Electronic Supplementary Information (ESI)

2D-to-disguised 3D materials with built-in acid sites: H^+-[Al]-RUB-18

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Unit Cell Determination

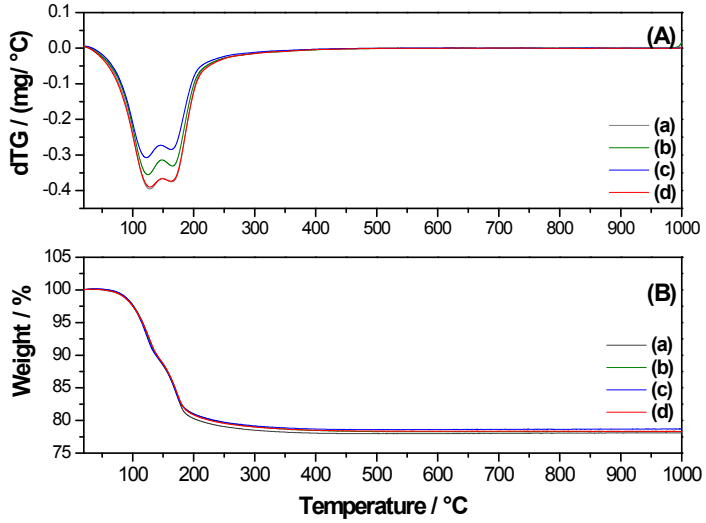

Figure S1. Thermogravimetry (A) and its derivative (B) of the (a) Na-R18, (b) Na-[Al15_22]-R18, (c) Na-[Al30_42]-R18, and (d) Na-[Al60_40]-R18.

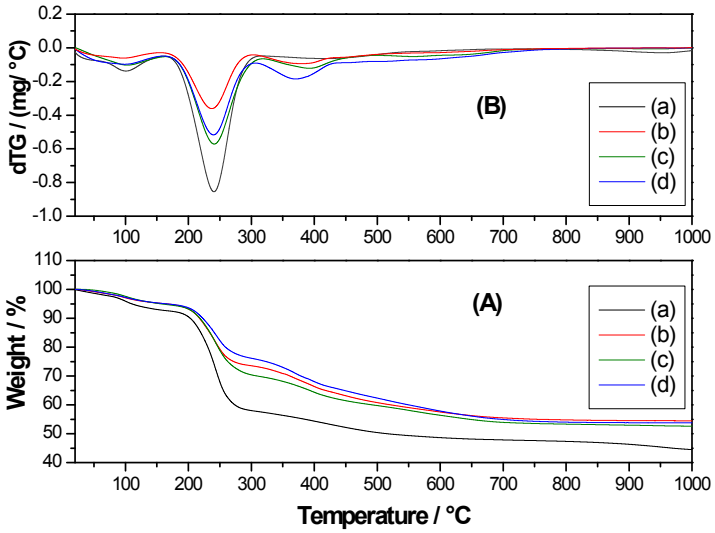

Figure S2. Thermogravimetry (A) and its derivative (B) of (a) CTA-R18, (b) CTA-[Al15_22]-R18, (c) CTA-[Al30_42]-R18, and (d) CTA-[Al60_40]-R18.

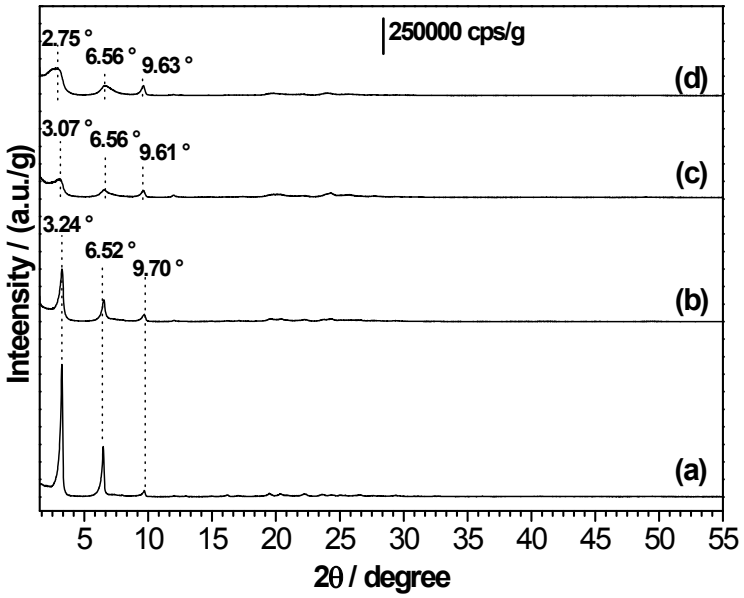

Figure S3. Powder XRD patterns of (a) CTA-RUB-18, (b) CTA-[Al15_22]-R18, (c) CTA-[Al30_42]-R18 and (d) CTA-[Al60_40]-R18.

