Electronic Supplementary Information for

Continuous solid solutions constructed from two isostructural octahedron-based molecular sieves: preparation, acidity regulation and catalytic application in Strecker reactions

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1. Spectral data for imines precursors and α-aminonitriles

The molecular structures of all obtained imines precursors and α-aminonitriles are known and also have been reported in previous references, therefore, only $^1$H NMR and $^{13}$C NMR spectra were used to qualitatively identify their structure. The results were proved to be identical to the standard sample in the literature, which were found in the Figures S1-S20, molt point found form these references were also included here. For some compounds, such as 1b, 1c, 2c 2e, the data of melt points were not available from the references, however their high resolution mass spectra data were acquired to further verify their molecular structure, which included also in the Figures S21-S24.
(1) 4-methyl-N-phenylmethylene-benzenesulfonamide (1a)

![Chemical structure of 1a](image)

**Data of 1a:** Melting point \[^{[1]}\], 109-111 °C; \(^1\)H NMR (400 MHz, DMSO-\(d_6\)) \(\delta\): 9.15 (s, 1H), 8.08-7.99 (m, 2H), 7.85 (d, \(J = 8.3\) Hz, 2H), 7.72 (t, \(J = 7.4\) Hz, 1H), 7.58 (t, \(J = 7.7\) Hz, 2H), 7.47 (d, \(J = 8.2\) Hz, 2H), 2.41 (s, 3H). \(^{13}\)C NMR (101 MHz, DMSO-\(d_6\)) \(\delta\): 171.98, 145.05, 135.67, 135.29, 132.59, 131.69, 130.52, 129.75, 128.10, 21.54.

![Figure S1. \(^1\)H NMR spectrum of 1a substrate](image)
Figure S2. $^{13}$C NMR spectrum of 1a substrate
(2) 4-methyl-N-[(2,4,6-trimethylphenyl)methylene]-benzenesulfonamide (1b)

Data of 1b: $^1$H NMR (400 MHz, DMSO-$d_6$) $\delta$: 9.28 (s, 1H), 7.81 (d, $J = 8.2$ Hz, 2H), 7.43 (d, $J = 8.3$ Hz, 2H), 6.99 (s, 2H), 2.41 (s, 6H), 2.37 (s, 3H), 2.25 (s, 3H). $^{13}$C NMR (151 MHz, DMSO-$d_6$) $\delta$: 170.08, 144.89, 144.78, 142.68, 135.72, 130.87, 130.45, 127.99, 126.20, 21.53, 21.52, 21.50. HRMS (M+Na$^+$) calculated for C$_{17}$H$_{19}$NNaO$_2$S 324.10287, found 324.10304.

Figure S3. $^1$H NMR spectrum of 1b substrate
Figure S4. $^{13}$C NMR spectrum of 1b substrate
(3) 4-methyl-N-[(3,4-dimethylphenyl)methylene]-benzenesulfonamide (1c)

Data of 1c: $^1$H NMR (400 MHz, DMSO-$d_6$) $\delta$: 9.00 (s, 1H), 7.85 – 7.63 (m, 4H), 7.42 (d, $J = 8.2$ Hz, 2H), 7.32 (t, $J = 7.3$ Hz, 1H), 2.35 (d, $J = 13.2$ Hz, 3H), 2.25 (d, $J = 16.4$ Hz, 6H). $^{13}$C NMR (101 MHz, DMSO-$d_6$) $\delta$ 171.63, 145.72, 144.87, 138.11, 135.54, 131.86, 130.83, 130.47, 130.33, 130.02, 128.00, 40.59, 40.39, 40.18, 39.97, 39.76, 39.55, 39.35, 21.52, 20.34, 19.53. HRMS (M+Na$^+$) calculated for C$_{16}$H$_{17}$NNaO$_2$S 310.08722

Figure S5. $^1$H NMR spectrum of 1c substrate
Figure S6. $^{13}$C NMR spectrum of 1c substrate
(4) 4-methyl-N-[(4-methylphenyl)methylene]-benzenesulfonamide (1d)

