Electronic Supplementary Material (ESI) for New Journal of Chemistry.

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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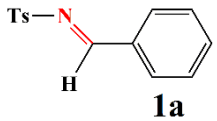
1. Spectral data for imines precursors and α-aminonitriles

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**)



1a

Data of 1a: Melting point [1], 109-111 °C; ^{1}H NMR (400 MHz, DMSO-d_6) δ: 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) δ: 171.98, 145.05, 135.67, 135.29, 132.59, 131.69, 130.52, 129.75, 128.10, 21.54.

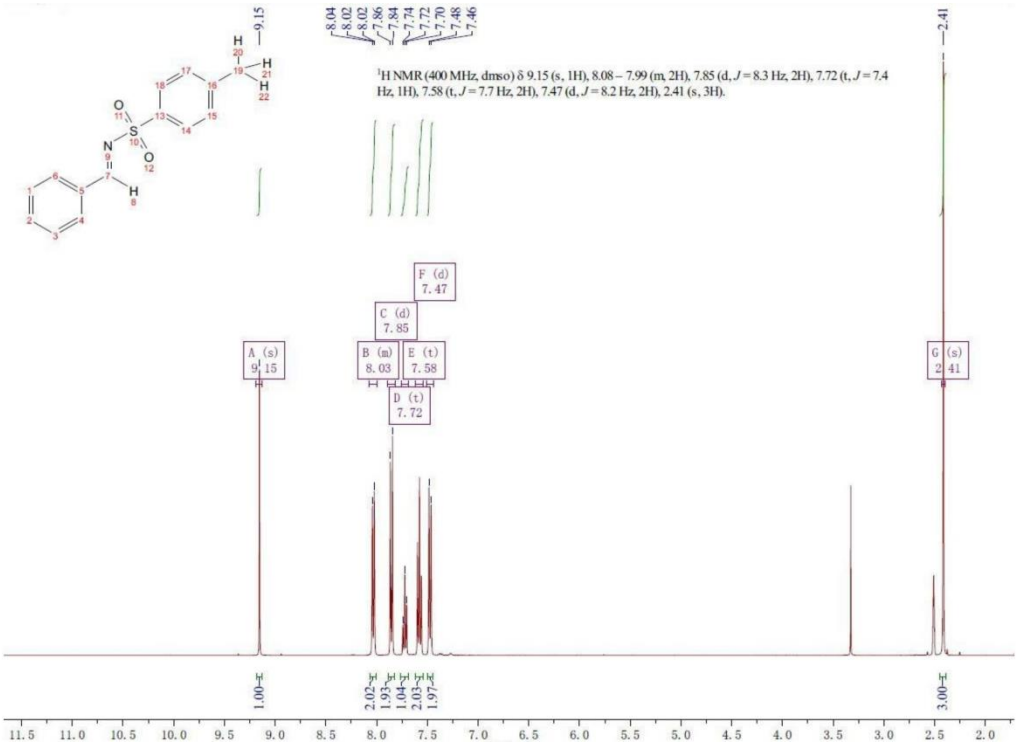


Figure S1. ^{1}H NMR spectrum of 1a substrate

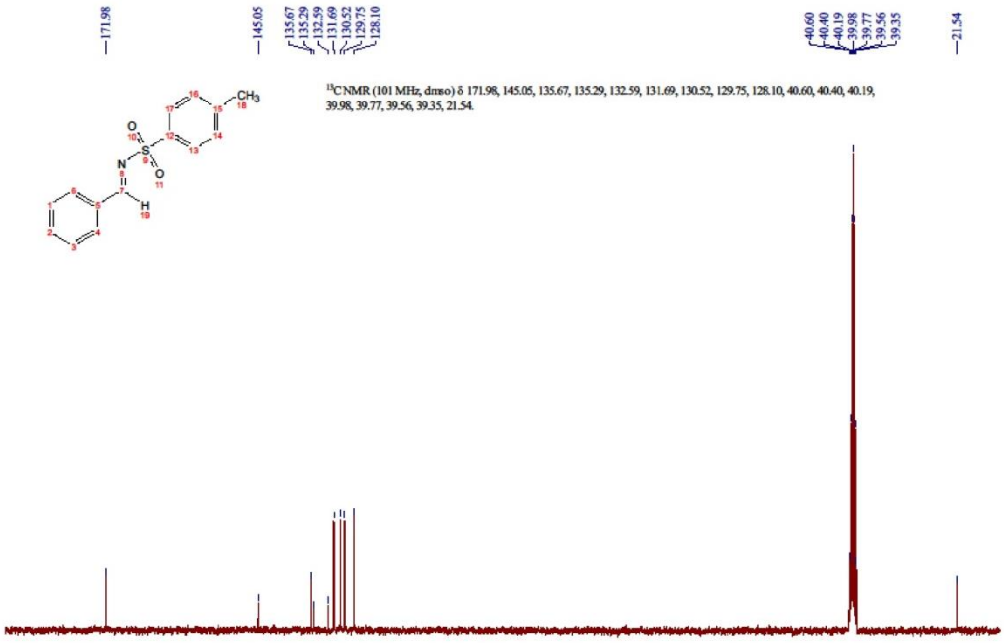
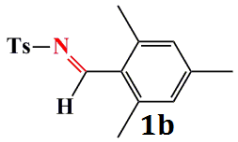


Figure S2. ^{13}C NMR spectrum of 1a substrate

(2) *4-methyl-N-[(2,4,6-trimethylphenyl)methylene]-benzenesulfonamide* (**1b**)



Data of 1b: ^{1}H NMR (400 MHz, DMSO-d_6) δ: 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) δ: 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 $C_{17}H_{19}NNaO_2S$ 324.10287, found 324.10304.

