Figure S1: $^{27}$Al MAS NMR spectra of (a) Al-MWW(P), (b) silylated Al-MWW(P), (c) Al-IEZ-MWW and (d) Al-MWW.
Electric Supporting Information 2

$^{29}$Si MAS NMR

Fig. S2  $^{29}$Si MAS NMR of (a) Al-MWW(P), (b) silylated Al-MWW(P) and (c) Al-IEZ-MWW.

D$^2$ and D$^1$ peaks in Fig. S2(b) are corresponding to the interlayer-modified dimethylsilane species, and the dimethylsilane species reacted with silanols on the external surface, respectively. Q$^2$ peak in Fig. S2(c) is corresponding to the monomeric silica puncheons. After the silylation and the following calcination, the intensity of Q$^3$ peak was clearly minimized, due to the interlayer-silylation.
Electric Supporting Information 3
Cyclohexane adsorption isotherms

Figure S3  Cyclohexane adsorption isotherms of Al-IEZ-MWW (●) and Al-MWW (○).
**Electric Supporting Information 4**

Micropore size distribution (from argon adsorption-desorption isotherm)

Figure S4  Micropore size distributions of (a) Al-MWW and (b) Al-IEZ-MWW.
Electric supporting Information 5
NH₃ temperature-programmed desorption (TPD) profiles

Figure S5  NH₃-TPD profiles of (a) Al-MWW, (b) Al-IEZ-MWW and (c) Al-beta.
Electric Supporting Information 6

Set up of autoclave

Figure S6  Set up of an autoclave for the vapor-phase silylation of Al-MWW(P).