

Supporting Information

Dendrimer crown ether tethered Multi-Wall Carbon Nanotubes support methyltrioxorhenium in the selective oxidation of olefins to epoxides

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SI#1 FT-IR of intermediates II A-D, III A-D and catalysts IV A-D and V E-H

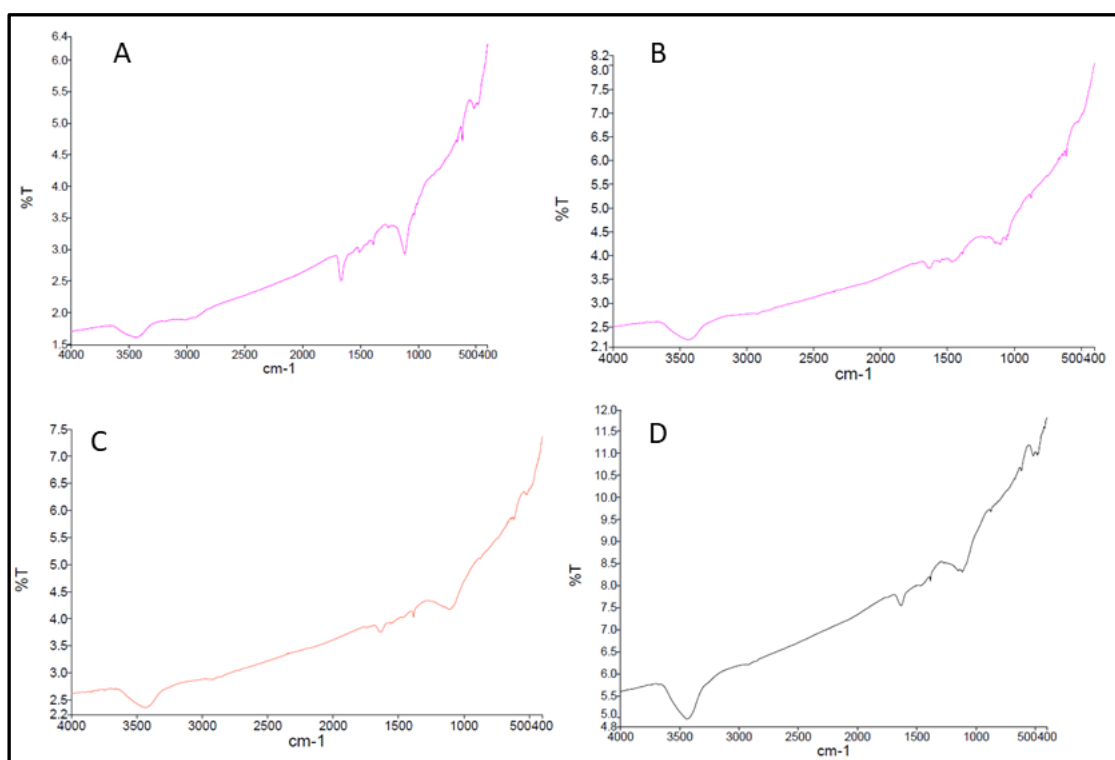


Figure S1. FT-IR of intermediates II A-D. Panel A: intermediate II A. Panel B: intermediates II B. Panel C : intermediates II C. Panel D: intermediates II D.

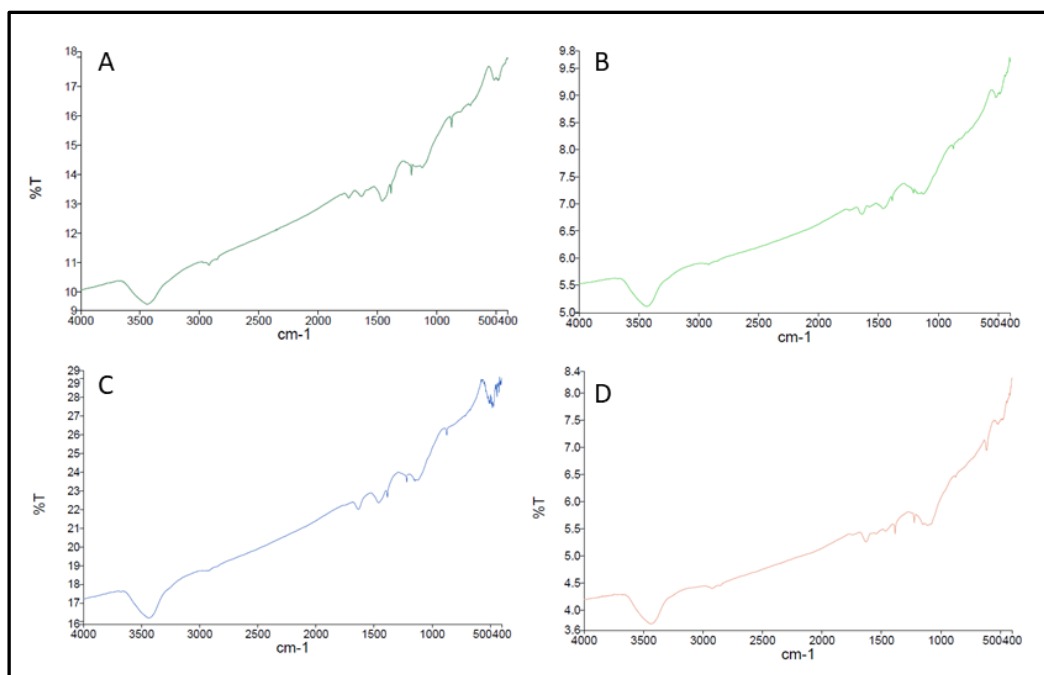


Figure S2. FT-IR of intermediates **III A-D**. Panel A: intermediate **III A**. Panel B: intermediates **III B**. Panel C: intermediates **III C**. Panel D: intermediates **III D**.

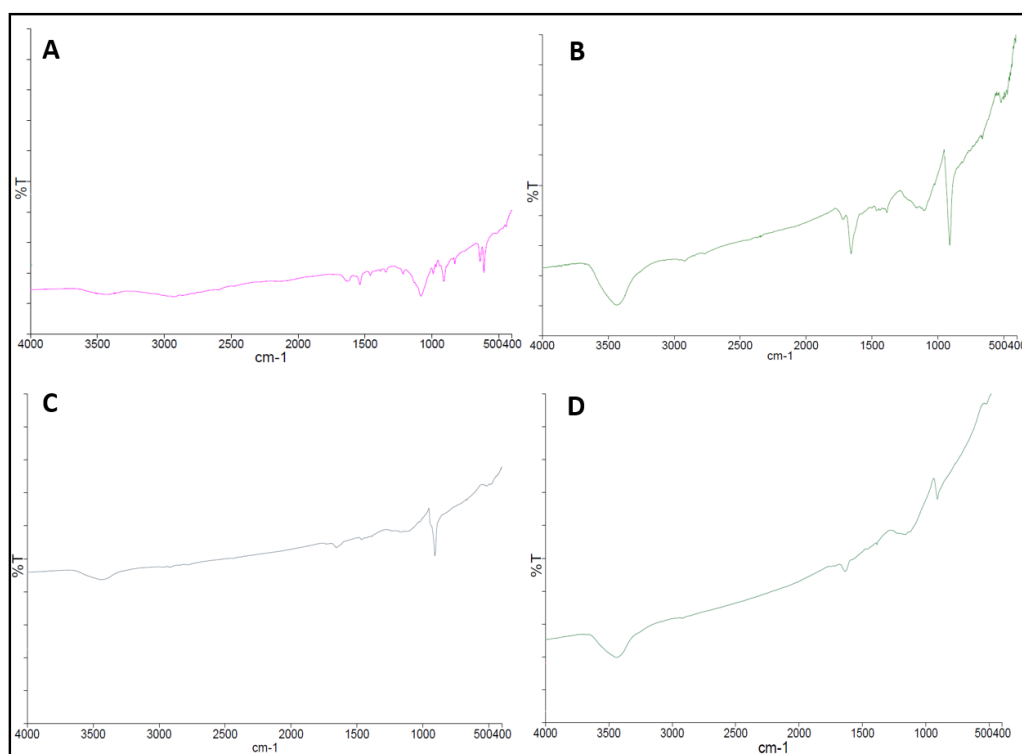


Figure S3. FT-IR of catalysts **IV A-D**. Panel A: catalyst **IV A**. Panel B: catalyst **IV B**. Panel C: catalyst **IV C**. Panel D: catalyst **IV D**.

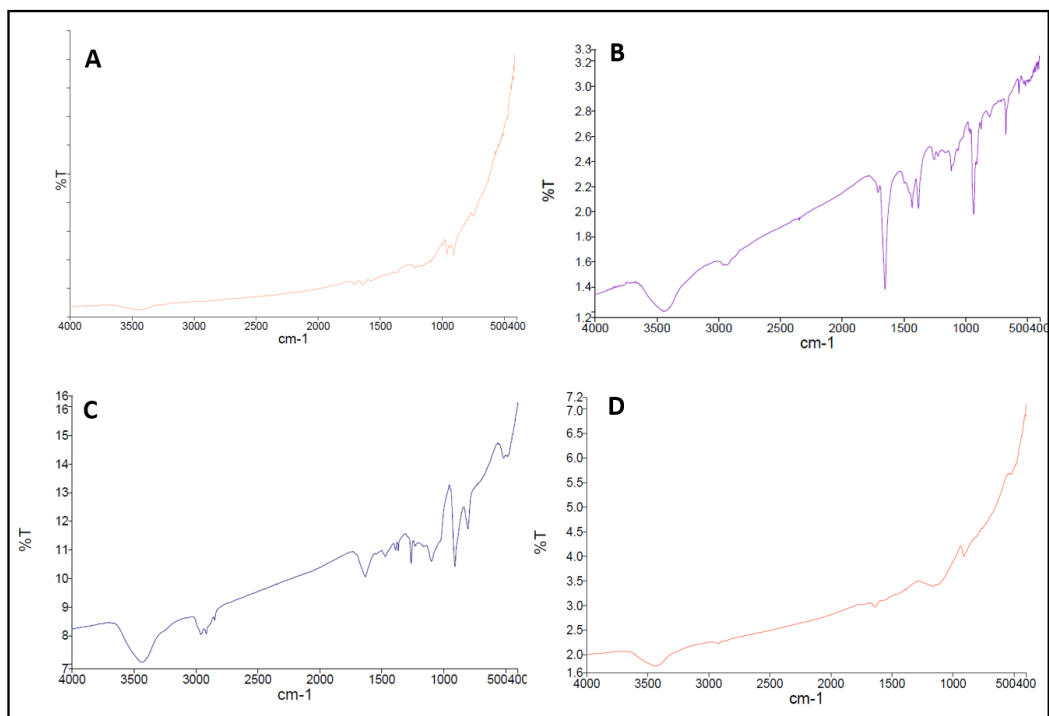


Figure S4. FT-IR of catalysts **V E-H**. Panel A: catalyst **V E**. Panel B: catalyst **V F**. Panel C : catalyst **V G**. Panel D: catalyst **V H**.

