Molecular insights into adsorption and diffusion mechanism of n-hexane in MFI zeolites with different Si-to-Al ratio and counterions

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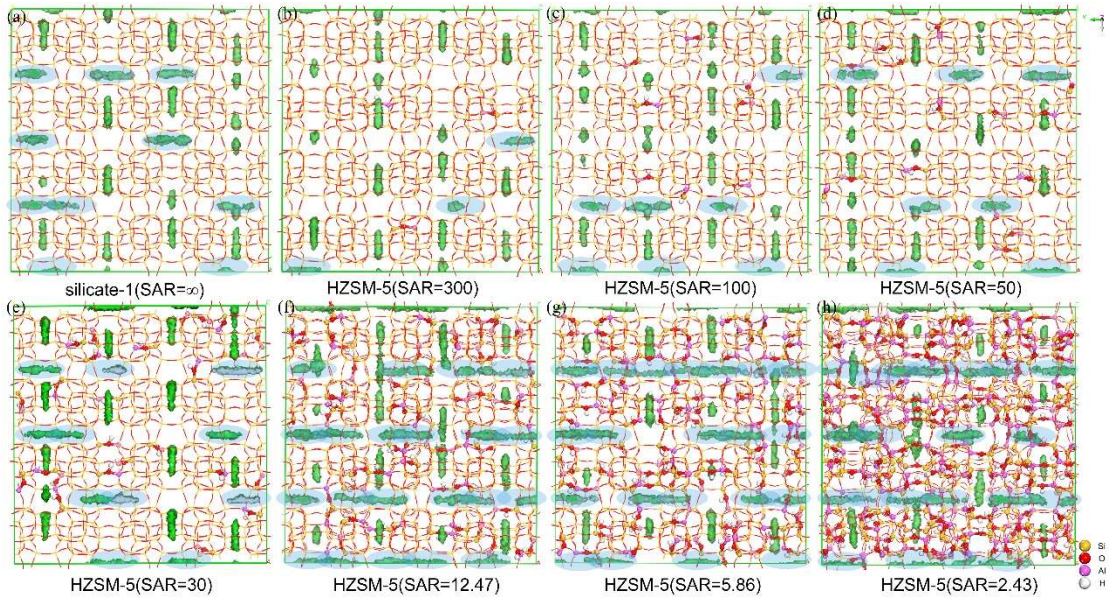

Figure S1. Density distribution of n-hexane on silicate-1 (a) and HZSM-5 with different SAR of 300 (b), 100 (c), 50 (d), 30 (e), 12.47 (f), 5.86 (g) and 2.43 (h) at 423 K. The X-axis direction (vertical direction in picture) represents the sinusoidal channels; the Y-axis direction (horizontal direction in picture) represents the straight channels (blue shadow area). The green region represents the lowest-energy, preferred adsorption positions for n-hexane molecules.