Data of 1d: Melting point \( ^\circ \text{C} \); 1H NMR (400 MHz, DMSO-\(d_6\)) \( \delta \): 9.09 (s, 1H), 7.88 (dd, \( J = 34.9 \text{ Hz}, 8.2 \text{ Hz} \), 4H), 7.43 (dd, \( J = 29.5 \text{ Hz}, 8.0 \text{ Hz} \), 4H), 2.41 (d, \( J = 2.2 \text{ Hz} \), 6H). 13C NMR (101 MHz, DMSO-\(d_6\)) \( \delta \): 171.59, 146.76, 144.92, 135.52, 131.80, 130.49, 130.40, 128.02, 21.93, 21.53.

Figure S7. \(^1\text{H NMR spectrum of 1d substrate}\)
Figure S8. $^{13}$C NMR spectrum of 1d substrate

(5) 4-methyl-N-[[4-(1-methylethyl)phenyl]methylene]-benzenesulfonamide (1e)

Data of 1e: Melting point [3]: 113-115 °C; $^1$H NMR (400 MHz, DMSO-$d_6$) $\delta$: 9.11 (s, 1H), 7.96 (d, $J = 8.2$ Hz, 2H), 7.84 (d, $J = 8.2$ Hz, 2H), 7.46 (dd, $J = 8.0$, 5.1 Hz, 4H), 2.98 (dt, $J = 13.8$, 6.9 Hz, 1H), 2.39 (d, $J = 12.5$ Hz, 3H), 1.21 (d, $J = 6.9$ Hz, 6H). $^{13}$C NMR (101 MHz, DMSO-$d_6$) $\delta$: 171.63, 157.13, 144.87, 135.55, 131.99, 130.46, 127.98, 127.80, 40.60, 40.39, 40.18, 39.97, 39.76, 39.56, 39.35, 34.19, 23.76, 21.51.
Figure S9. $^1$H NMR spectrum of 1e substrate

Figure S10. $^{13}$C NMR spectrum of 1e substrate
2. $^1$H NMR and $^{13}$C NMR spectra of 5 products

(1) $N$-[cyano(phenyl)methyl]-4-methyl-benzenesulfonamide (2a)

Data of 2a: Melting point $^{[4]}$, 153-154 °C; $^1$H NMR (500 MHz, DMSO-$d_6$) $\delta$: 9.22 (d, $J = 9.3$ Hz, 1H), 7.75 (d, $J = 8.2$ Hz, 2H), 7.40 (dt, $J = 9.3$ Hz, 7H), 5.84 (d, $J = 9.3$ Hz, 1H), 2.40 (s, 3H). $^{13}$C NMR (101 MHz, DMSO-$d_6$) $\delta$: 143.79, 137.67, 134.37, 130.13, 129.44, 129.35, 127.41, 118.17, 47.47, 21.47.

Figure S11. $^1$H NMR spectrum of 2a product
Figure S12. $^{13}$C NMR spectrum of 2a product
(2) \(N\)-cyano(2,4,6-trimethylphenyl)methyl]-4-methyl-benzenesulfonamide (2b)

Data of 2b: Melting point \(^{[4]}\), 139-140 °C; \(^1\)H NMR (400 MHz, DMSO-\(d_6\)) \(\delta\): 8.71 (s, 1H), 7.75 (d, \(J = 7.4\) Hz, 2H), 7.39 (d, \(J = 7.7\) Hz, 2H), 6.83 (s, 2H), 5.48 (s, 1H), 2.37 (s, 3H), 2.17 (d, \(J = 10.2\) Hz, 9H). \(^{13}\)C NMR (101 MHz, DMSO-\(d_6\)) \(\delta\): 143.88, 139.18, 137.35, 137.09, 130.23, 130.08, 127.41, 126.90, 118.27, 42.83, 40.58, 40.37, 40.16, 39.95, 39.74, 39.53, 39.32, 21.45, 20.82, 19.98.