SI#2: Difference method of neutron diffraction and isotopic substitution

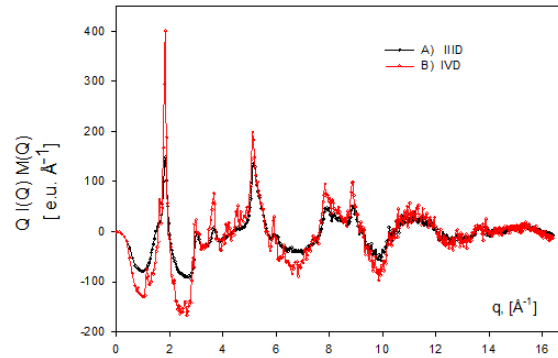


Figure S5. Experimental structure functions in the $QI(Q)M(Q)$ form: A) **III D** sample, B) **IV D** sample

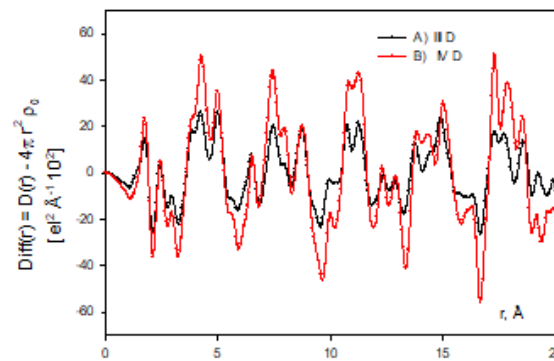


Figure S6. Experimental radial distribution functions in the $\text{Diff}(r) = D(r) - 4\pi r^2 \rho_0$ form: A) **III D** sample, B) **IV D** sample.

The experimental data are transformed into electronic units by the following equation:

$$I(Q) = I_{\text{exp}}(Q) - \sum_{i=1}^N x_i f_i(Q)^2$$

The radial distribution $D(r)$ is calculated as

$$D(r) = 4\pi r^2 \rho_0 + 2r \pi^{-1} \int_0^{S_{\text{max}}} QI(Q)M(Q)\sin(rQ)dQ$$

where q is the scattering parameter, ρ_0 the samples average electronic density $\rho_0 = (\sum_h n_h f_h(0))^2 V^{-1}$, V the stoichiometric unit volume and $M(q)$ a modification function defined by:

$$M(q) = f_{\text{Re}}^2(0) / f_{\text{Re}}^2(Q) \exp(-0.01Q^2)$$

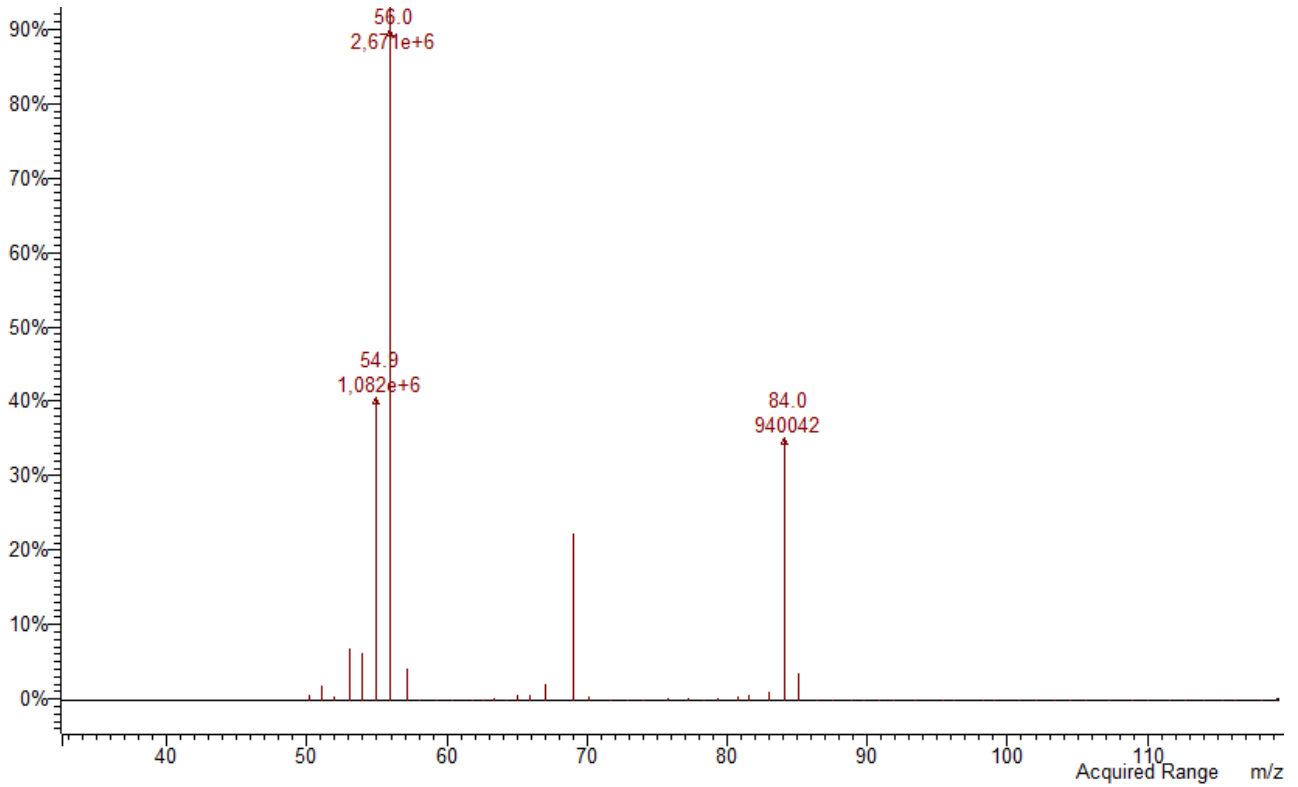
The experimental radial distribution functions are shown also as $\text{Diff}(r) = D(r) - 4\pi r^2 \rho_0$. Theoretical peaks were calculated by Fourier transformation of the theoretical intensities for the pair interactions,

$$I_{ij} = \sum f_i f_j \sin(r_{ij} Q) (r_{ij} Q)^{-1} \exp(-1/2\sigma_{ij}^2 Q^2)$$

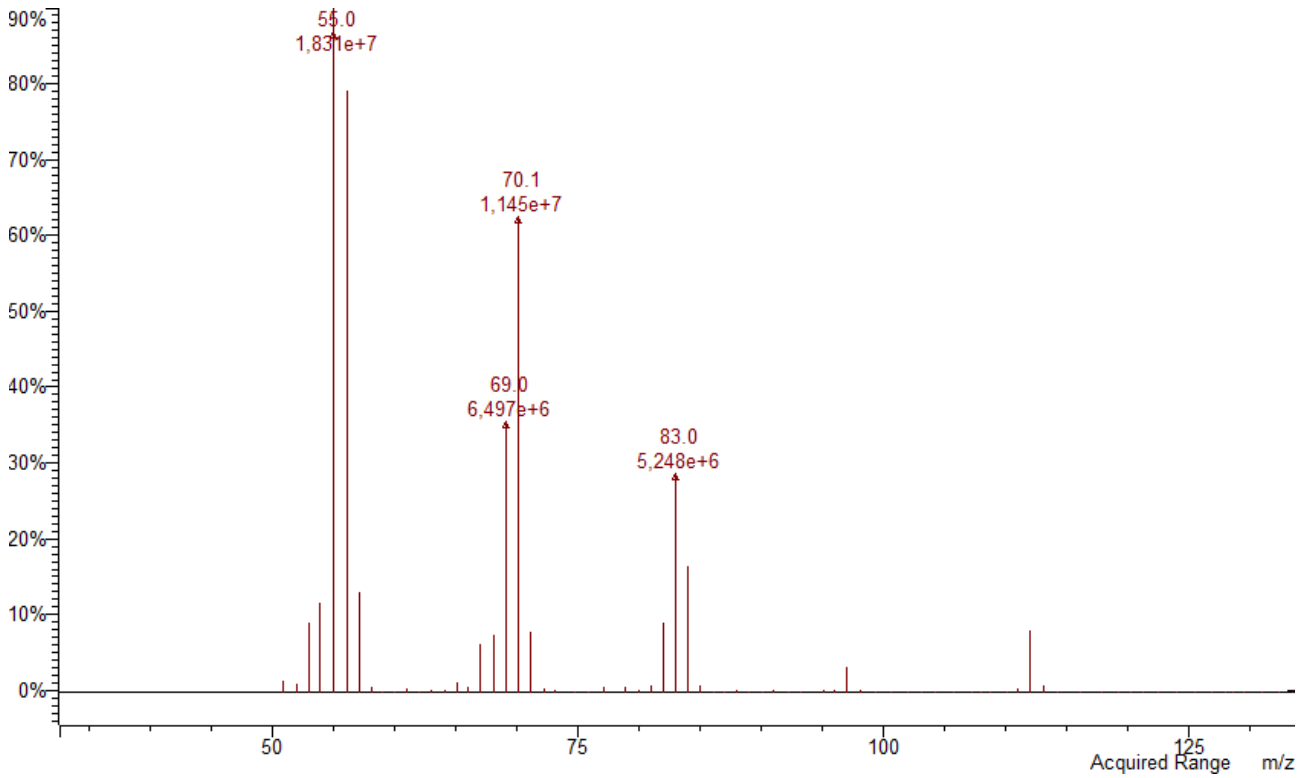
using the same sharpening function as for the experimental data, and assuming the root-mean-square variation over distance to be σ_{ij} . The experimental radial distribution functions of **III D** and **IV D** in the $\text{Diff}(r)$ form (Figure 2) showed peaks at about 1.70, 2.40 and 3.0 Å and broadened peaks at greater distances related to MWCNT structure. The shoulder at 1.45 Å and the peak at 2.45 Å can be attributed to the main interactions of MWCNT and B15C5 moieties. The first value represents the distances between directly bounded atoms, while the second is representative of the interactions between two non-directly bounded atoms, one of which is located in the network of MWCNT and the other in B15C5. To apply the differences method, the weighted amount of **III D** in the experimental distribution radial function $D(r)$ curve was subtracted from the respective $D(r)$ curve of **IV D** to extract the MTO/B15C5 interactions.