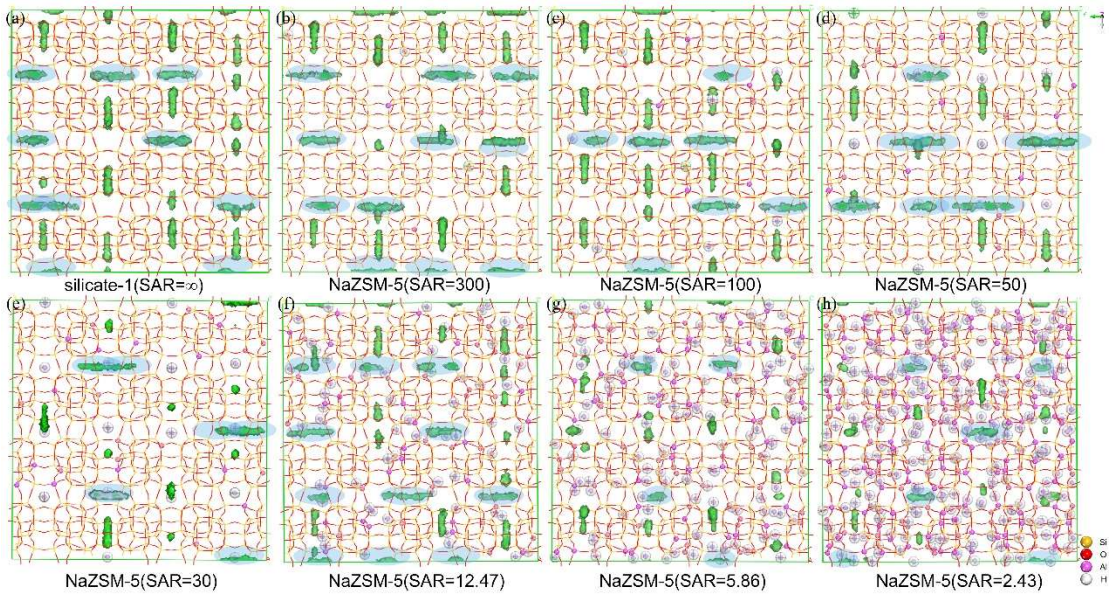

Figure S2. Density distribution of n-hexane on silicate-1 (a) and NaZSM-5 with different SAR of 300 (b), 100 (c), 50 (d), 30 (e), 12.47 (f), 5.86 (g) and 2.43 (h) at 423 K. The X-axis direction (vertical direction in picture) represents the sinusoidal channels; the Y-axis direction (horizontal direction in picture) represents the straight channels (blue shadow area). The green region represents the lowest-energy, preferred adsorption positions for n-hexane molecules.

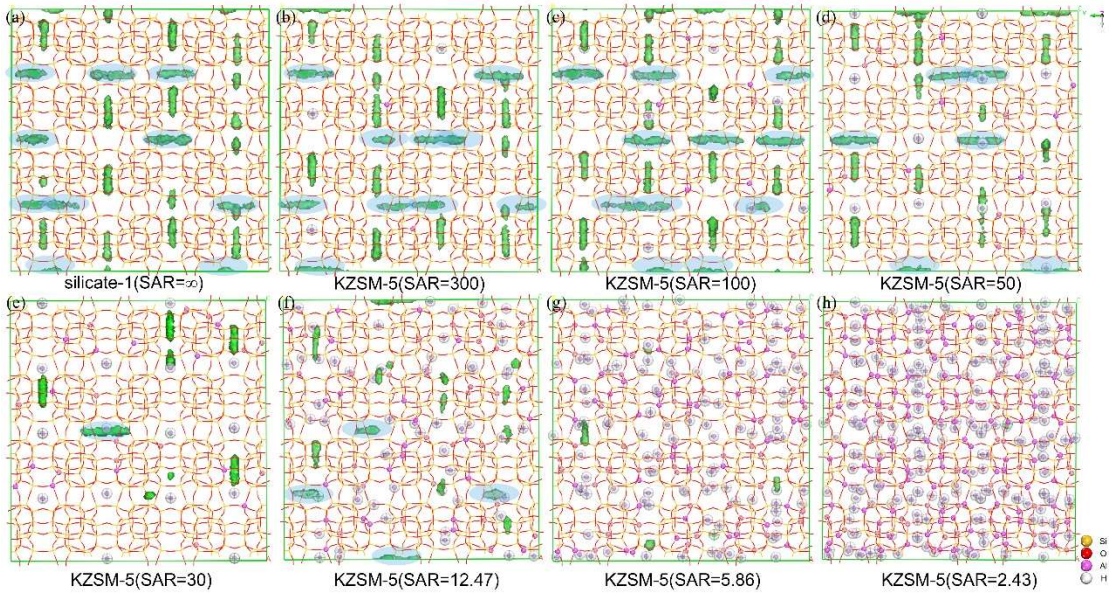

Figure S3. Density distribution of n-hexane on silicate-1 (a) and KZSM-5 with different SAR of 300 (b), 100 (c), 50 (d), 30 (e), 12.47 (f), 5.86 (g) and 2.43 (h) at 423 K. The X-axis direction (vertical

direction in picture) represents the sinusoidal channels; the Y-axis direction (horizontal direction in picture) represents the straight channels (blue shadow area). The green region represents the lowest-energy, preferred adsorption positions for n-hexane molecules.