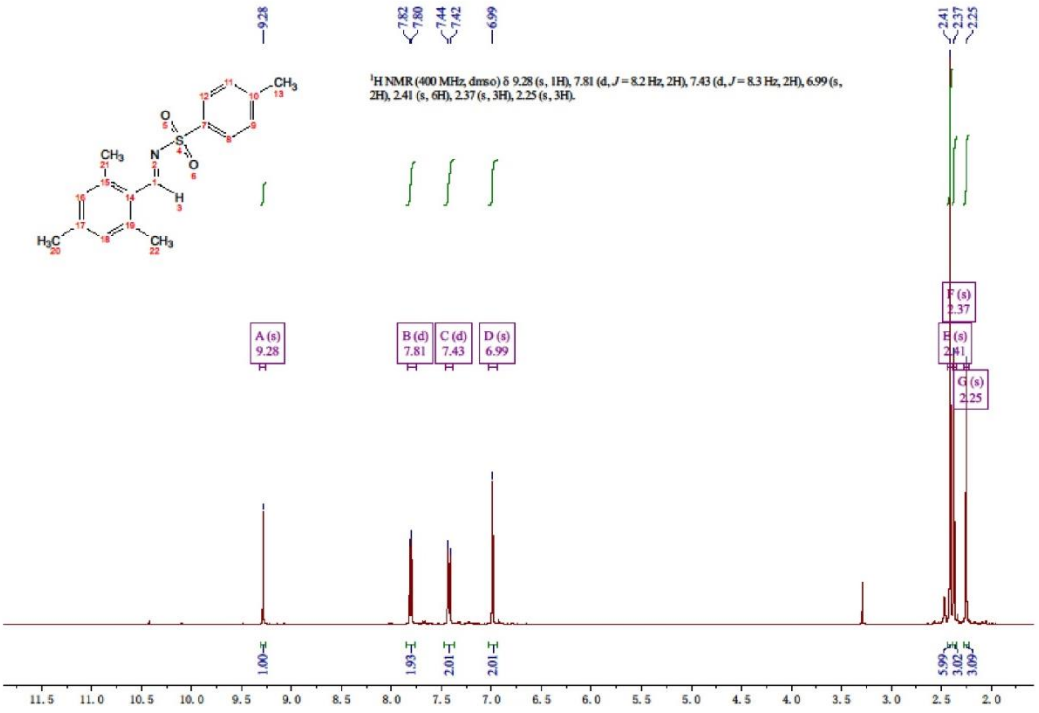


Figure S3. ^{1}H NMR spectrum of 1b substrate

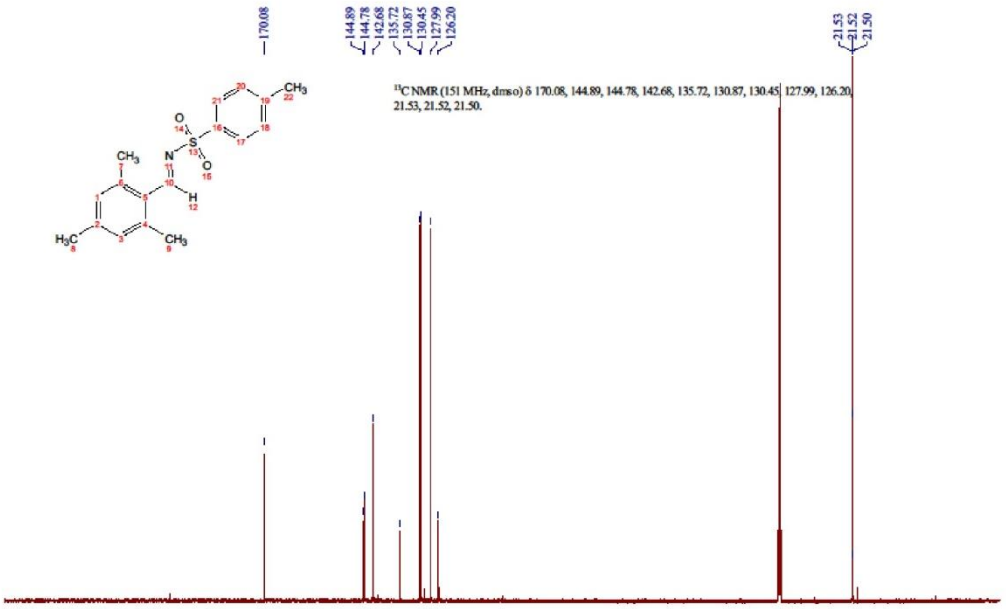
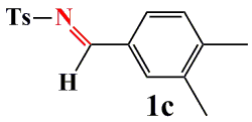


Figure S4. ^{13}C NMR spectrum of 1b substrate

(3) *4-methyl-N-[(3,4-dimethylphenyl)methylene]-benzenesulfonamide* (**1c**)



1c

Data of 1c: ^{1}H NMR (400 MHz, DMSO-d_6) δ: 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) δ 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 $C_{16}H_{17}NNaO_2S$ 310.08722

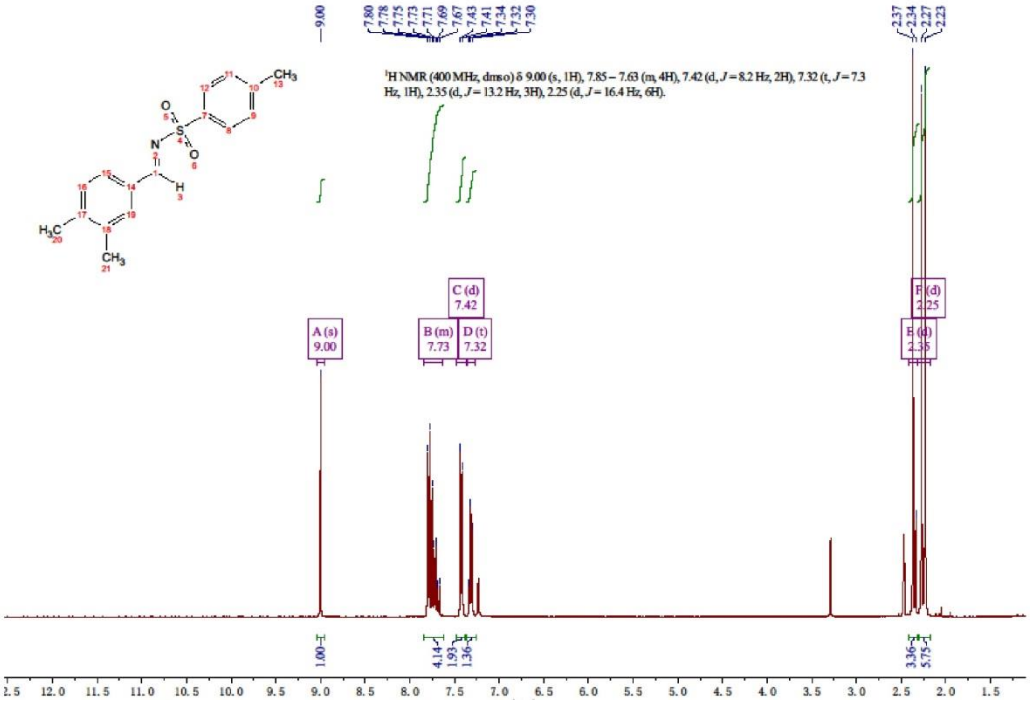


Figure S5. ^{1}H NMR spectrum of 1c substrate

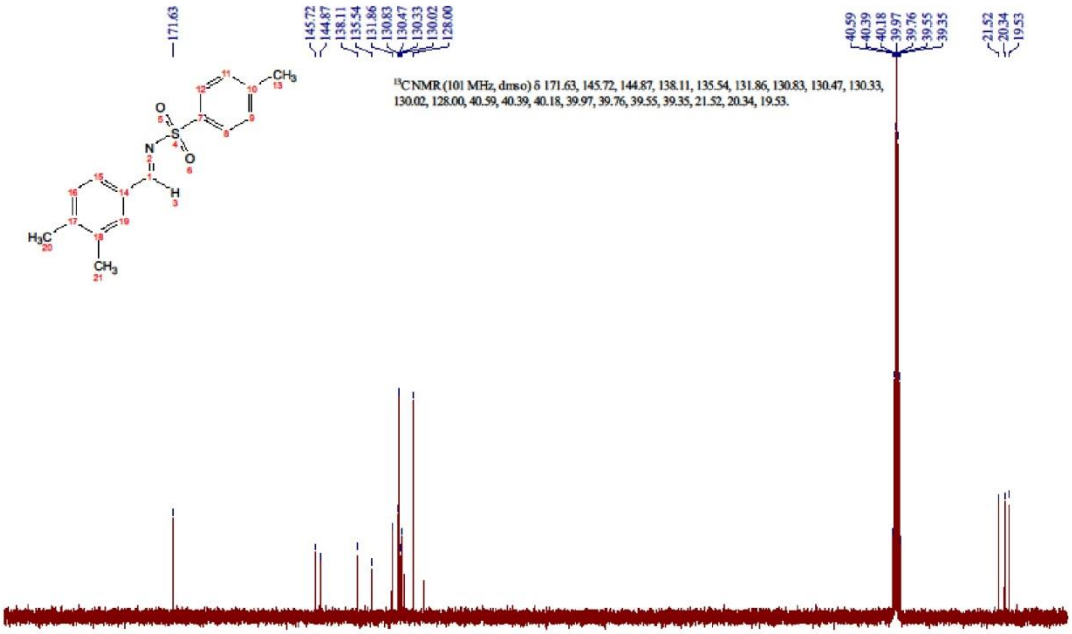
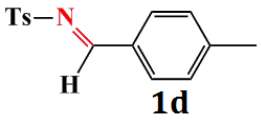


Figure S6. ^{13}C NMR spectrum of 1c substrate

(4) *4-methyl-N-[(4-methylphenyl)methylene]-benzenesulfonamide* (**1d**)



1d

Data of 1d: Melting point [2], 114-115 °C; 1H NMR (400 MHz, DMSO-d_6) δ: 9.09 (s, 1H), 7.88 (dd, *J* = 34.9 Hz, 8.2Hz, 4H), 7.43 (dd, *J* = 29.5 Hz, 8.0 Hz, 4H), 2.41 (d, *J* = 2.2 Hz, 6H). ^{13}C NMR (101 MHz, DMSO-d_6) δ: 171.59, 146.76, 144.92, 135.52, 131.80, 130.49, 130.40, 128.02, 21.93, 21.53.