SI#3: Mass spectrometry profiles of compounds 1-21

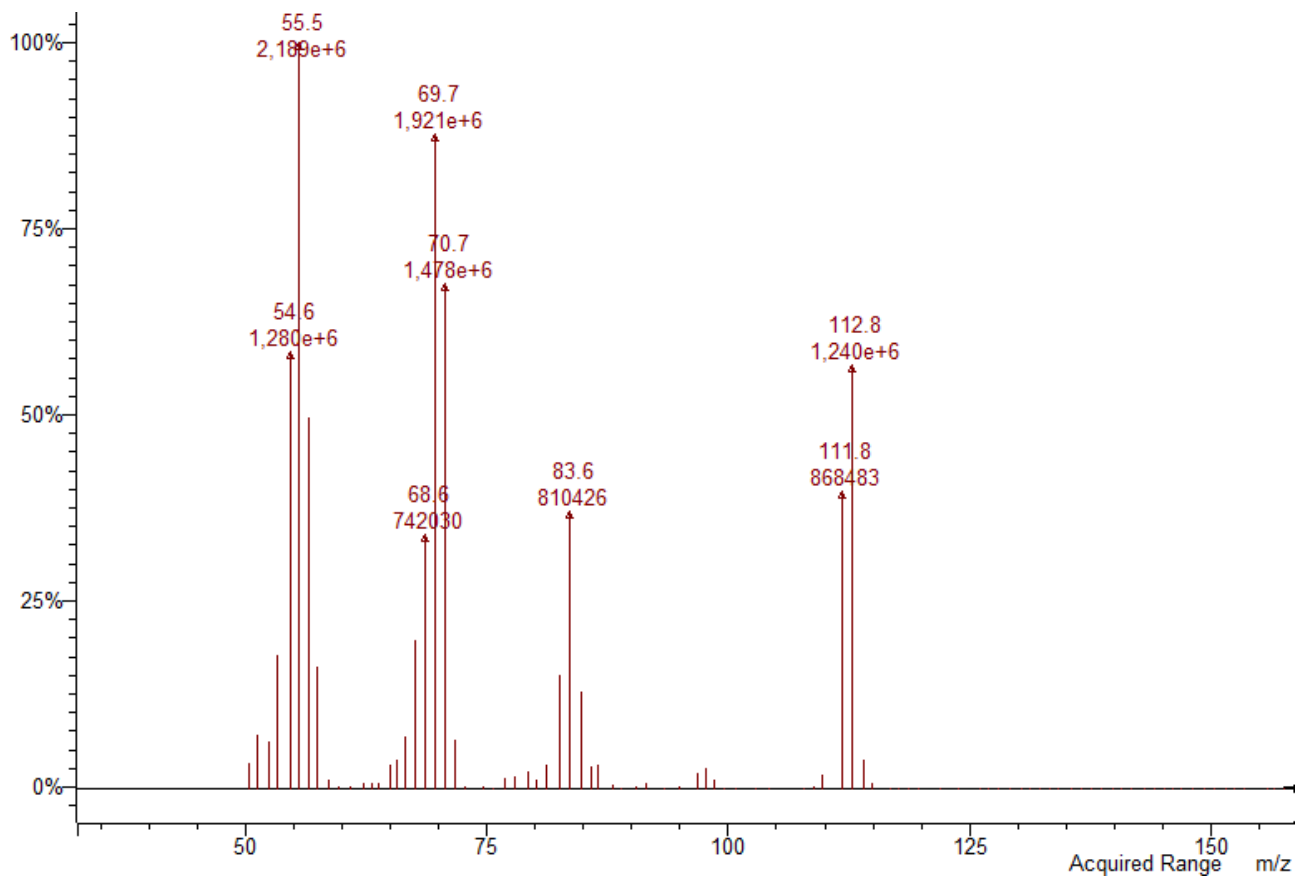
Mass spectrometry profiles of Hexene (1).



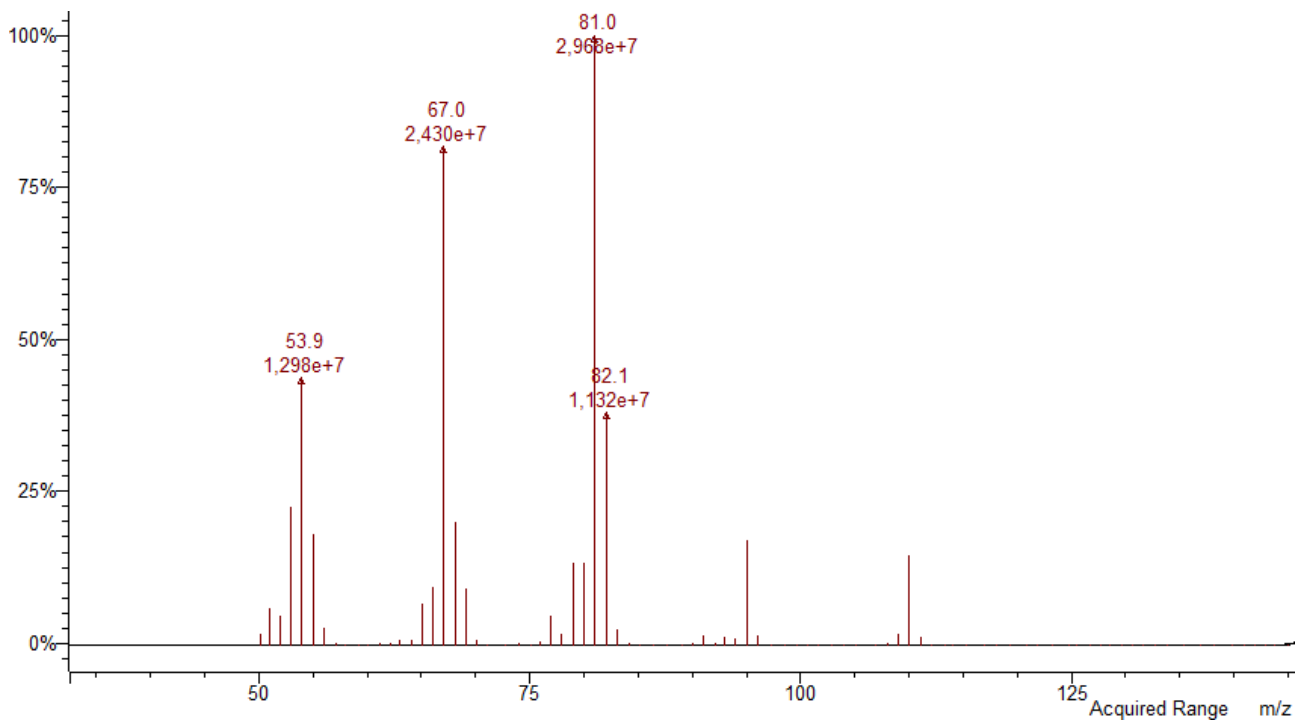
Mass spectrometry profiles of Octene (2).



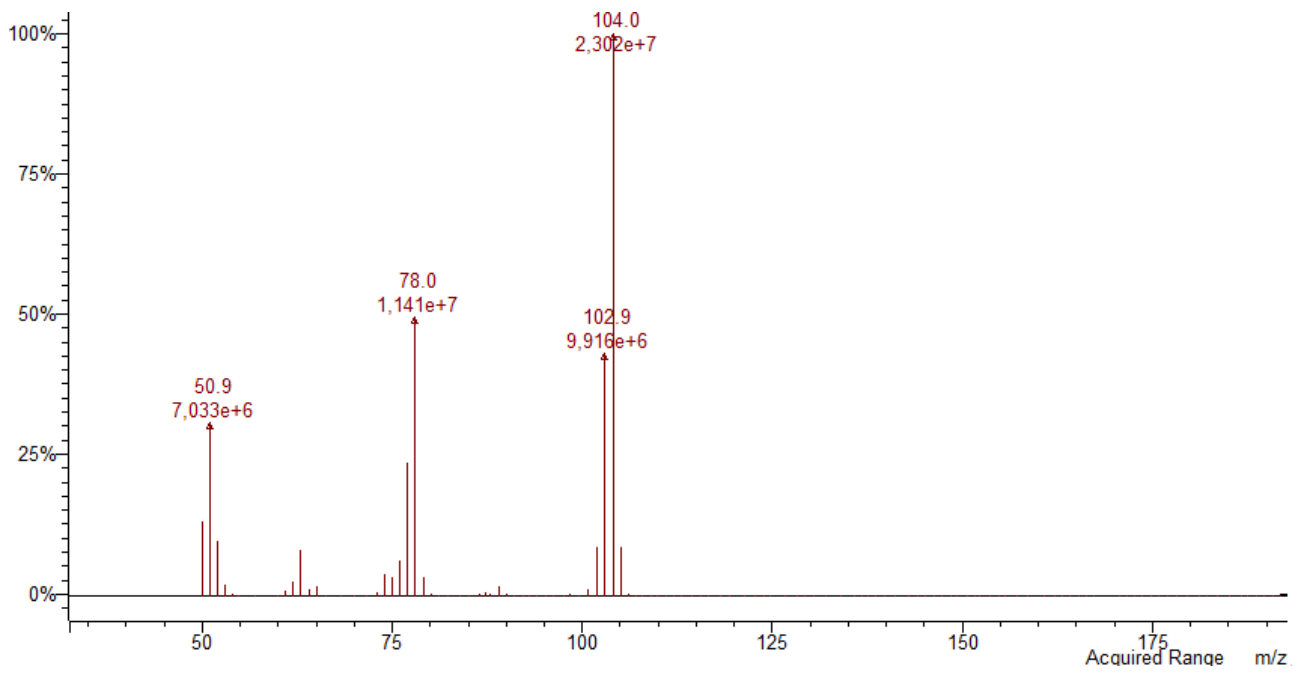
Mass spectrometry profiles of Trans-3-Octene (3).



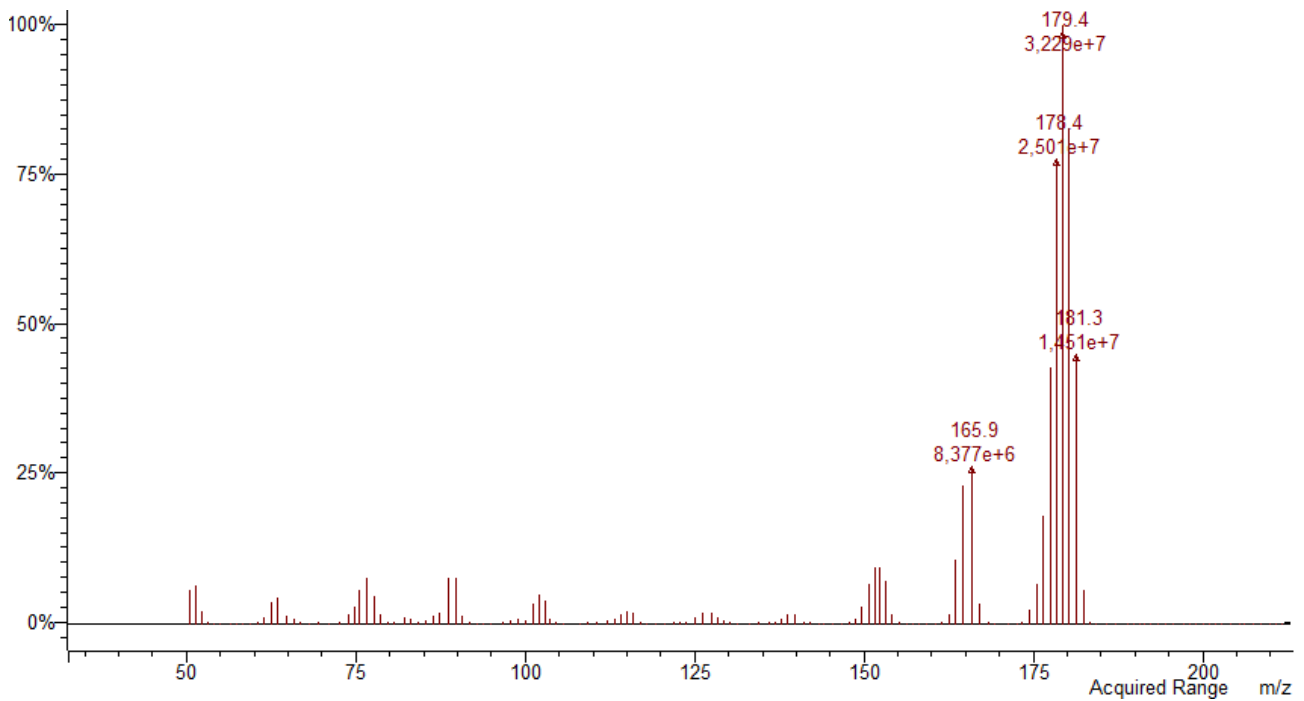
Mass spectrometry profiles of Vinylcyclohexane (4).