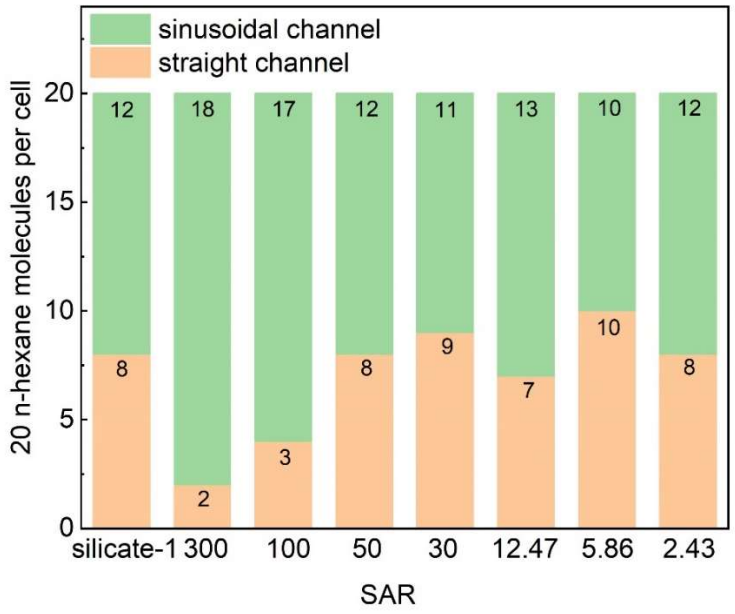

Figure S4 Number of n-hexane molecules in straight (see yellow rectangle) and sinusoidal (see green rectangle) channels in the HMFI and silicate-1 models used for MD calculations

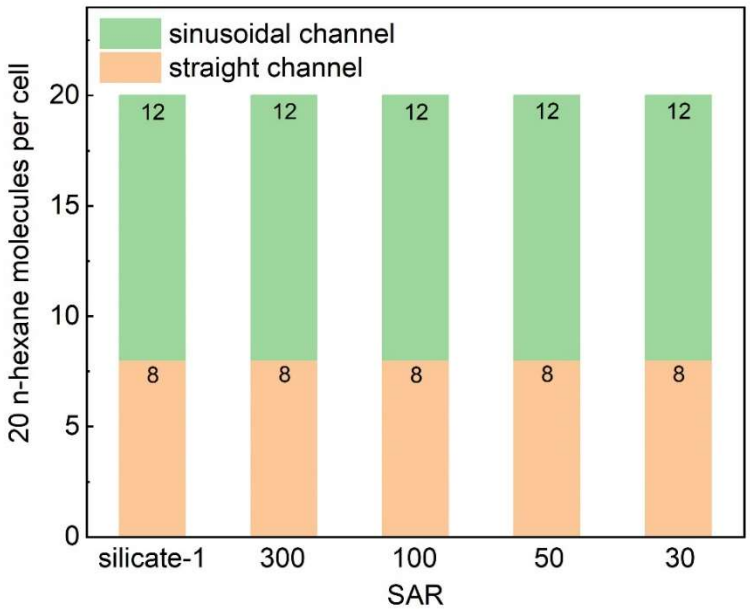

Figure S5 Number of n-hexane molecules in straight (see yellow rectangle) and sinusoidal (see green rectangle) channels in the NaMFI and silicate-1 models used for MD calculations