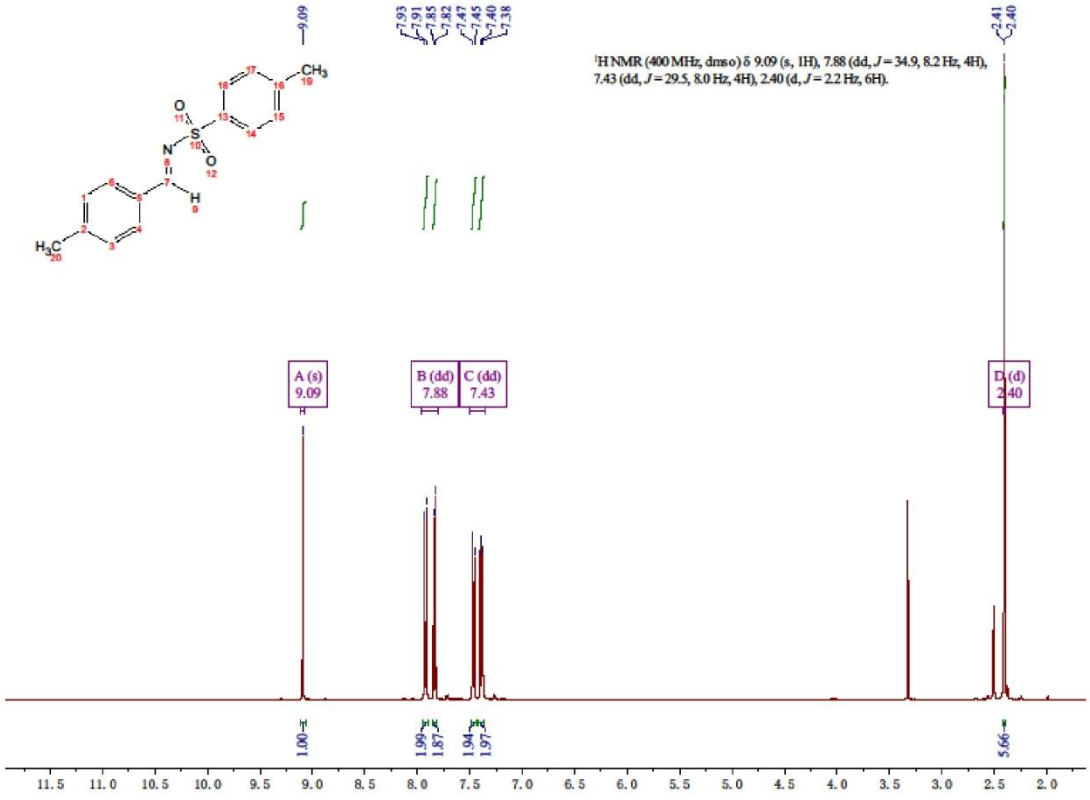


Figure S7. ^{1}H NMR spectrum of 1d substrate

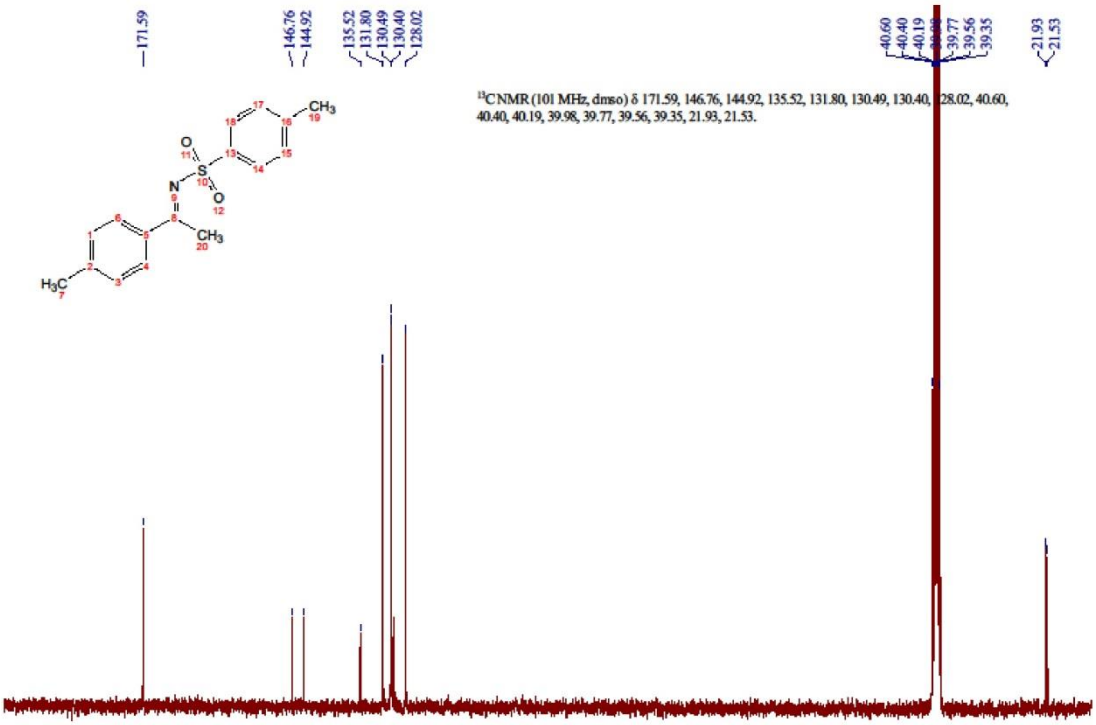
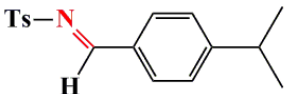


Figure S8. ^{13}C NMR spectrum of 1d substrate

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



1e

Data of 1e: Melting point [3], 113-115 °C; ^{1}H NMR (400 MHz, DMSO-d_6) δ: 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) δ: 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.