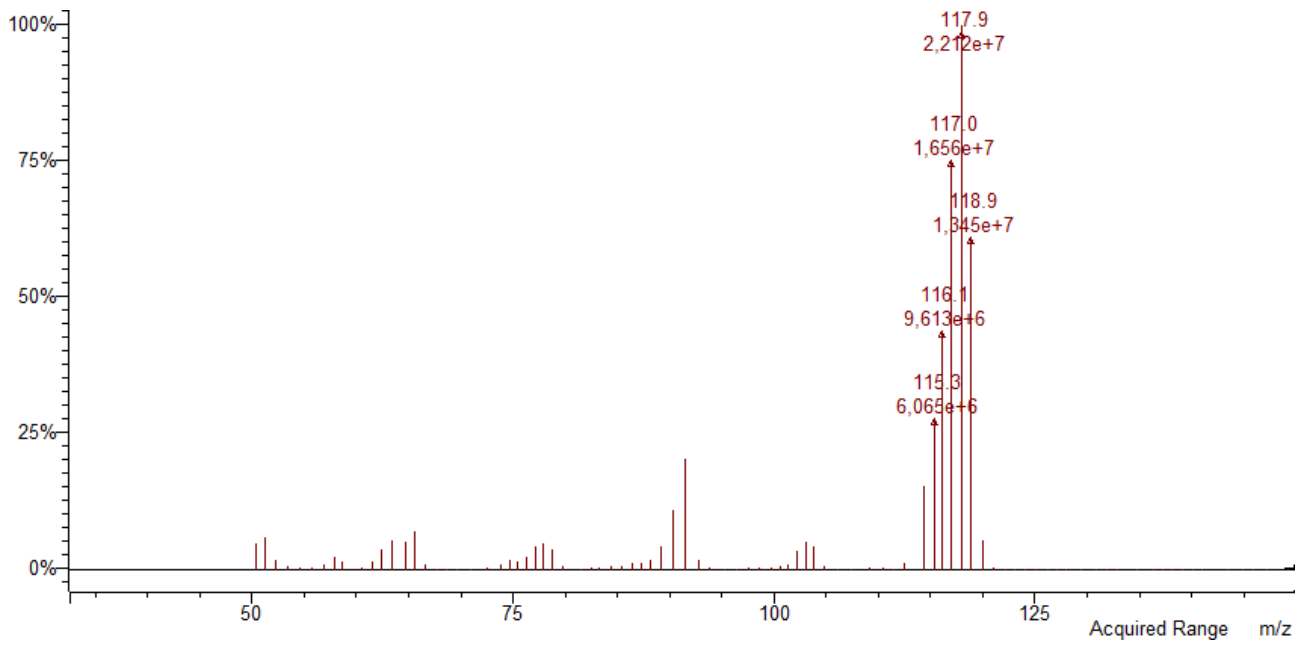
Mass spectrometry profiles of Styrene (5).



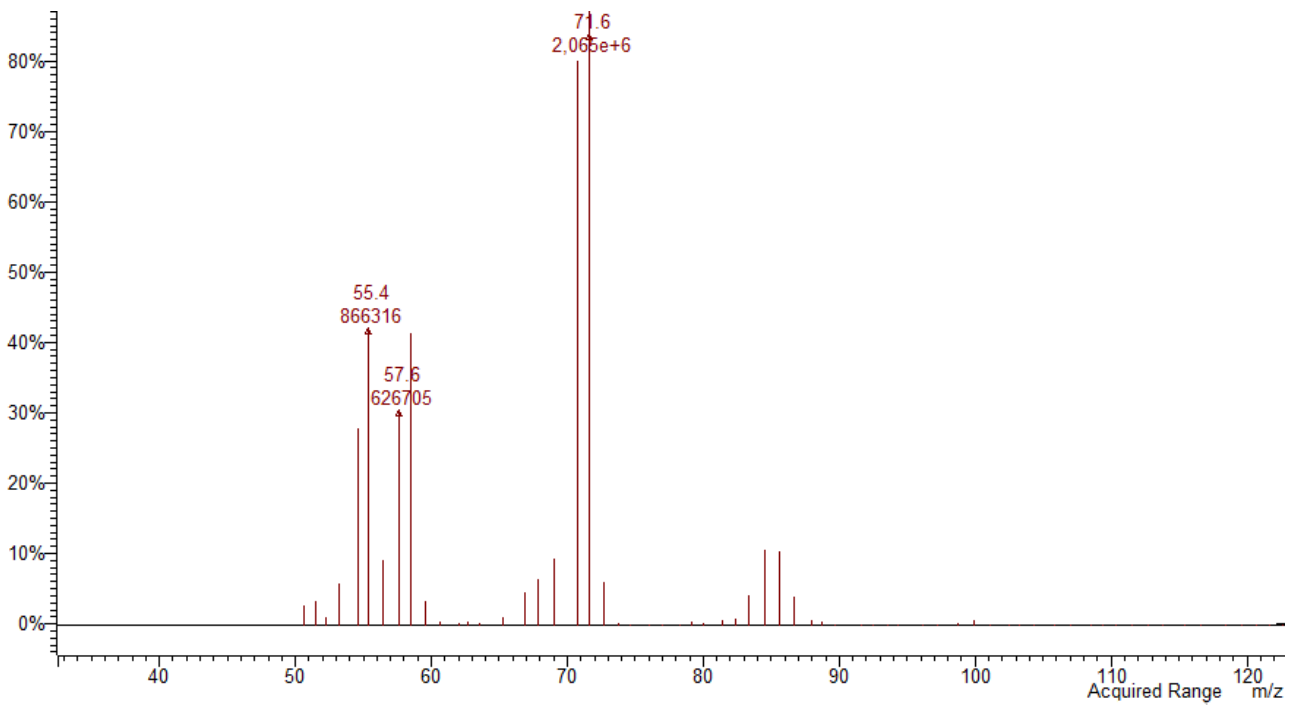
Mass spectrometry profiles of Cis-stilbene (6).



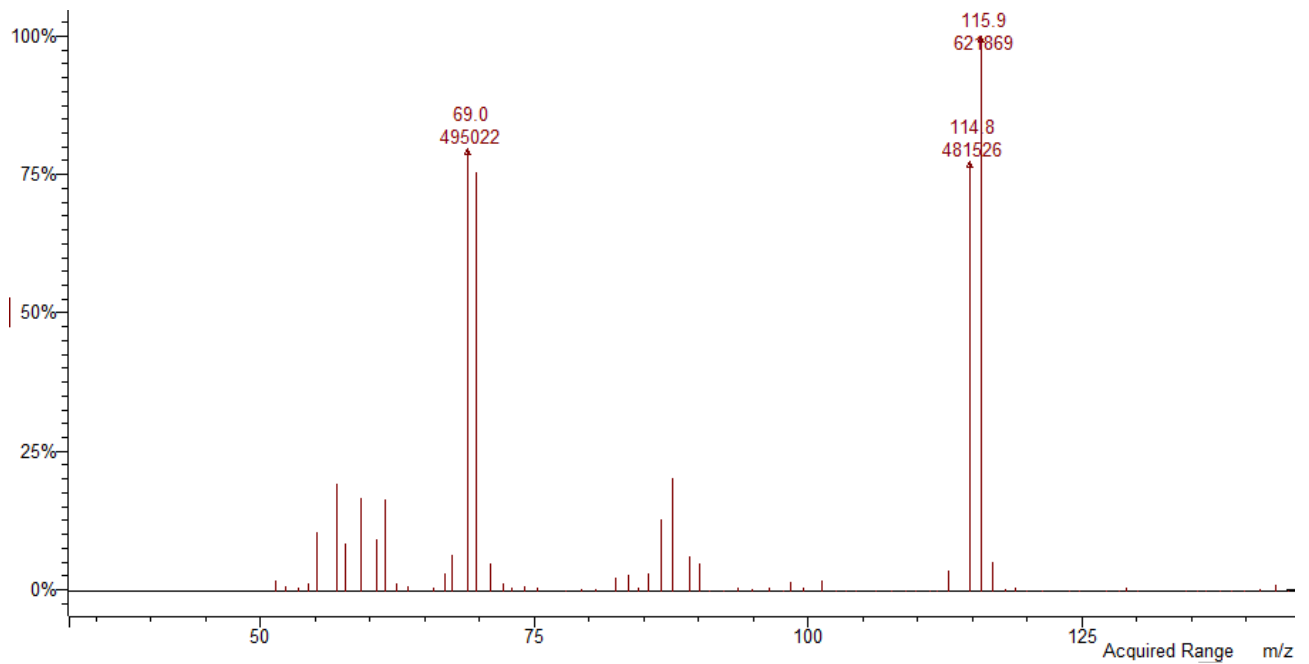
Mass spectrometry profiles of Allylbenzene (7).



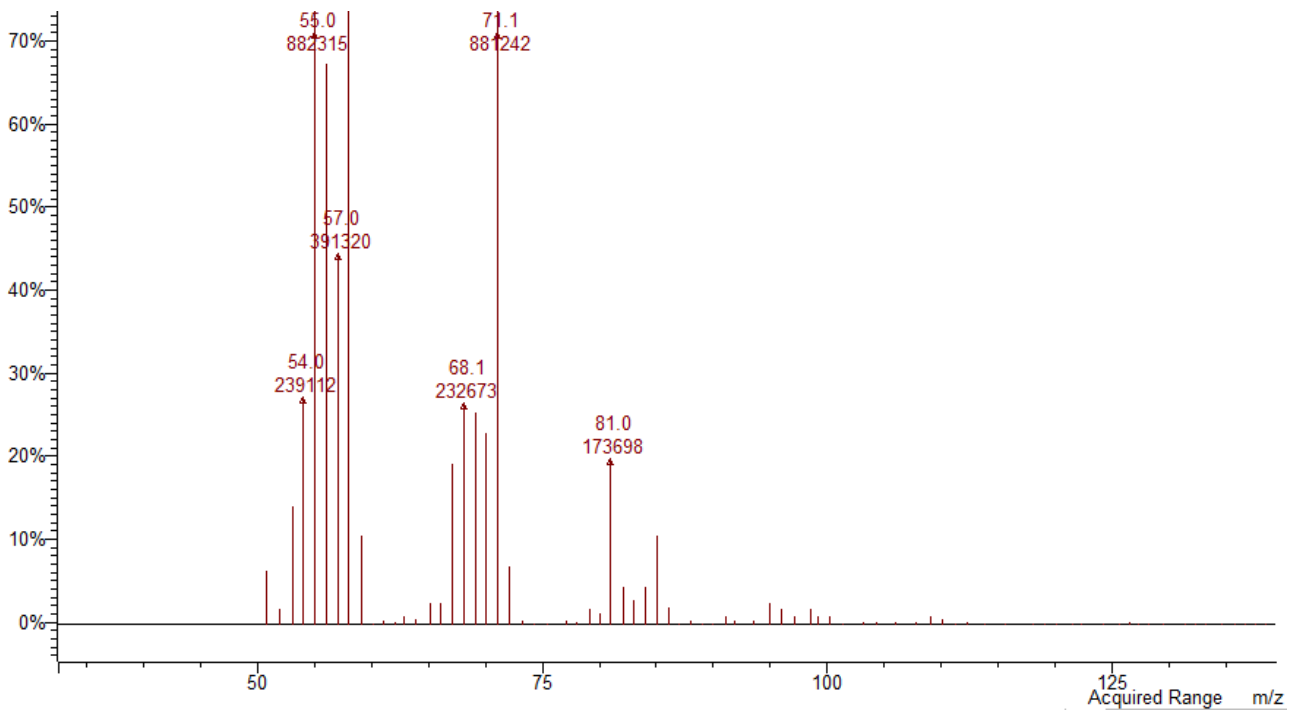
Mass spectrometry profiles of 1,2-Epoxyhexane (8).