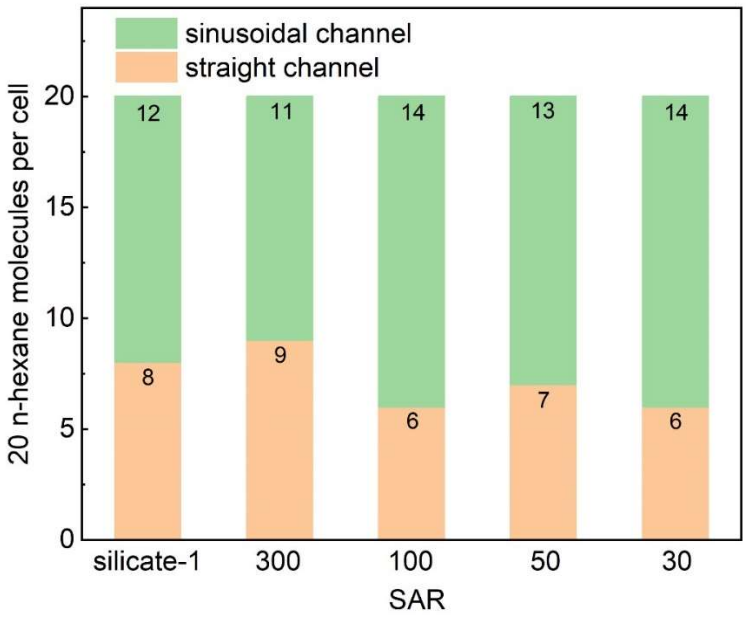

Figure S6 Number of n-hexane molecules in straight (see yellow rectangle) and sinusoidal (see green rectangle) channels in the KMFI and silicate-1 models used for MD calculations

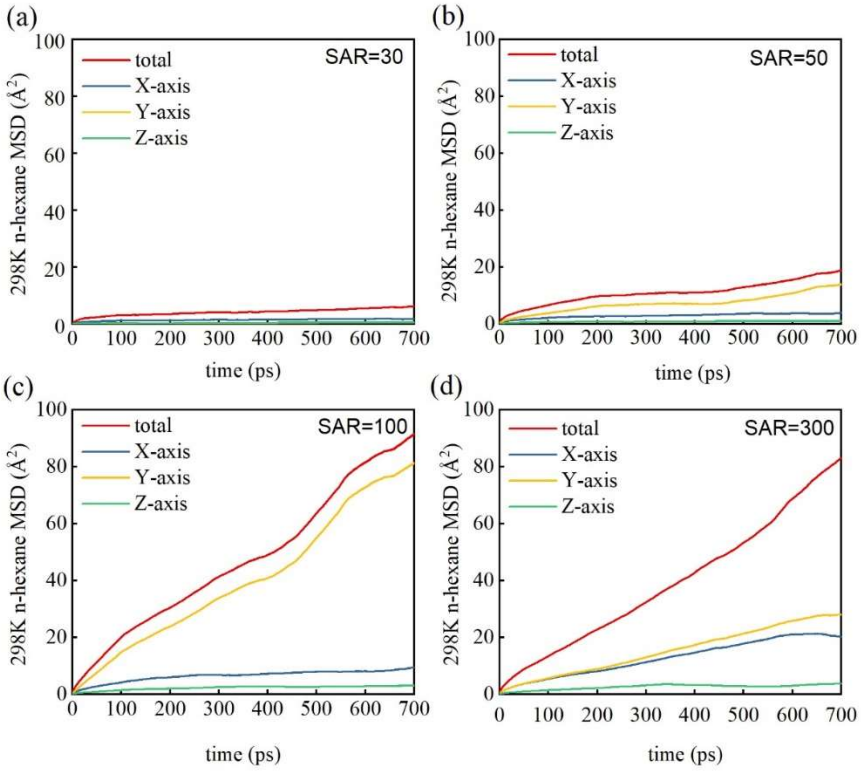

Figure S7. MSD of anisotropic diffusion coefficient of n-hexane molecules on NaZSM-5 with different SAR of 300 (d), 100 (c), 50 (b) and 30 (a) at 298 K

