-1</sup>. The filled circles indicate adsorption and the hollow circles indicate desorption.

Table S1 Surface area and pore volume of the as-calcined samples in Figure S8.

Sample	Surface area (m <sup>2</sup> g <sup>-1</sup> )	Pore volume (cm <sup>3</sup> g <sup>-1</sup> )
a: Entry 1	405	0.21
b: Entry 8	387	0.20
c: Entry 12	338	0.18
d: Entry 13	267	0.15
e: Entry 14	258	0.15
f: Entry 15	248	0.14
g: Entry 16	232	0.12