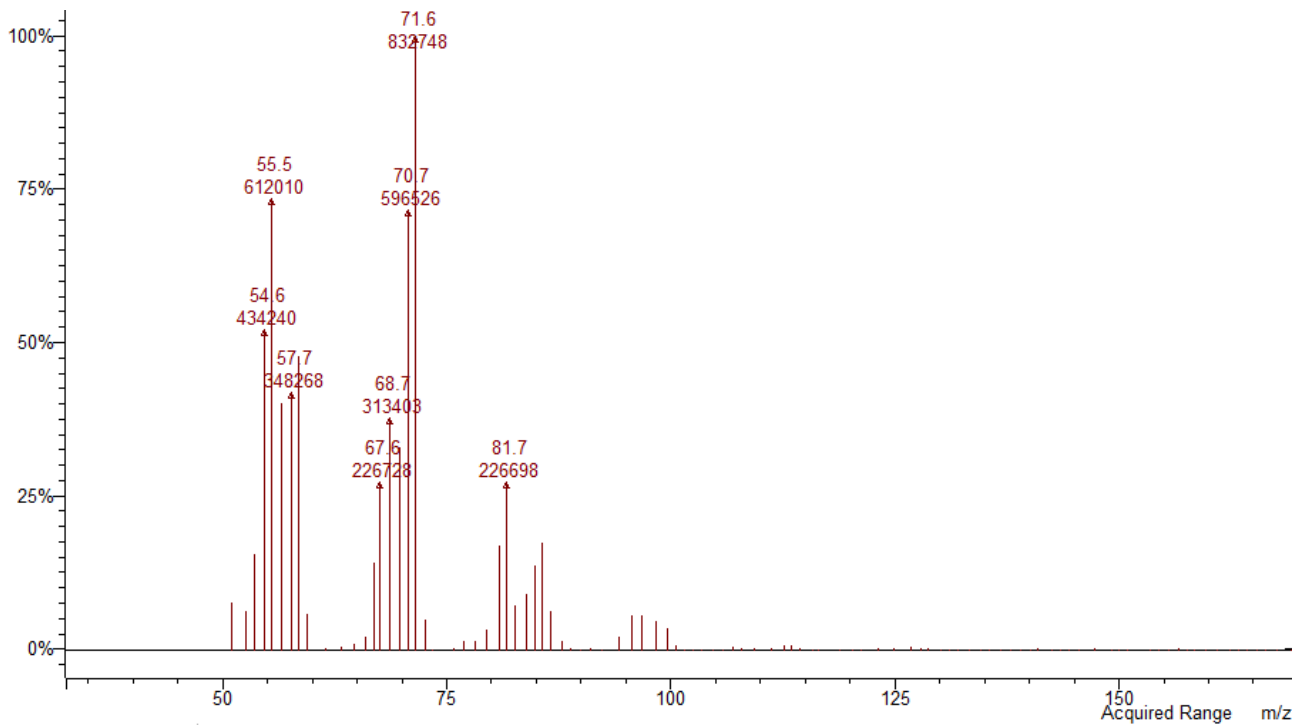
Mass spectrometry profiles of Hexane-1,2-diol (9).



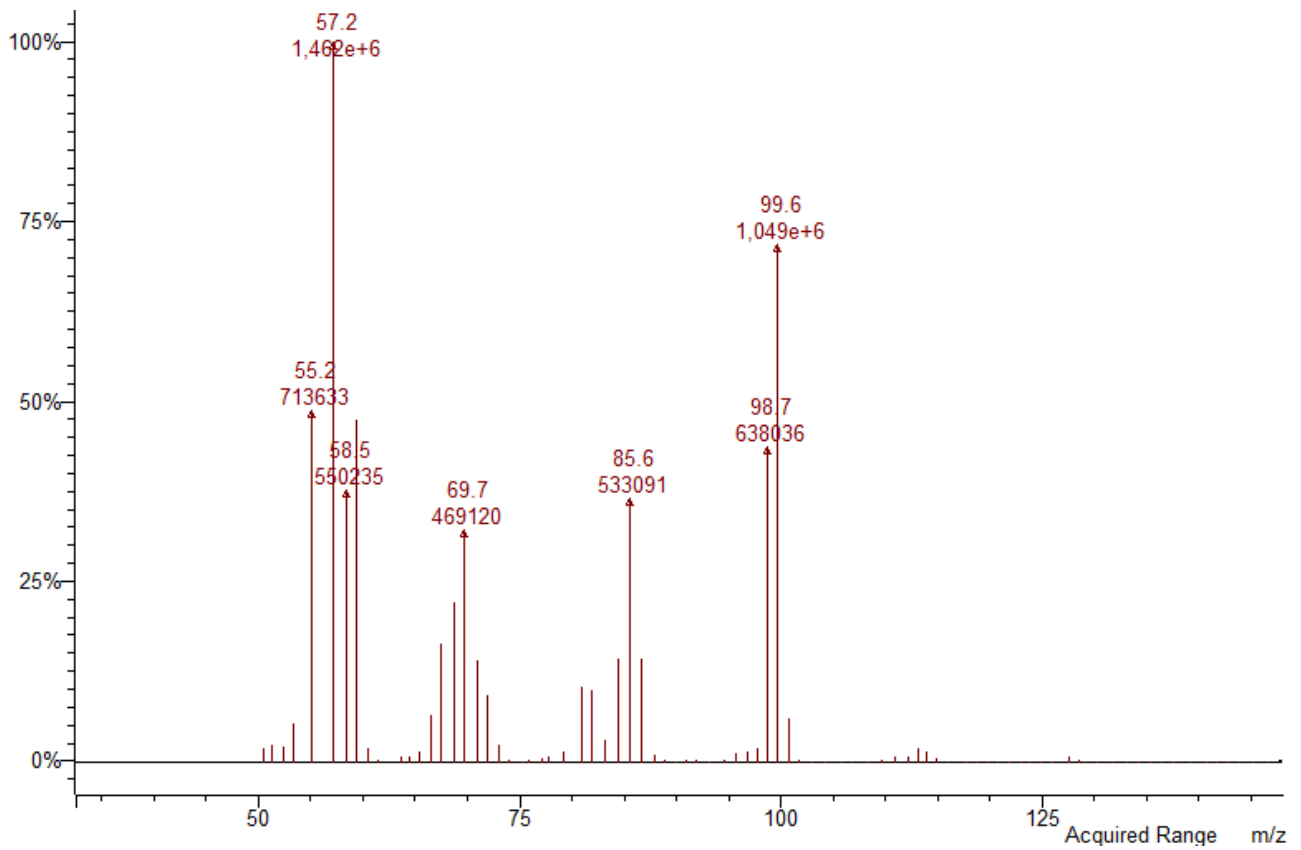
Mass spectrometry profiles of 1,2-Epoxyoctane (10).



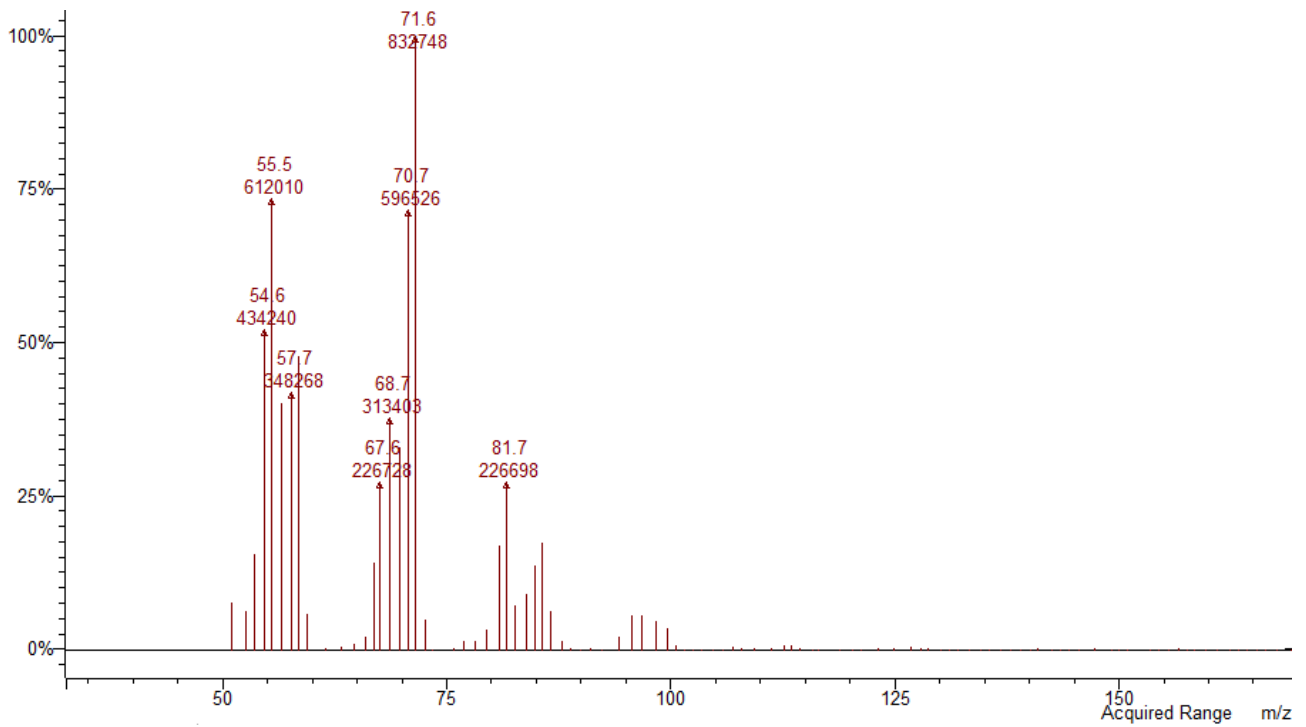
Mass spectrometry profiles of Octane-1,2-diol (11).