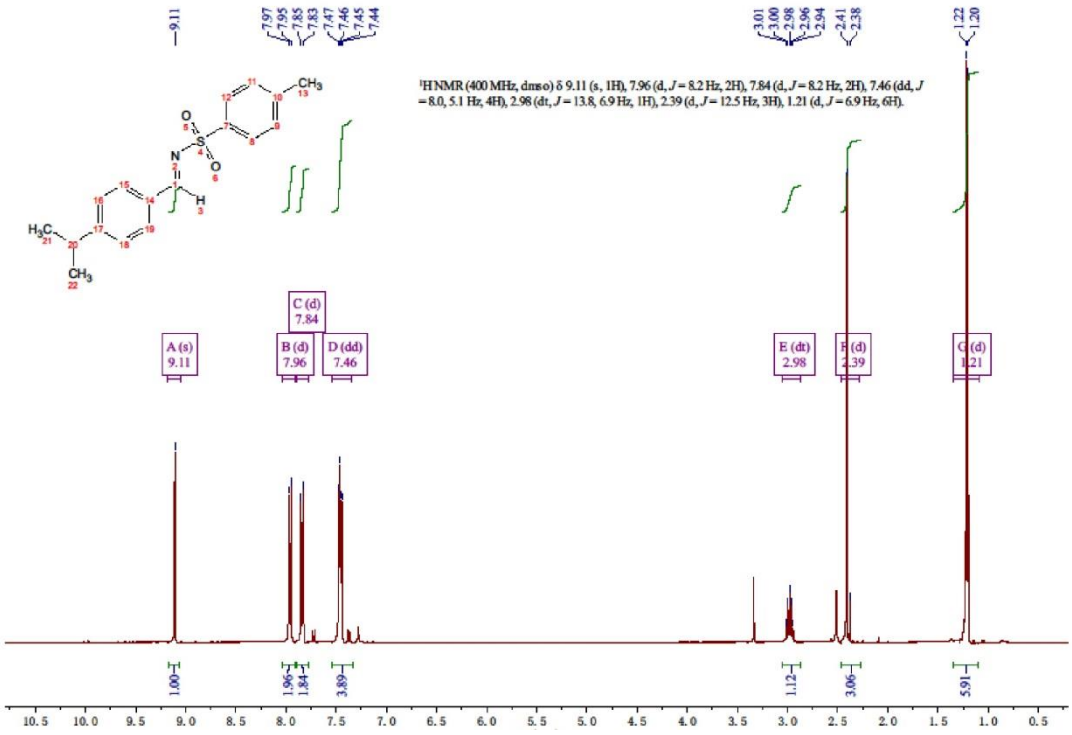


Figure S9. ^{1}H NMR spectrum of 1e substrate

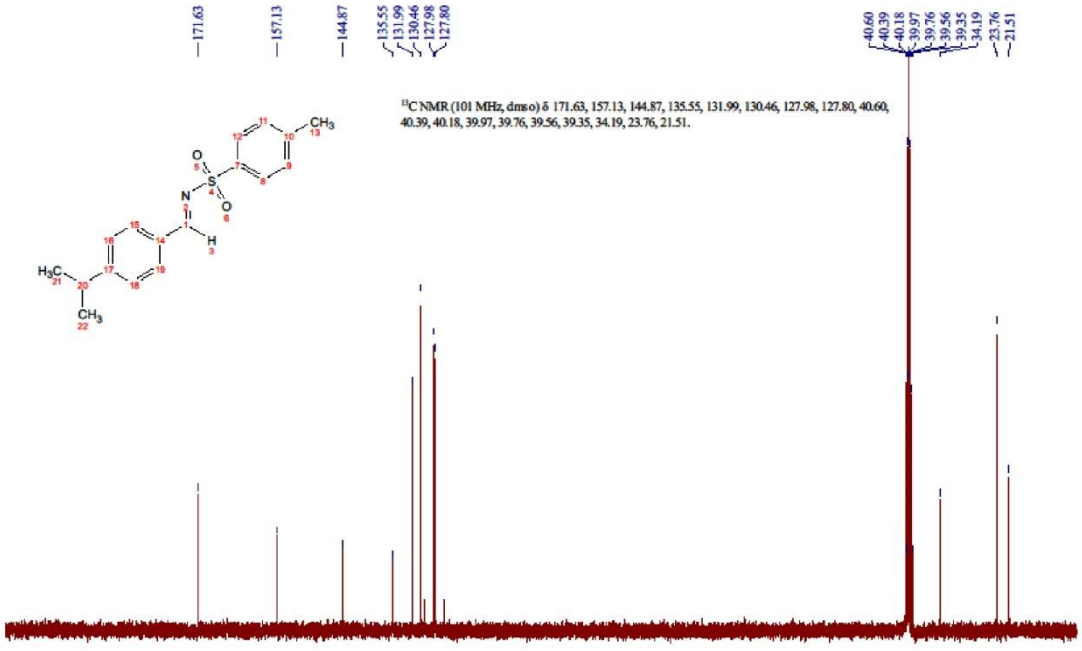
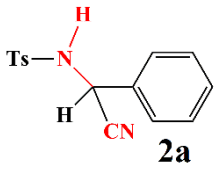


Figure S10. ^{13}C NMR spectrum of 1e substrate

2. ^{1}H NMR and ^{13}C NMR spectra of 5 products

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



2a

Data of 2a: Melting point [4], 153-154 ºC; ^{1}H NMR (500 MHz, DMSO-d_6) δ: 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) δ: 143.79, 137.67, 134.37, 130.13, 129.44, 129.35, 127.41, 118.17, 47.47, 21.47.