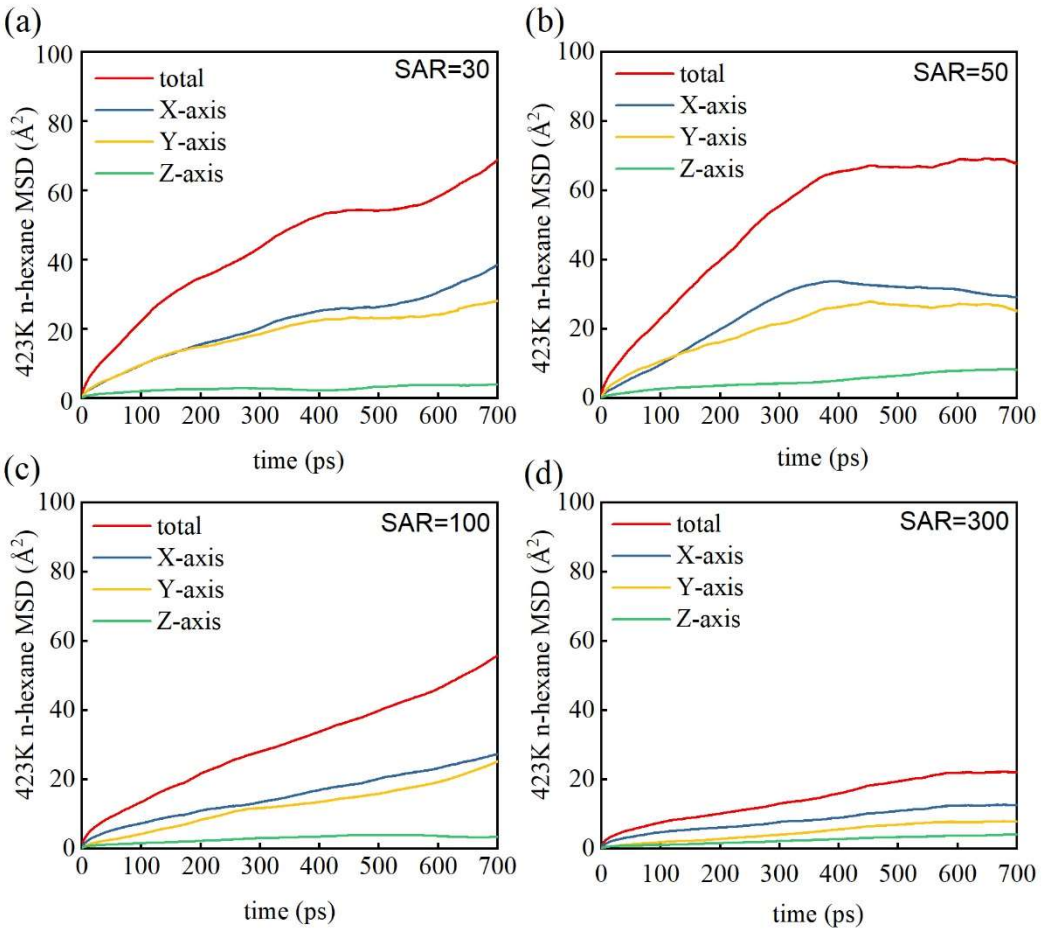

Figure S8. MSD of anisotropic diffusion coefficient of n-hexane molecules on NaZSM-5 with different SAR of 300 (d), 100 (c), 50 (b) and 30 (a) at 423 K

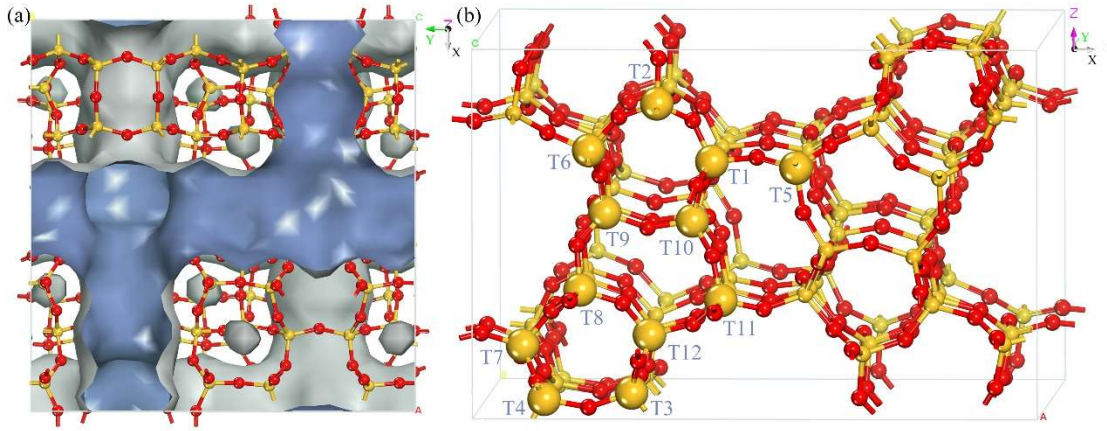

Figure S9. Unite cell structure of the orthorhombic MFI framework with intersectional straight and sinusoidal ten-membered-ring channels (a) and all 12 T sites (b). The X-axis direction represents the sinusoidal channels; the Y-axis direction represents the straight channels.