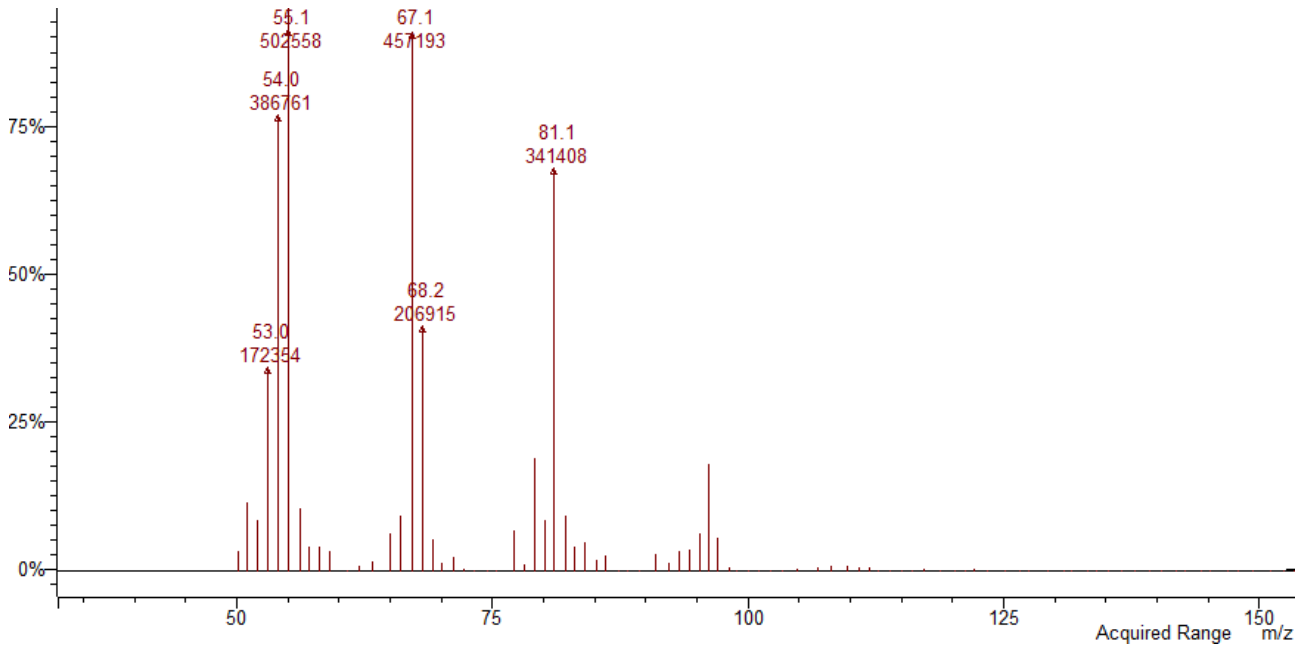
Mass spectrometry profiles of Trans-3,4-Epoxyoctane(12).



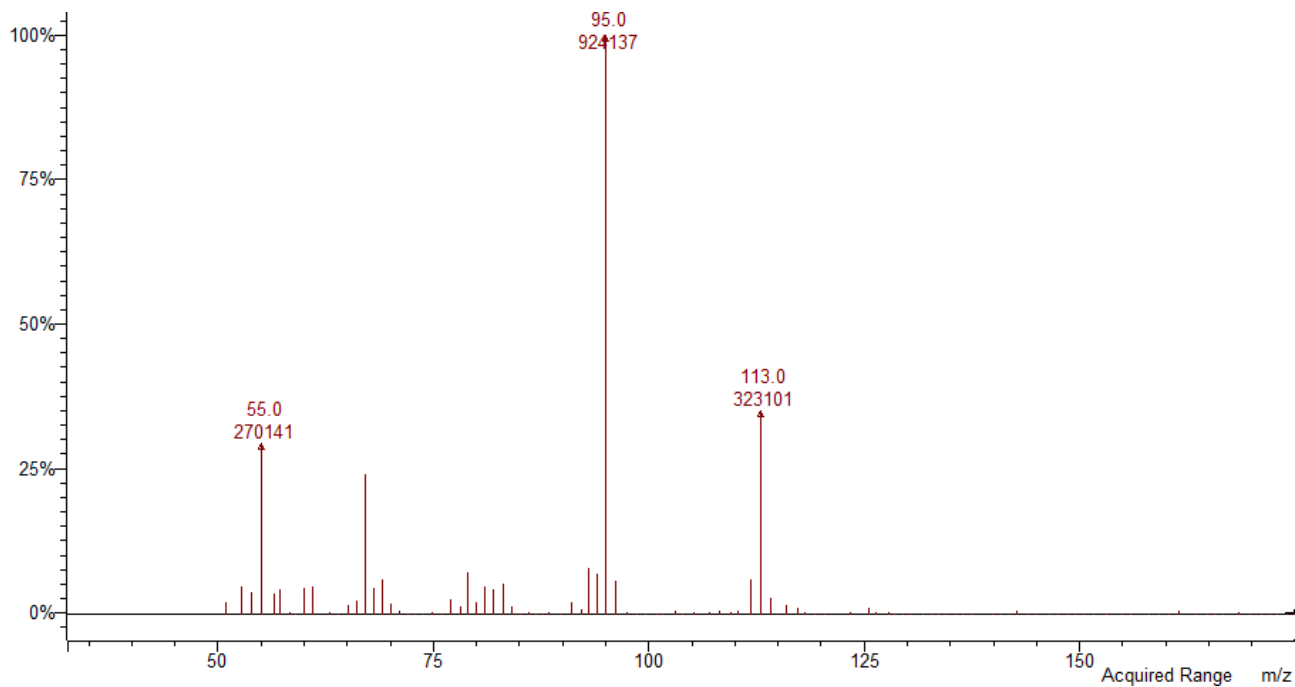
Mass spectrometry profiles of Octane-3,4-diol (13).



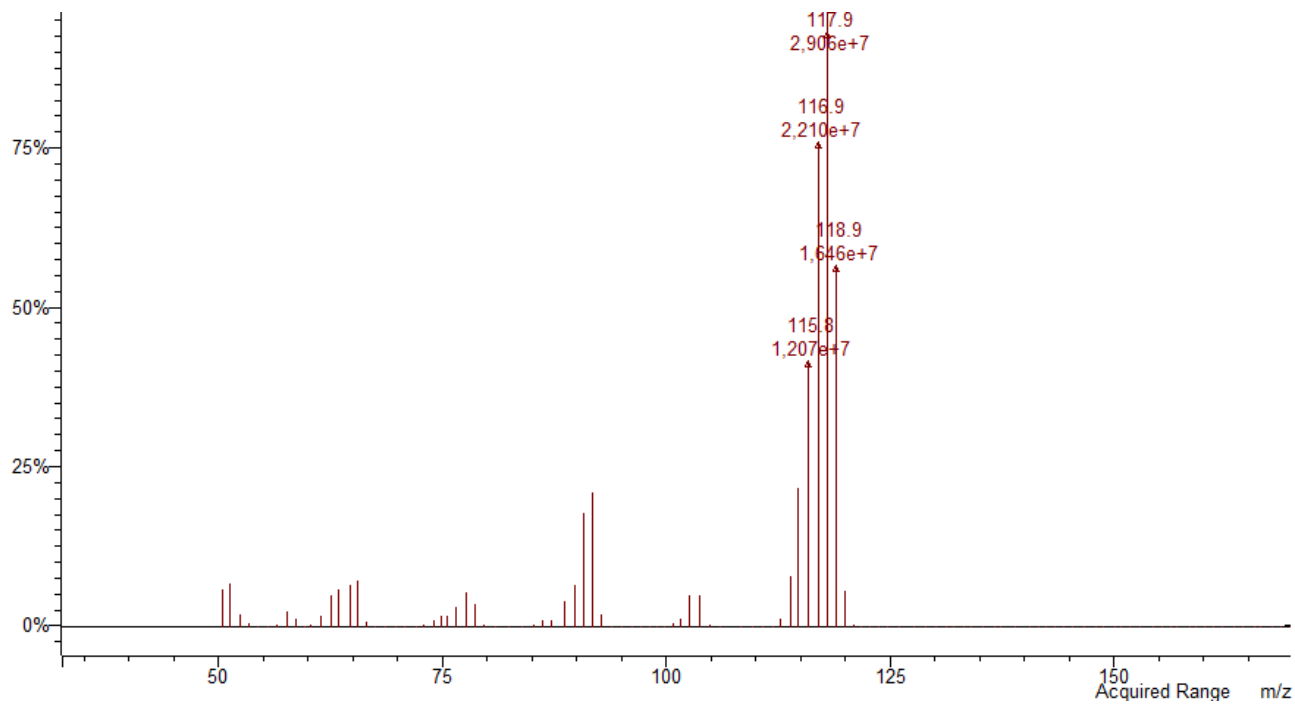
Mass spectrometry profiles of 2-Cyclohexyloxirane (14).



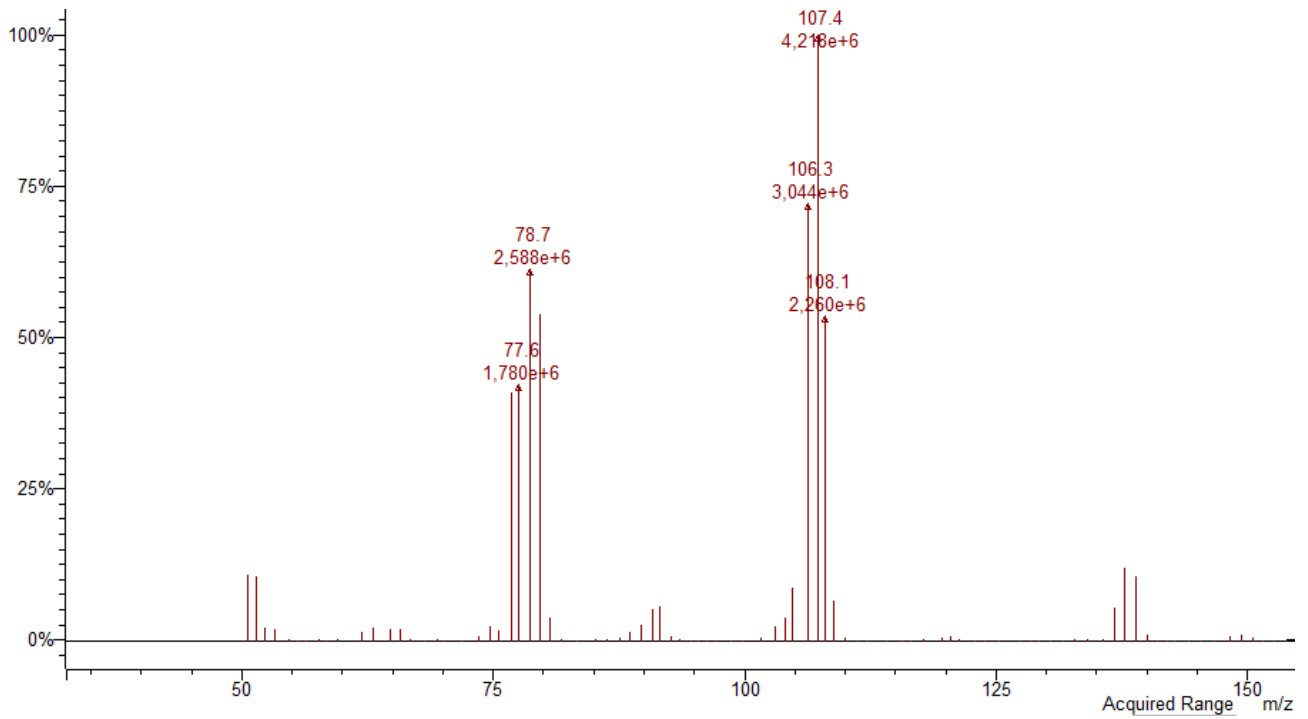
Mass spectrometry profiles of 1-Cyclohexylethane-1,2-diol (15).