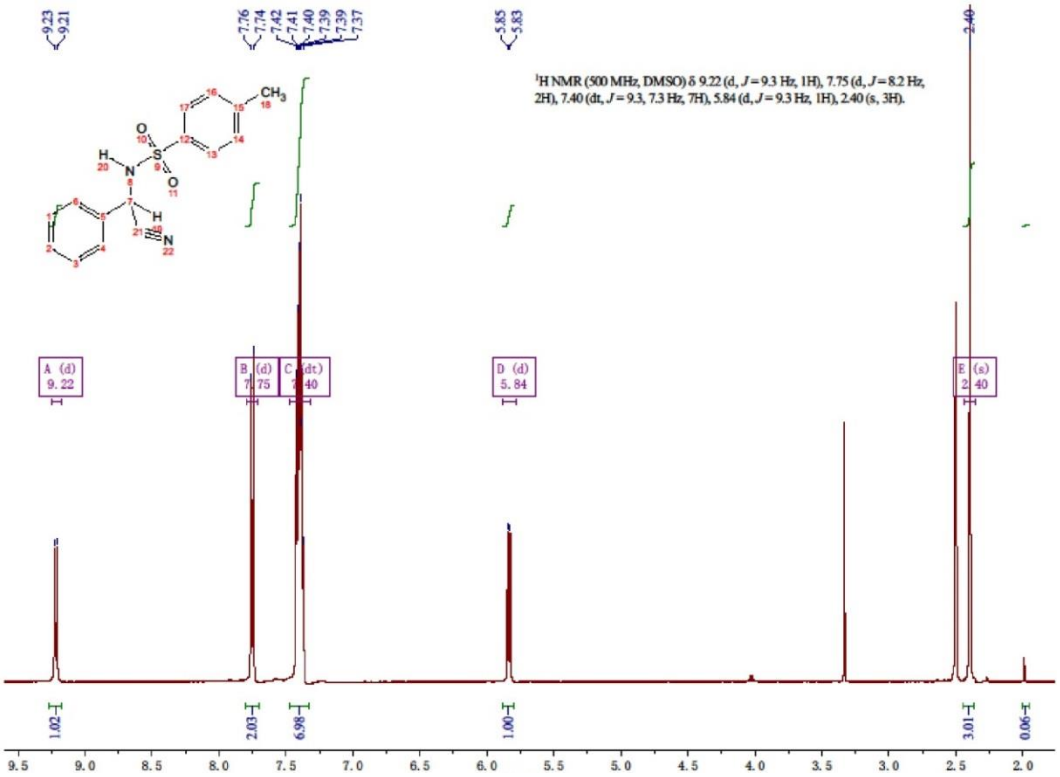


Figure S11. ^{1}H NMR spectrum of 2a product

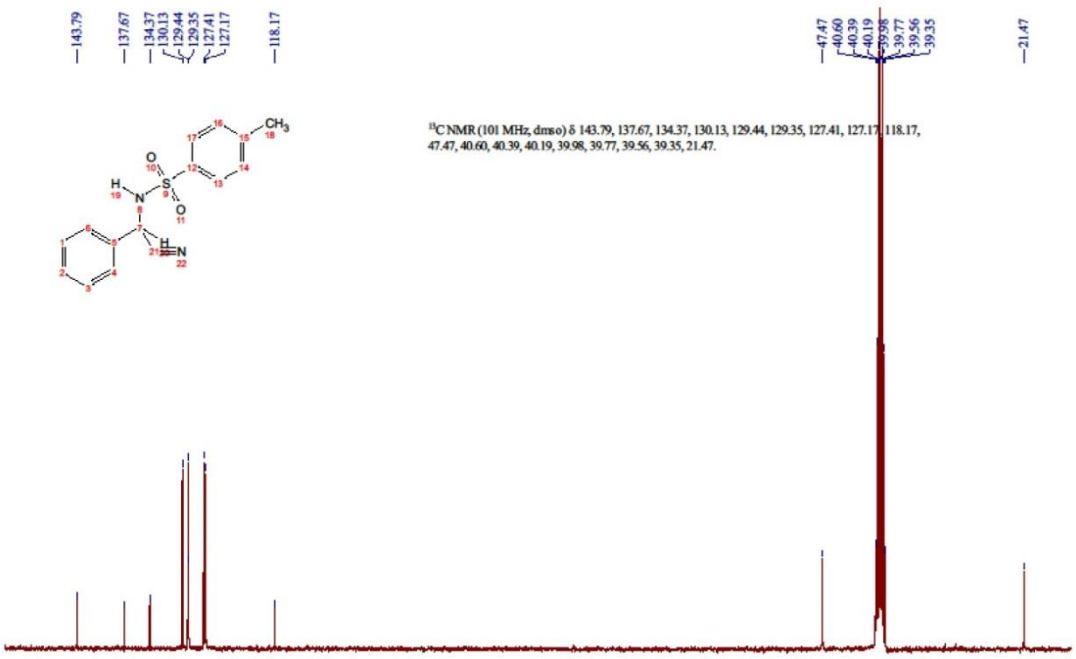
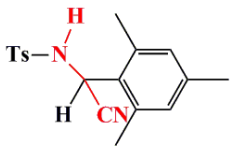


Figure S12. ^{13}C NMR spectrum of 2a product

(2) *N-[cyano(2,4,6-trimethylphenyl)methyl]-4-methyl-benzenesulfonamide* (**2b**)



2b

Data of 2b: Melting point [4], 139-140 °C; ^{1}H NMR (400 MHz, DMSO-d_6) δ: 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) δ: 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.

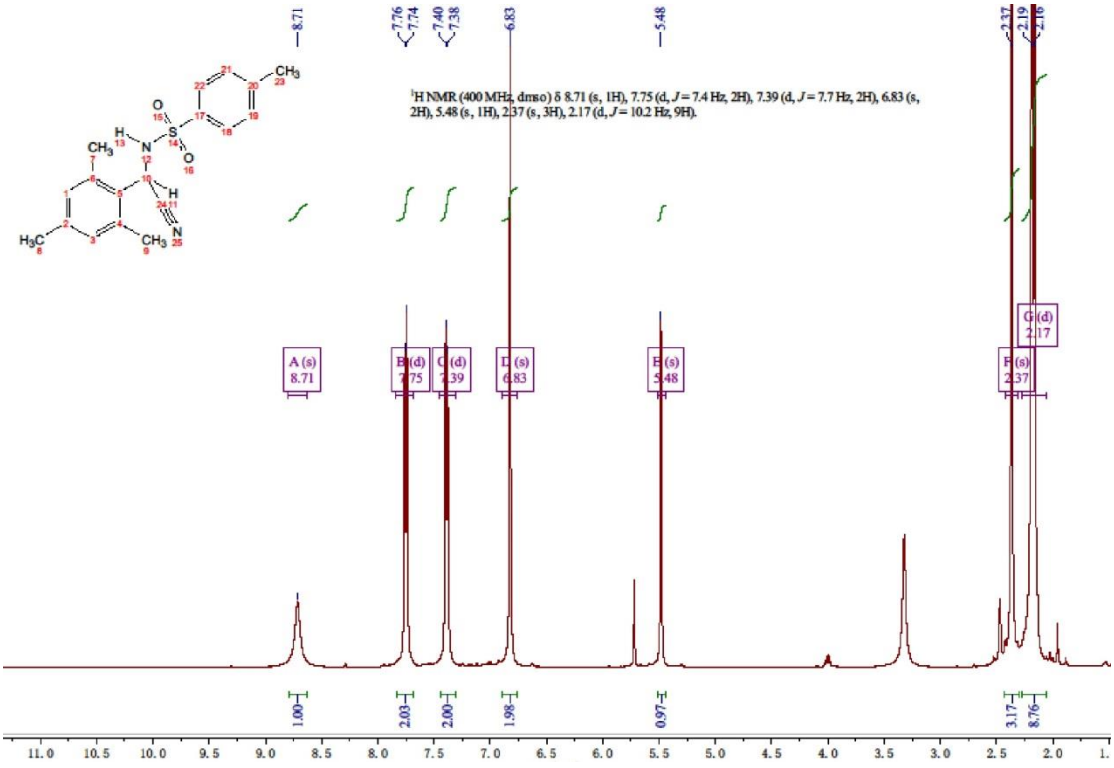


Figure S13. ^{1}H NMR spectrum of 2b product

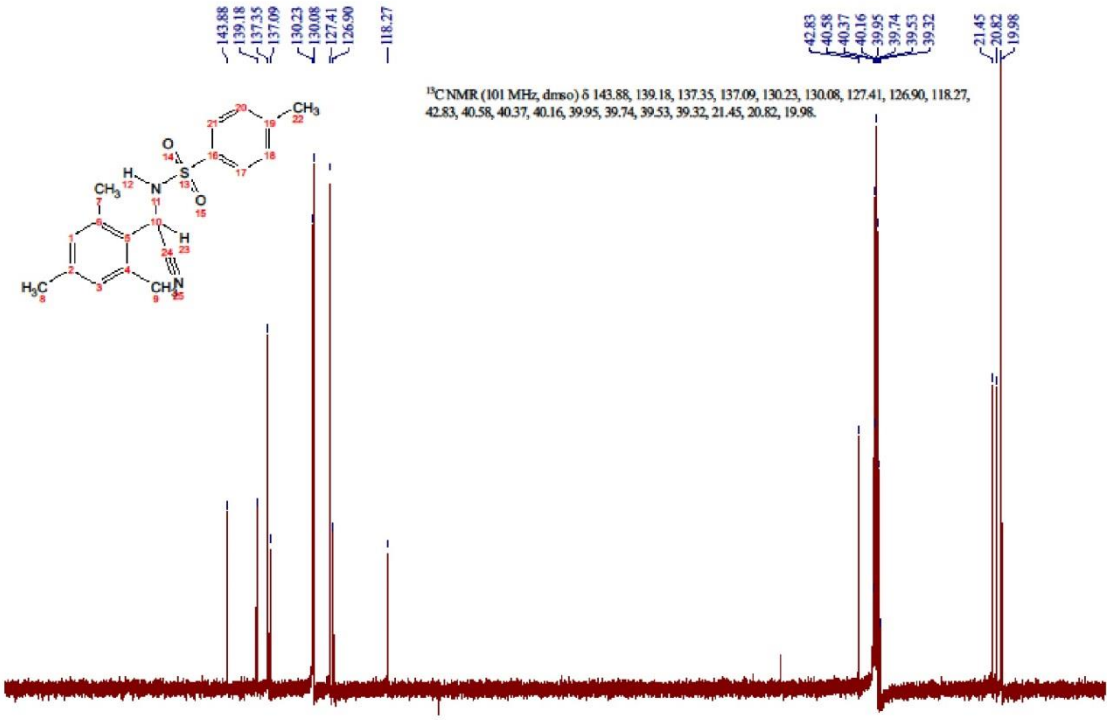
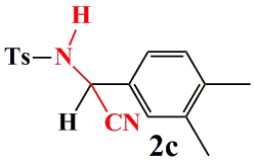


Figure S14. ^{13}C NMR spectrum of 2b product

(3) *N-[cyano(3,4-dimethylphenyl)methyl]-4-methyl-benzenesulfonamide* (**2c**)



2c

Data of 2c: ^{1}H NMR (400 MHz, DMSO-d_6) δ: 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) δ: 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 $C_{17}H_{18}N_2NaO_2S$ 337.09812, found 337.09838.