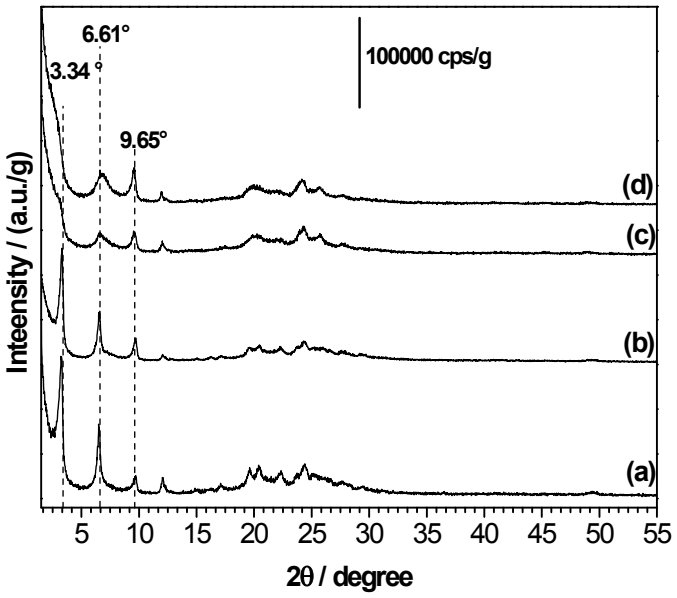

Figure S4 Powder XRD patterns of the as synthesized pillared materials (a) R18-pil, (b) [Al15_22]-R18-pil, (c) [Al30_42]-R18-pil and (d) [Al60_40]-R18-pil.

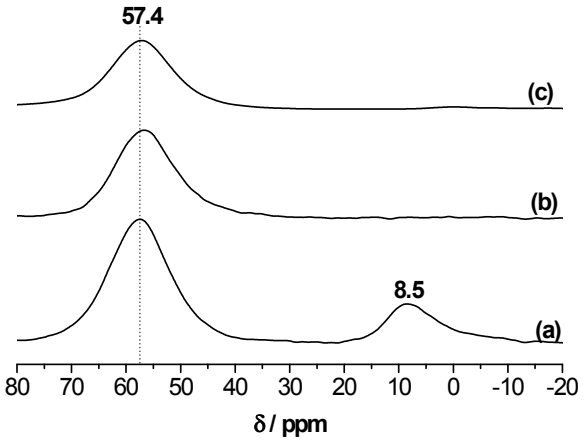

Figure S5. ^{27}Al NMR spectra of (a) CTA-[Al15_22]-R18, (b) CTA-[Al30_42]-R18 and (c) CTA-[Al60_40]-R18.

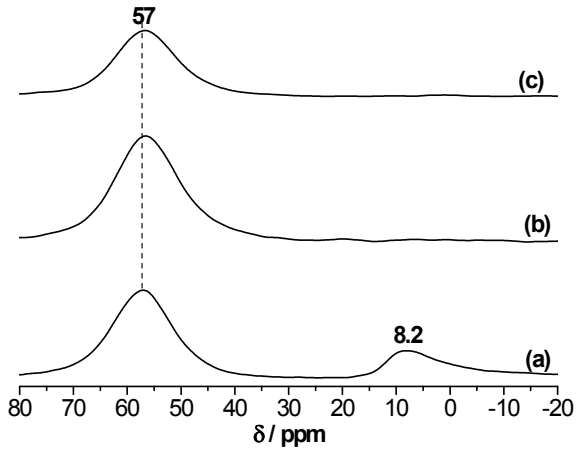

Figure S6. ^{27}Al NMR spectra of (a) [Al15_22]-R18-pil, (b) [Al30_42]-R18-pil and (c) [Al60_40]-R18-pil.