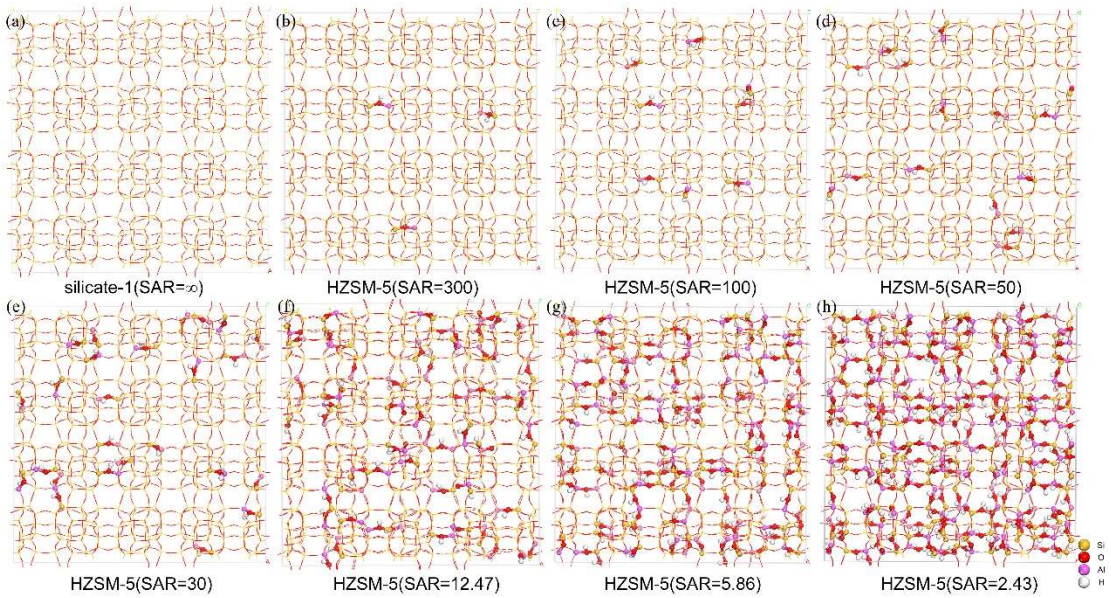

Figure S10. Optimized structures of silicate-1 (a) and HZSM-5 with different SAR of 300 (b), 100 (c), 50 (d), 30 (e), 12.47 (f), 5.86 (g) and 2.43 (h).