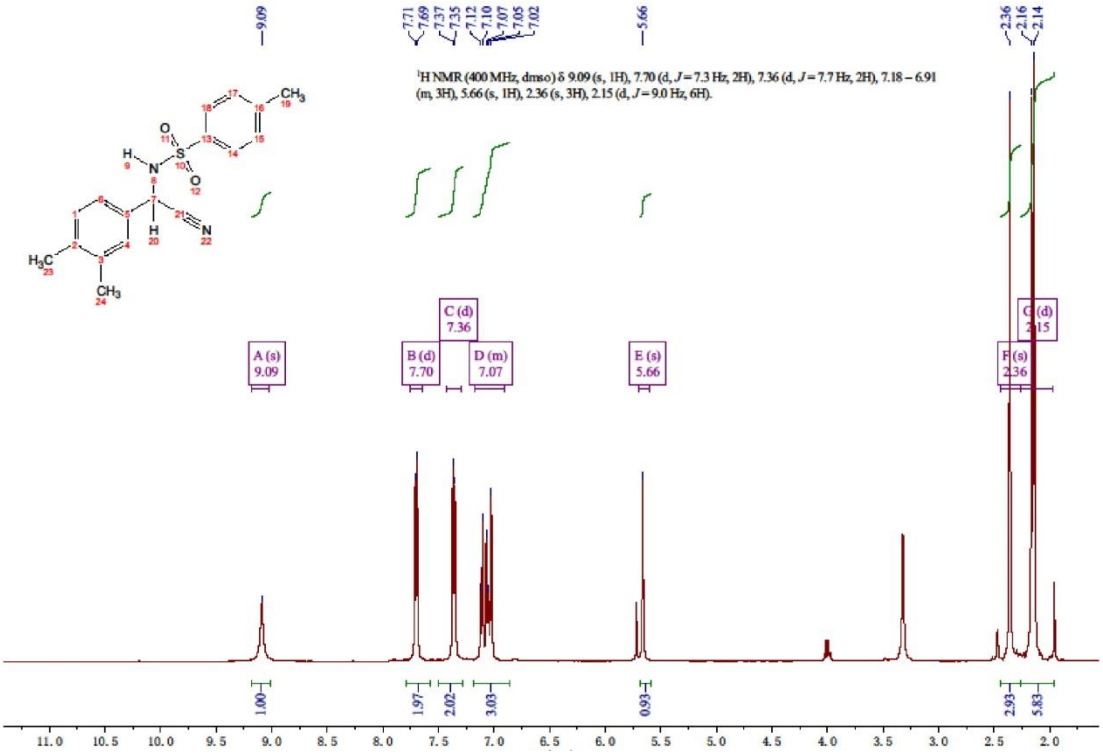


Figure S15. ^{1}H NMR spectrum of 2c product

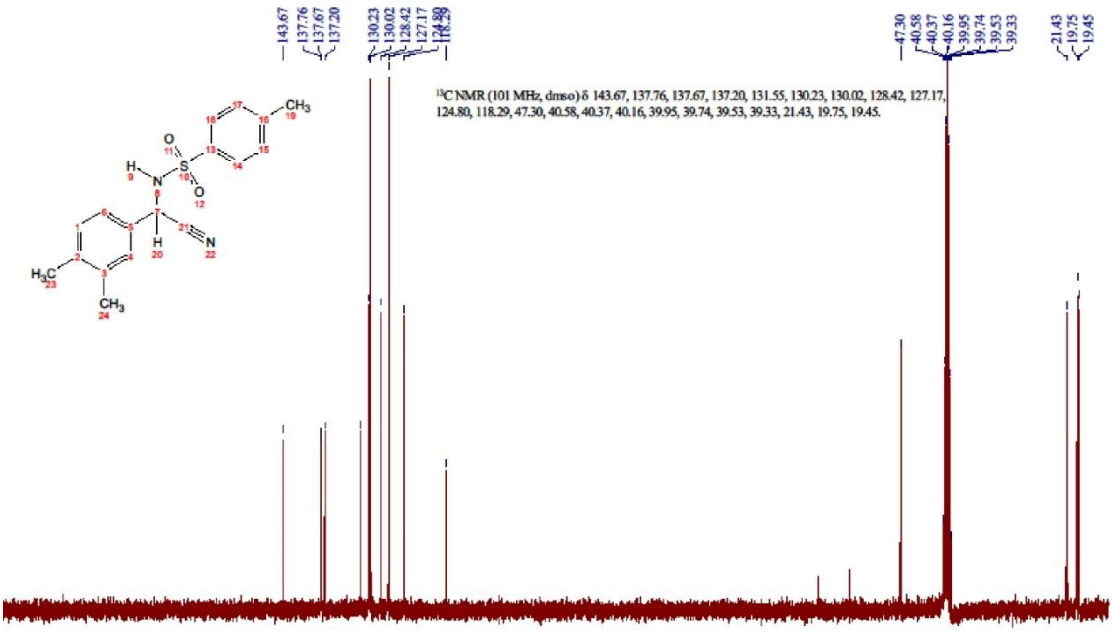
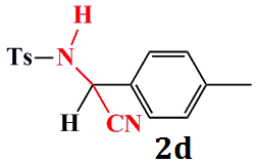


Figure S16. ^{13}C NMR spectrum of 2c product

(4) *N-[cyano(p-tolyl)methyl]-4-methyl-benzenesulfonamide* (**2d**)



2d

Data of 2d: Melting point [4], 140-142 °C; ^{1}H NMR (600 MHz, DMSO-d_6) δ: 9.16 (s, 1H), 7.73 (dd, *J* = 17.8 Hz, 8.0Hz, 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) δ: 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.