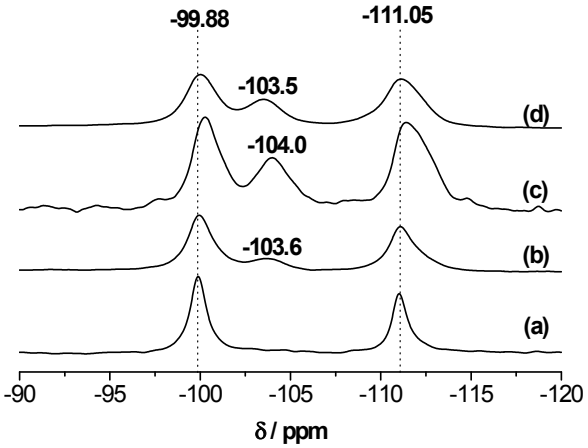

Figure S7. ^{29}Si NMR spectra of (a) CTA-R18, (b) CTA-[Al15_22]-R18, (c) CTA-[Al30_42]-R18, and(d) CTA-[Al60_40]-R18.

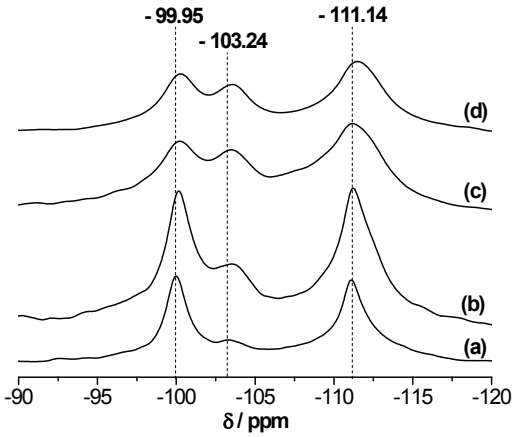

Figure S8. ^{29}Si NMR spectra of (a) [Al15_22]-R18-pil, (b) [Al30_42]-R18-pil and (c) [Al60_40]-R18-pil.

Table S1. Empirical Formula of the Na-[Al]-RUB-18 aluminosilicates prepared through ICP OES and TGA data.

Sample	Empirical Formula
Na-[Al15_22]-R18	$Na_{7,4}[Si_{30,63}Al_{1,37}O_{64}(OH)_8]\cdot 25H_2O$
Na-[Al30_42]-R-18	$Na_{8,1}[Si_{31,25}Al_{0,75}O_{64}(OH)_8]\cdot 24H_2O$
Na-[Al60_40]-R-18	$Na_{9,8}[Si_{31,22}Al_{0,78}O_{64}(OH)_8]\cdot 26H_2O$

Table S2. Chemical Analysis of the samples exchanged with CTA^+ ions, CTA^+/unit cell and ion exchange degree for each sample.

CTA-sample	C / %*	N / %*	H /%*	CTA / mol	Ion Exchange degree / %[a]
CTA-R18	35.20	1.63	6.79	3.5	45
CTA-[Al15_22]-R18	31.02	1.43	6.71	3.2	42
CTA-[Al30_42]-R18	28.87	1.19	6.27	3.1	38
CTA-[Al60_40]-R18	21.15	1.04	6.07	3.0	31

*%= m/m; [a]100% = all Na^+ ions were replaced by CTA^+ ions

Unit cells calculated from CHN (Table S1) and TGA (Fig S1):

- **CTA-RUB-18:**

$Na_{4,4}CTA_{3,6}[Si_{32}O_{64}(OH)_8]\cdot 14{,}91H_2O$

- **CTA-[Al15_22]RUB-18:**

$Na_{4,3}CTA_{3,2}[Si_{30,63}Al_{1,37}O_{64}(OH)_8]\cdot 14{,}67H_2O$

- **CTA-[Al30_42]RUB-18:**

$Na_{3,1}CTA_5[Si_{31,25}Al_{0,75}O_{64}(OH)_8]\cdot 12{,}32H_2O$

- **CTA-[Al60_40]RUB-18:**

$Na_{6,8}CTA_3[Si_{31,22}Al_{0,78}O_{64}(OH)_8]\cdot 14{,}12H_2O$