Figure S13. \(^1\)H NMR spectrum of 2b product
Figure S14. $^{13}$C NMR spectrum of 2b product
(3) N-[cyano(3,4-dimethylphenyl)methyl]-4-methyl-benzenesulfonamide (2c)

Data of 2c: $^1$H NMR (400 MHz, DMSO-$d_6$) $\delta$: 9.09 (s, 1H), 7.70 (d, $J = 7.3$ Hz, 2H), 7.36 (d, $J = 7.7$ Hz, 2H), 7.28-6.91 (m, 3H), 5.66 (s, 1H), 2.36 (s, 3H), 2.15 (d, $J = 9.0$ Hz, 6H). $^{13}$C NMR (101 MHz, DMSO-$d_6$) $\delta$: 143.67, 137.76, 137.67, 137.20, 131.55, 130.23, 130.02, 128.42, 127.17, 124.80, 118.29, 47.30, 40.58, 40.37, 40.16, 39.95, 39.74, 39.53, 39.33, 21.43, 19.75, 19.45. HRMS (M+Na$^+$) calculated for C$_{17}$H$_{18}$N$_2$NaO$_2$S 337.09812, found 337.09838.

Figure S15. $^1$H NMR spectrum of 2c product
Figure S16. $^{13}$C NMR spectrum of 2c product
(4) N-[cyano(p-tolyl)methyl]-4-methyl-benzenesulfonamide (2d)

Data of 2d: Melting point \[^{[4]}\]: 140-142 °C; \(^1\)H NMR (600 MHz, DMSO-d\(_6\)) \(\delta: 9.16 (s, 1H), 7.73 (dd, J = 17.8 \text{ Hz}, 8.0 \text{ Hz}, 2H), 7.49-7.01 (m, 6H), 5.76 (s, 1H), 2.46-2.17 (m, 6H). \(^{13}\)C NMR (151 MHz, DMSO-d\(_6\)) \(\delta: 143.75, 138.97, 137.70, 131.42, 130.11, 129.82, 127.35, 127.16, 118.28, 47.25, 31.1, 21.46, 21.10.

![Figure S17. \(^1\)H NMR spectrum of 2d product](image-url)
Figure S18. $^{13}$C NMR spectrum of 2d product
(5) N-{cyano[4-(1-methylethyl)phenyl]methyl}-4-methyl-benzensulfonamide (2e)

![Image of chemical structure]

**Data of 2e:** $^1$H NMR (400 MHz, DMSO-d$_6$) $\delta$: 9.17 (s, 1H), 7.73 (d, $J = 7.5$ Hz, 2H), 7.38 (d, $J = 7.7$ Hz, 2H), 7.27 (dd, $J = 16.0$, 7.8 Hz, 3H), 5.75 (d, $J = 3.2$ Hz, 1H), 3.35 (s, 1H), 2.87 (dt, $J = 13.5$, 6.8 Hz, 1H), 2.38 (s, 2H), 1.17 (d, $J = 6.8$ Hz, 5H). $^{13}$C NMR (101 MHz, DMSO-d$_6$) $\delta$: 149.85, 143.64, 137.75, 131.67, 130.05, 127.50, 127.20, 127.14, 118.26, 47.32, 40.58, 40.37, 40.16, 39.95, 39.74, 39.53, 39.33, 33.58, 24.14, 21.44. HRMS (M+Na$^+$) calculated for C$_{18}$H$_{20}$N$_2$NaO$_2$S 351.11377, found 351.11382.

**Figure S19.** $^1$H NMR spectrum of 2e product
Figure S20. $^{13}$C NMR spectrum of 2e product
3. High resolution mass spectrum for some imines substrates and products

Figure S21. High resolution mass spectrum for substrate 1b

Figure S22. High resolution mass spectrum for substrate 1c

Figure S23. High resolution mass spectrum for product 2c
Figure S24 High resolution mass spectrum for product 2e

Reference