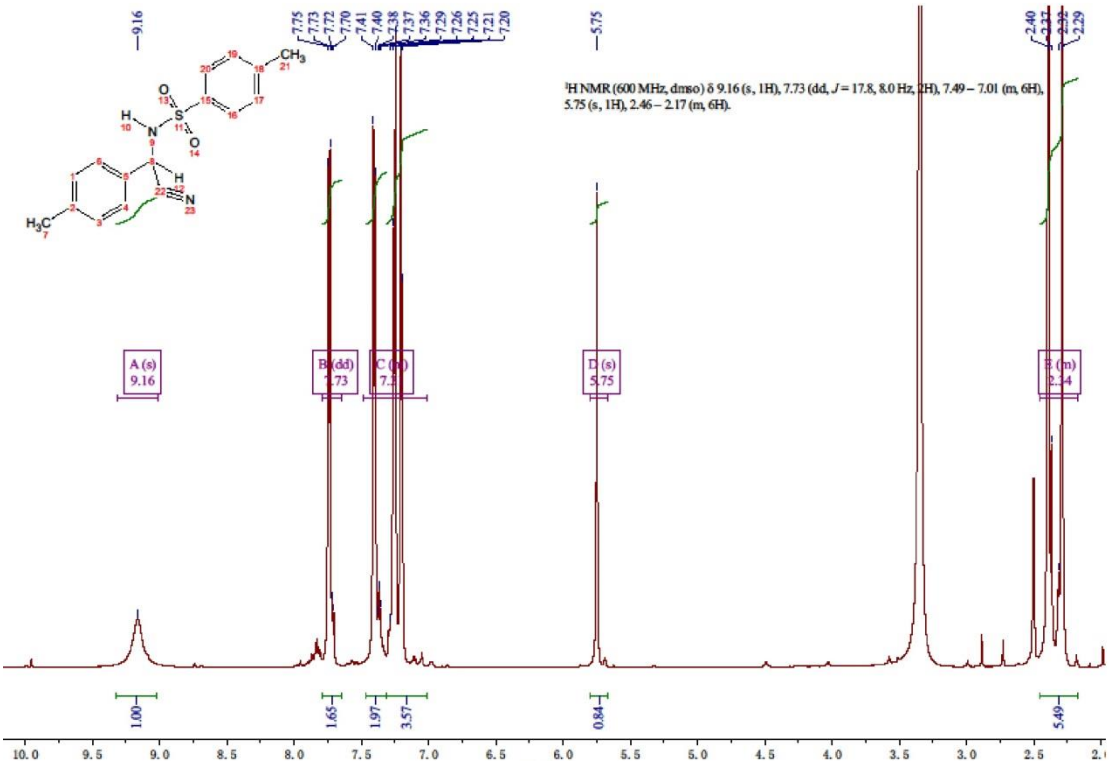


Figure S17. ^{1}H NMR spectrum of 2d product

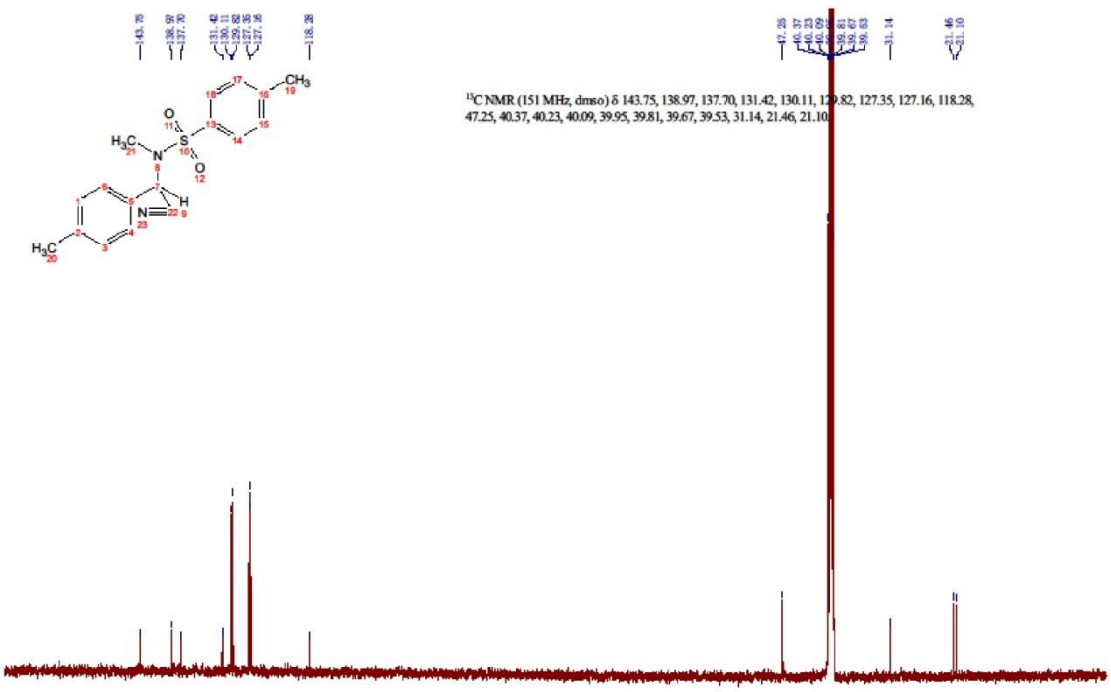
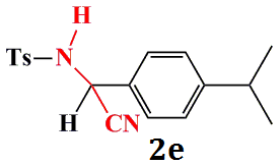


Figure S18. ^{13}C NMR spectrum of 2d product

(5) *N-{cyano[4-(1-methylethyl)phenyl]methyl}-4-methyl-benzenesulfonamide* (**2e**)



2e

Data of 2e: ^{1}H NMR (400 MHz, DMSO-d_6) δ: 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) δ: 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 $C_{18}H_{20}N_2NaO_2S$ 351.11377, found 351.11382