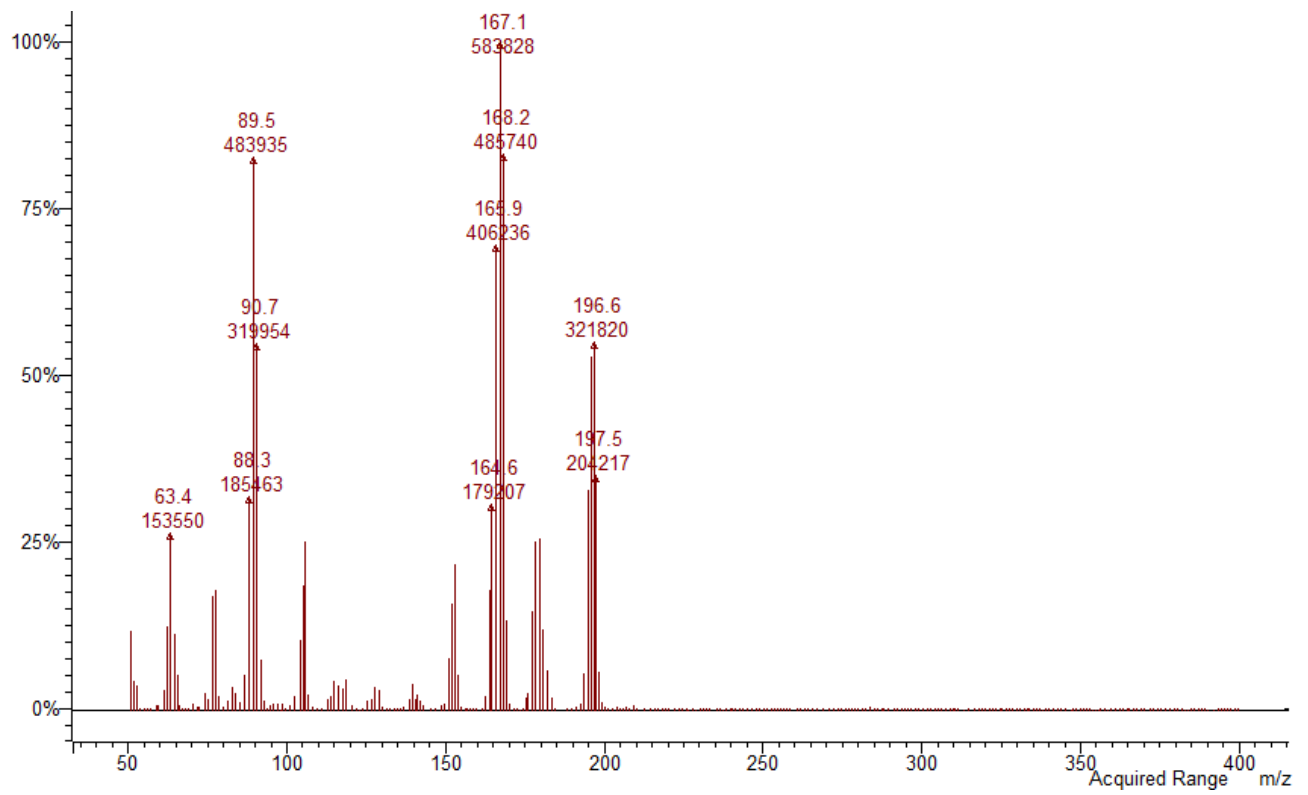
Mass spectrometry profiles of 2-Phenyloxirane (16).



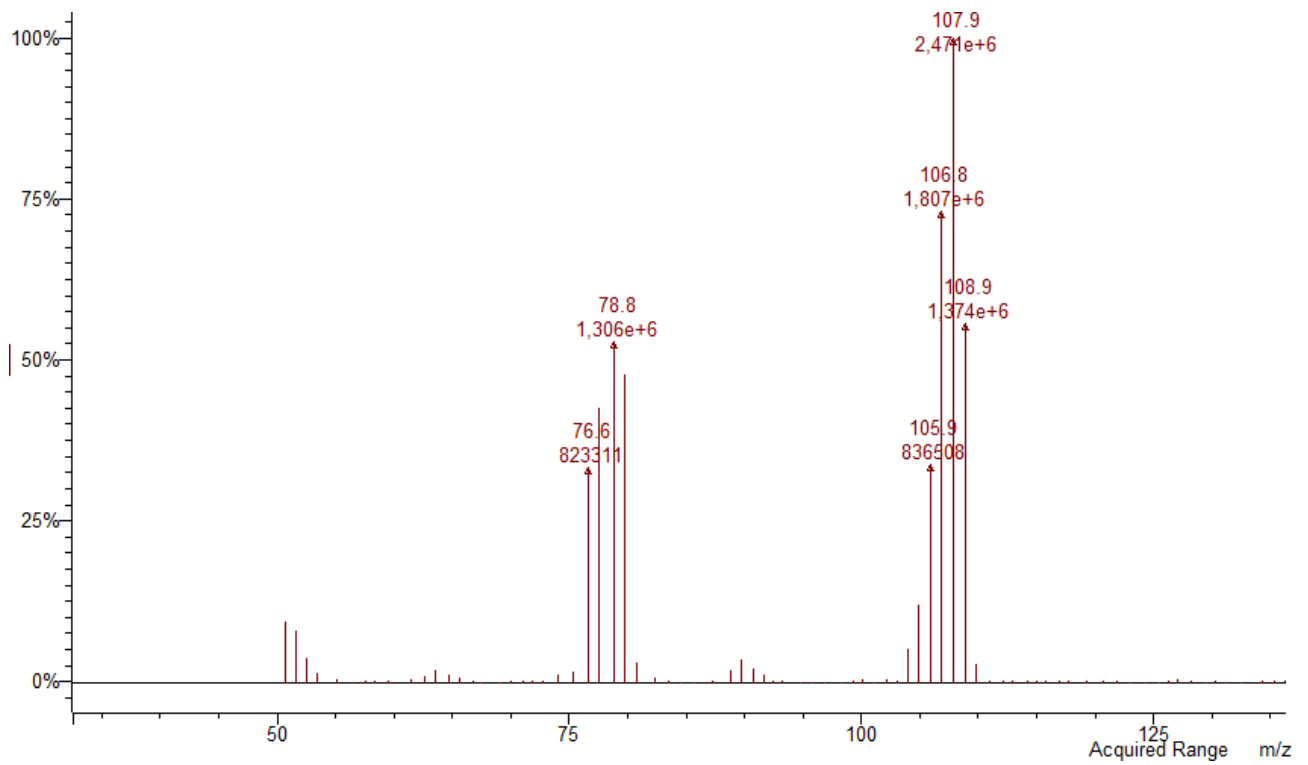
Mass spectrometry profiles of 1-Phenylethane-1,2-diol (17).



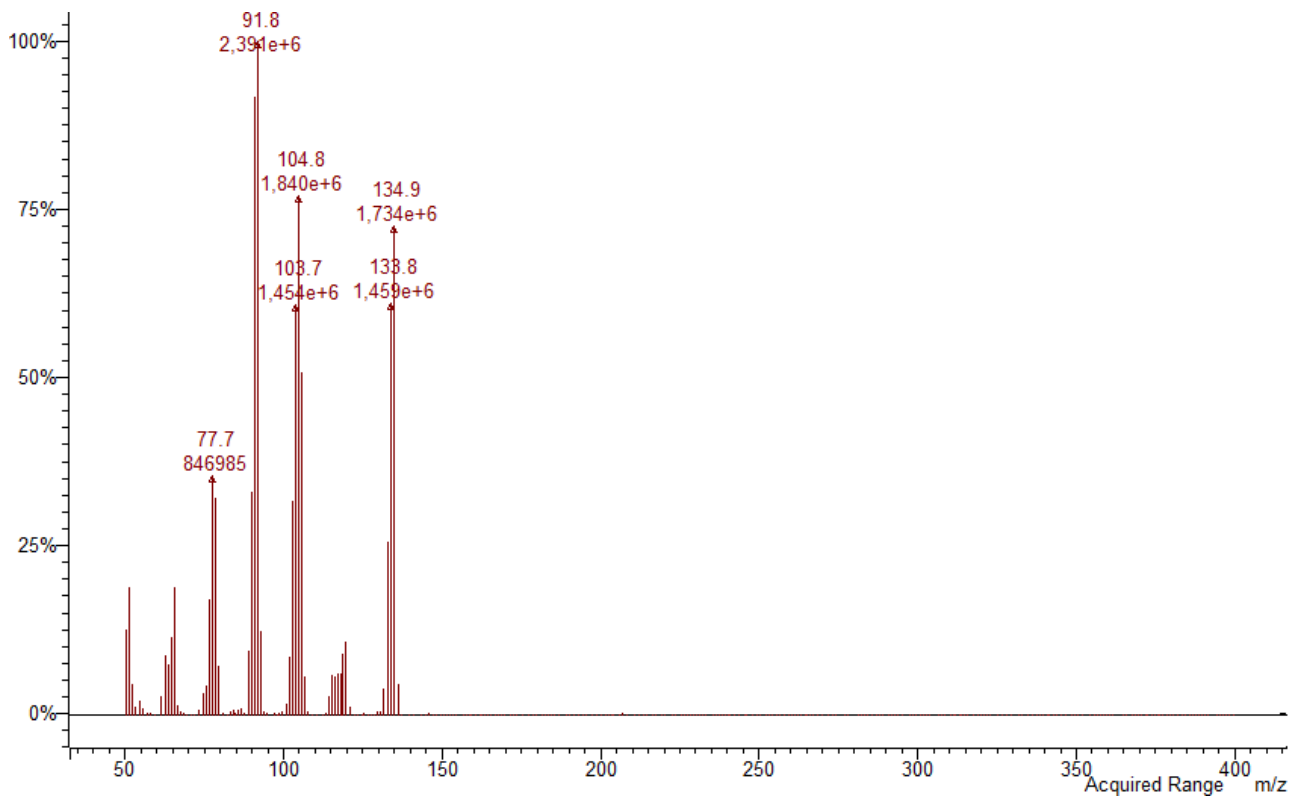
Mass spectrometry profiles of 2,3-Diphenyloxirane (18).