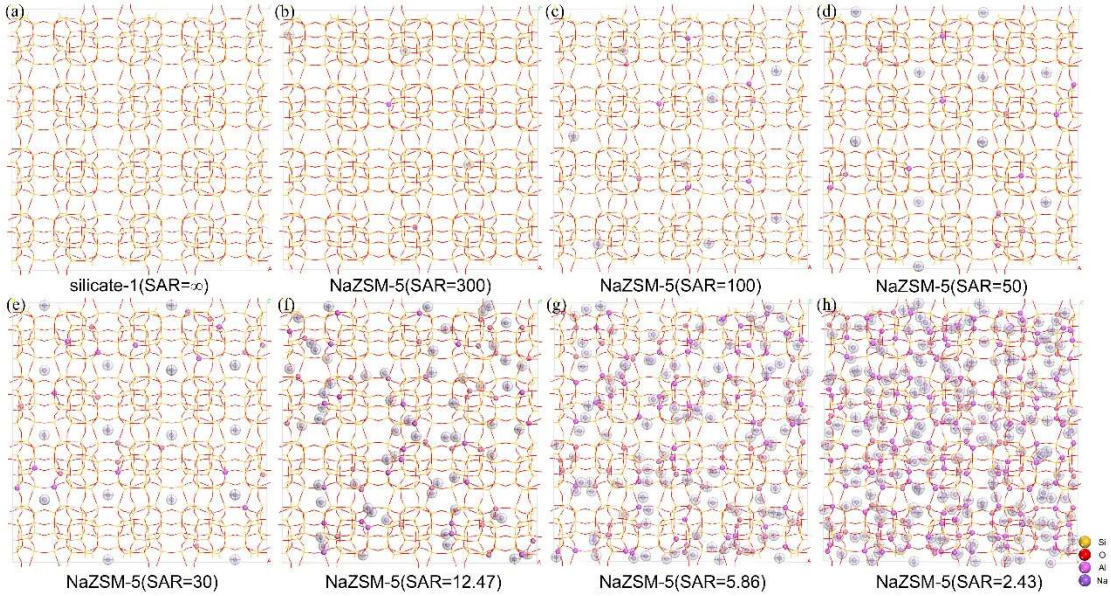

Figure S11. Optimized structures of silicate-1 (a) and NaZSM-5 with different SAR of 300 (b), 100 (c), 50 (d), 30 (e), 12.47 (f), 5.86 (g) and 2.43 (h).

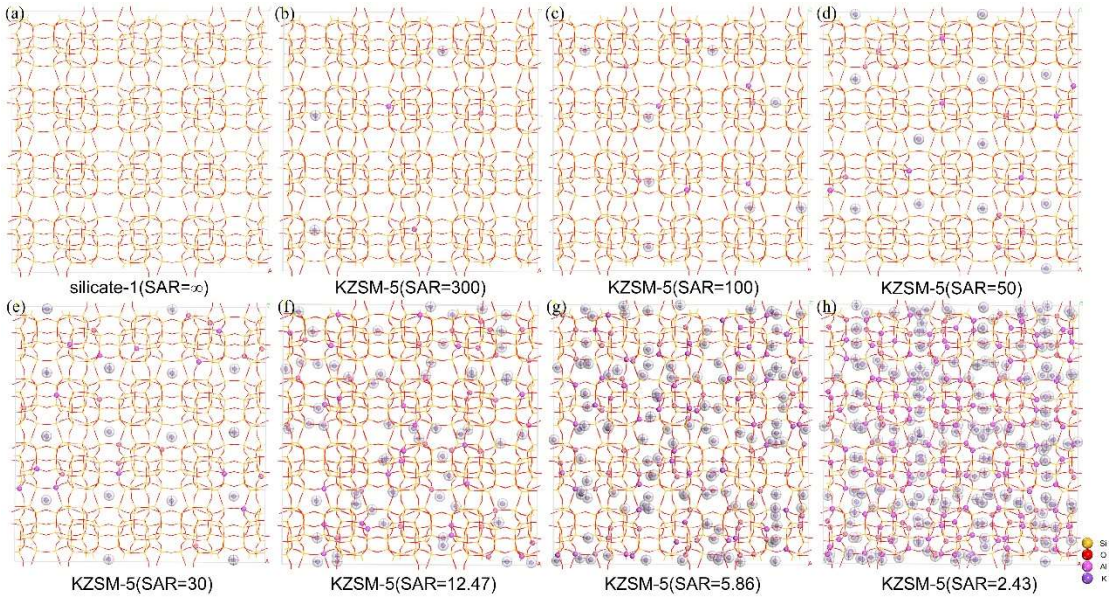

Figure S12. Optimized structures of silicate-1 (a) and KZSM-5 with different SAR of 300 (b), 100 (c), 50 (d), 30 (e), 12.47 (f), 5.86 (g) and 2.43 (h).

Table S1. The R^2 values of linear fitting data of adsorption heats

Zeolite/Temperature	298K	423K
Silicate-1	0.94	0.98
HMFI(300)	0.65	0.85
HMFI(100)	0.90	0.83
HMFI(50)	0.93	0.93
HMFI(30)	0.97	0.98