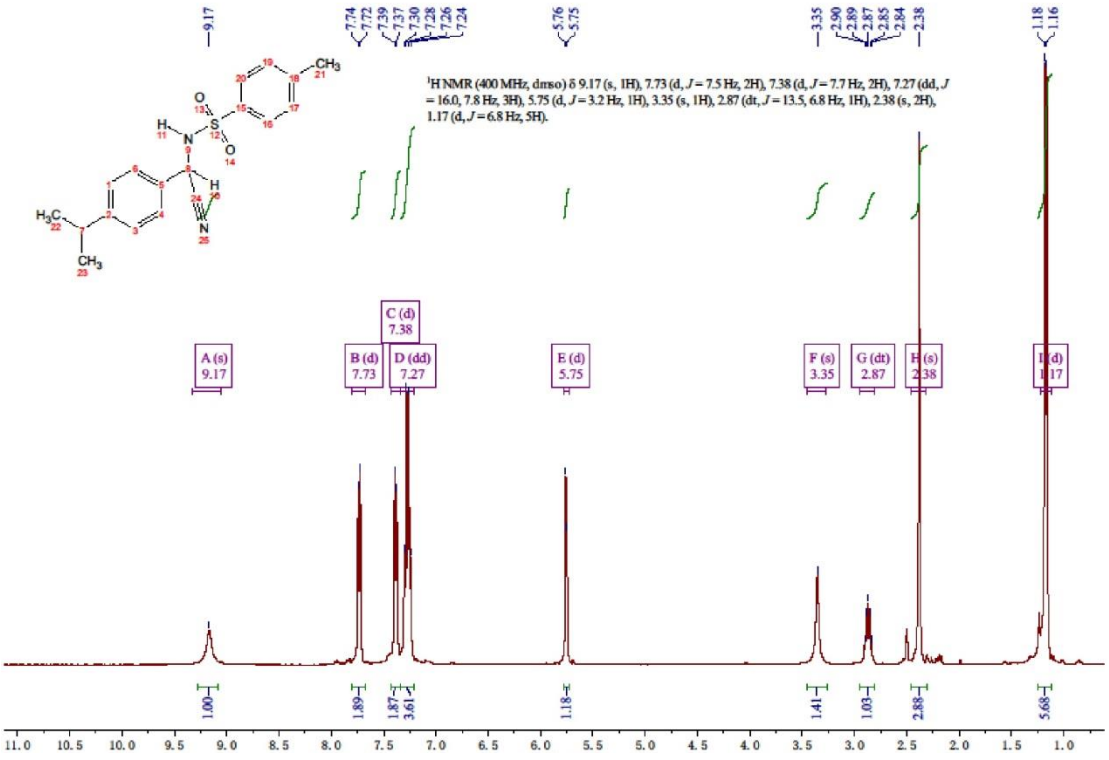


Figure S19. ^{1}H NMR spectrum of 2e product

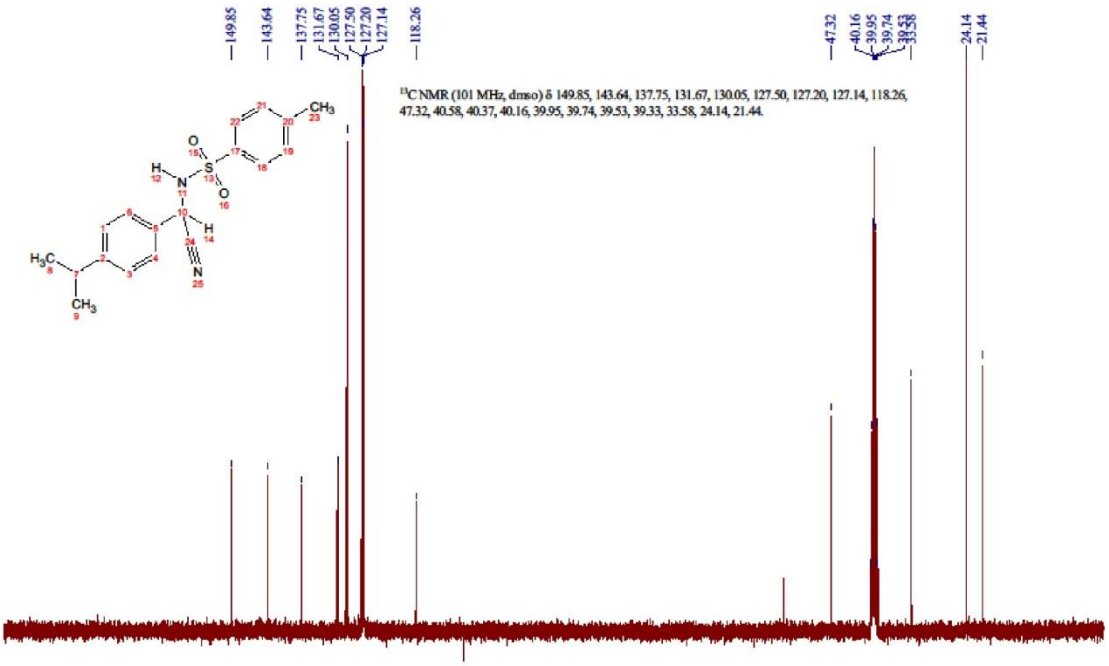
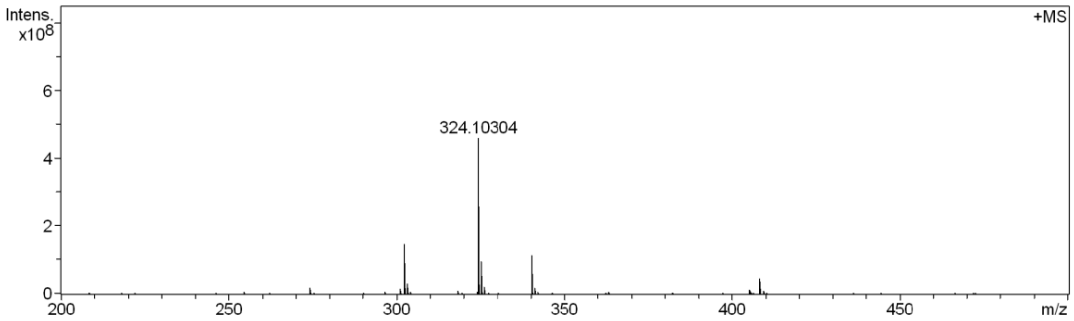


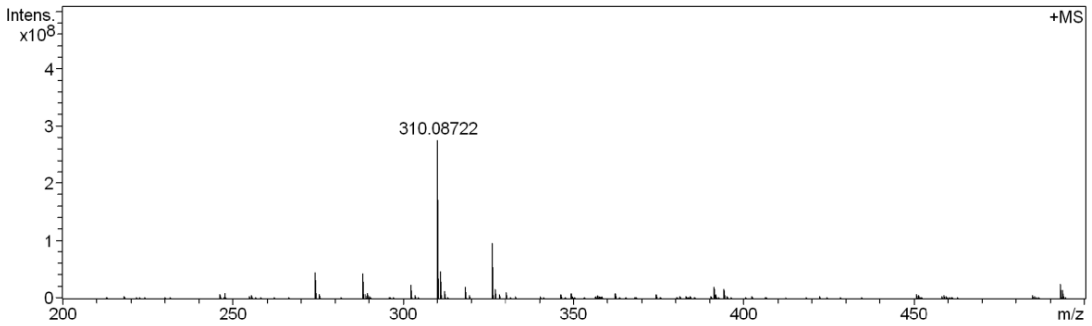
Figure S20. ^{13}C NMR spectrum of 2e product

3. High resolution mass spectrum for some imines substrates and products



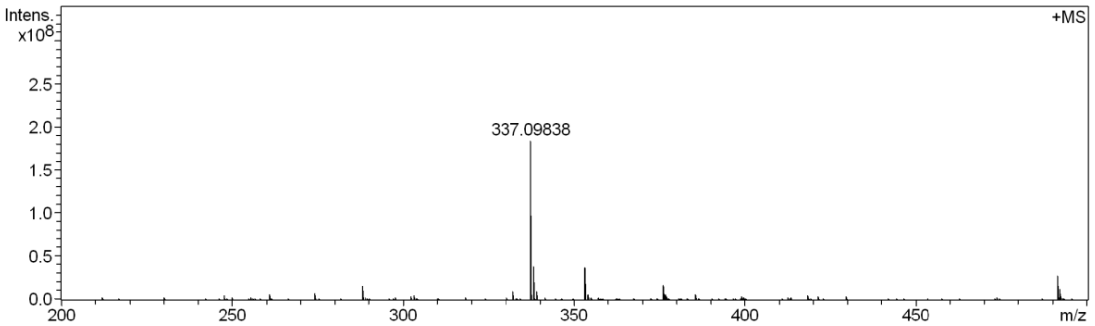
Meas. m/z	#	Formula	Score	m/z	err [ppm]	Mean err [ppm]	mSigma	rdb	e⁻ Conf	N-Rule
324.10304	1	C 17 H 19 N Na O 2 S	100.00	324.10287	-0.52	-0.73	11.7	8.5	even	ok

Figure S21. High resolution mass spectrum for substrate 1b



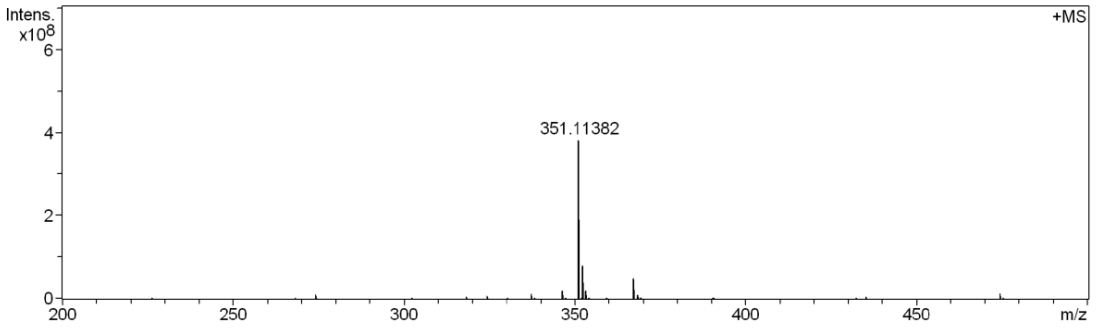
Meas. m/z	#	Formula	Score	m/z	err [ppm]	Mean err [ppm]	mSigma	rdb	e⁻ Conf	N-Rule
310.08722	1	C 16 H 17 N Na O 2 S	100.00	310.08722	0.01	-0.10	11.3	8.5	even	ok

Figure S22. High resolution mass spectrum for substrate 1c



Meas. m/z	#	Formula	Score	m/z	err [ppm]	Mean err [ppm]	mSigma	rdb	e⁻ Conf	N-Rule
337.09838	1	C 17 H 18 N 2 Na O 2 S	100.00	337.09812	-0.79	-0.82	9.4	9.5	even	ok

Figure S23. High resolution mass spectrum for product 2c



Meas. m/z	#	Formula	Score	m/z	err [ppm]	Mean err [ppm]	mSig ma	rdb	e⁻ Conf	N-R ule
351.11382	1	C 18 H 20 N 2 Na O 2 S	100.00	351.11377	-0.13	-0.44	8.2	9.5	even	ok

Figure S24 High resolution mass spectrum for product 2e

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