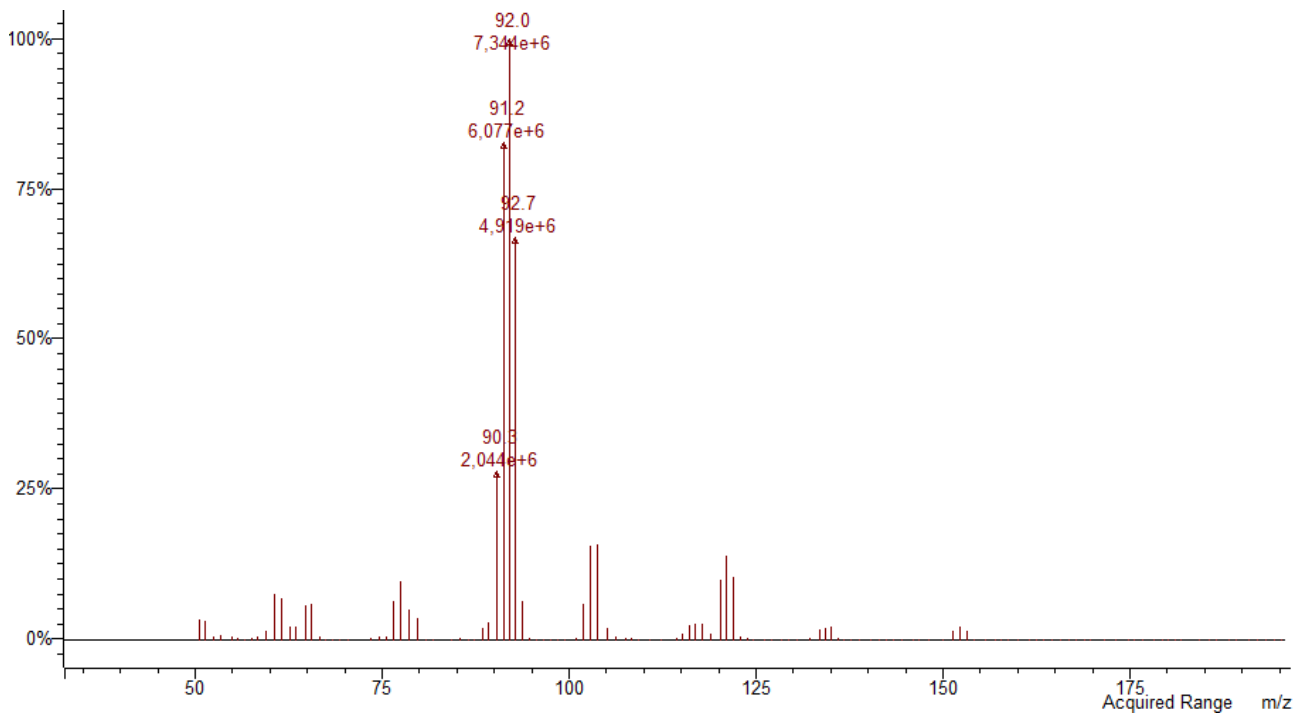
Mass spectrometry profiles of 1,2-Diphenylethane-1,2-diol (19).



Mass spectrometry profiles of 2-benzyloxirane (20).



Mass spectrometry profiles of 3-phenylpropane-1,2-diol (21).



SI#4: Nuclear magnetic resonance of compounds 1-21

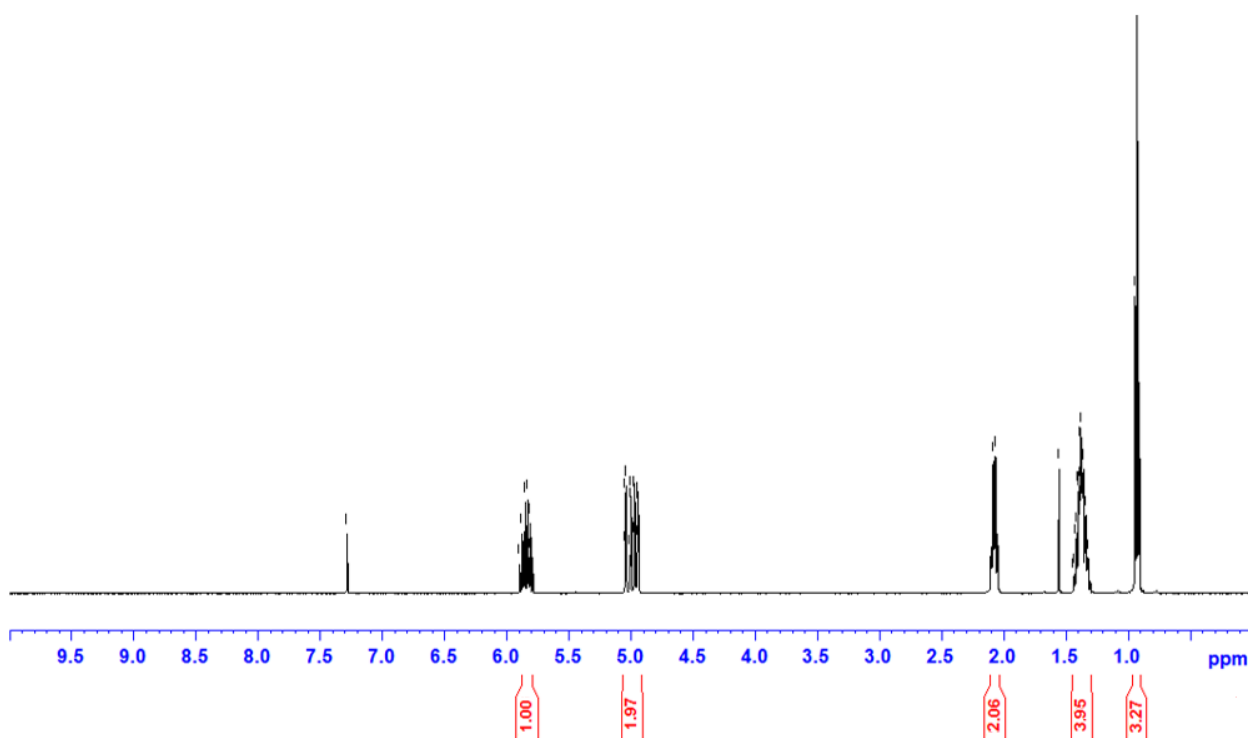


Figure SI#4-1. ^1H NMR of 1-hexene (**1**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 5.79-5.89 (m, 1H, $-\text{CH}=\text{}$), 4.93-5.04 (m, 2H, $=\text{CH}_2$), 2.07 (m, 2H, $-\text{CH}_2-$), 1.36-1.43 (m, 4H, 2 x $-\text{CH}_2-$), 0.90 (m, 3H, CH_3-).

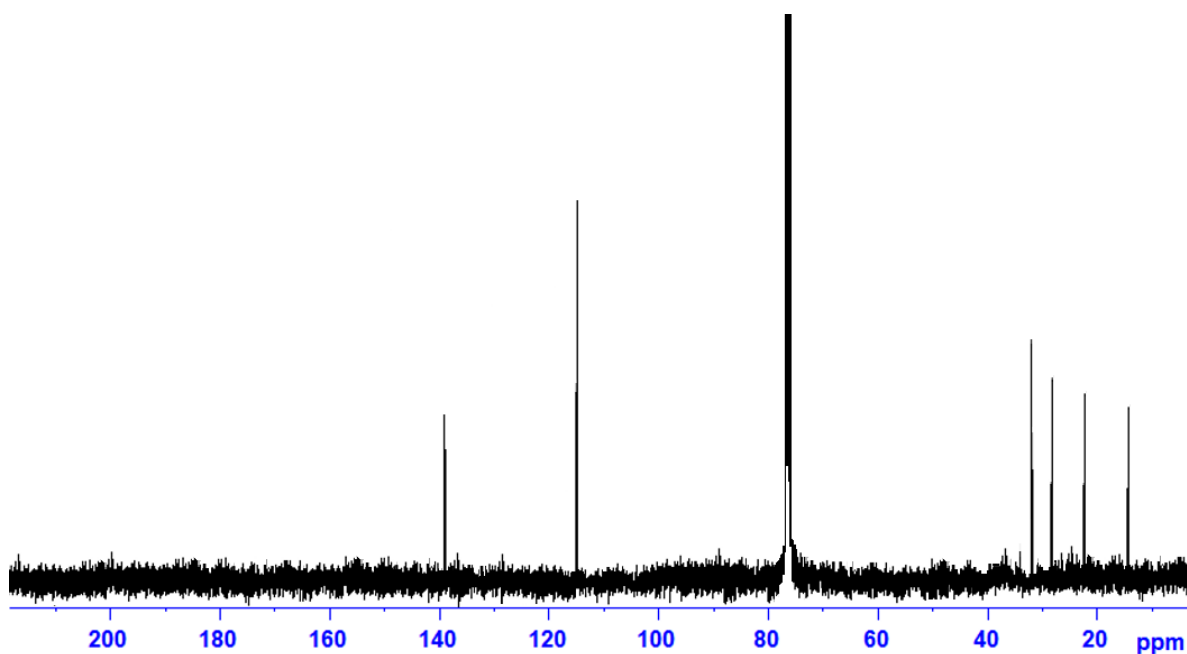


Figure SI#4-2. ^{13}C NMR of 1-hexene (**1**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 139.1 ($-\text{CH}=\text{}$), 115.7 ($=\text{CH}_2$), 32.5, 28.4, 22.8 ($-\text{CH}_2-$), 14.1 (CH_3-).

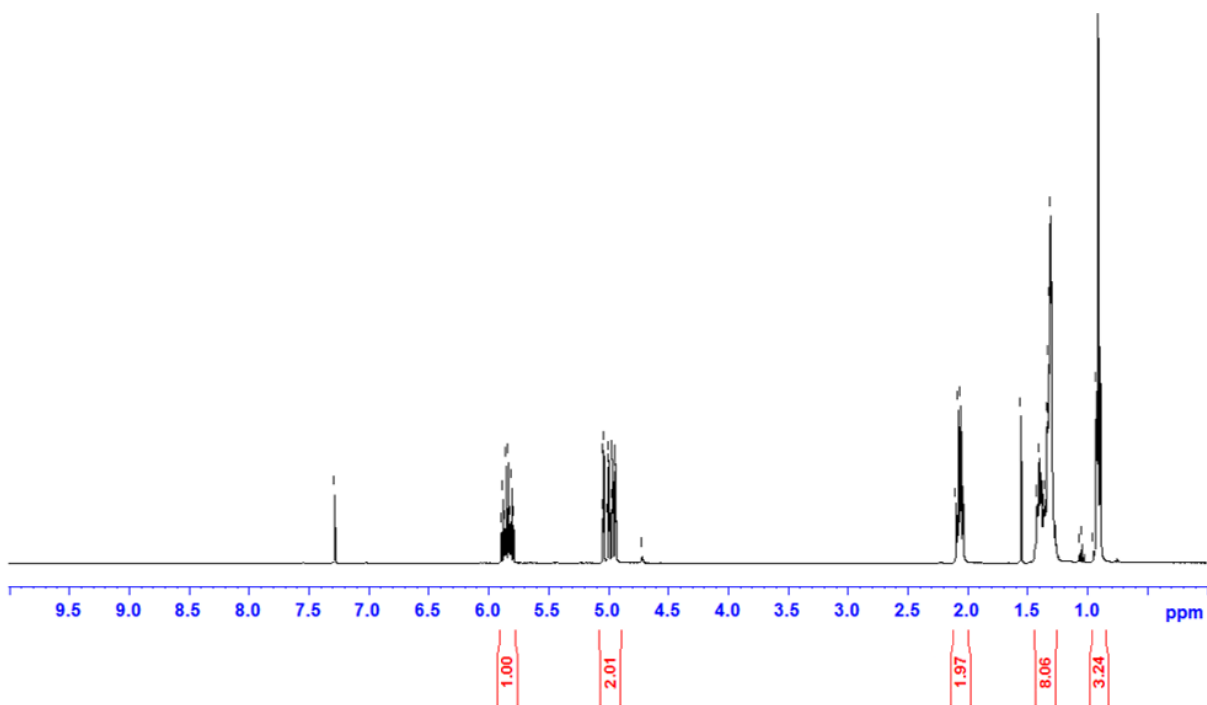


Figure SI#4-3. ^1H NMR of 1-octene (**2**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 5.79-5.87 (m, 1H, $-\text{CH}=\text{}$), 4.93-5.04 (m, 2H, $=\text{CH}_2$), 2.06 (m, 2H, $-\text{CH}_2-$), 1.28-1.42 (m, 8H, 4 \times $-\text{CH}_2-$), 0.91 (m, 3H, CH_3 -).

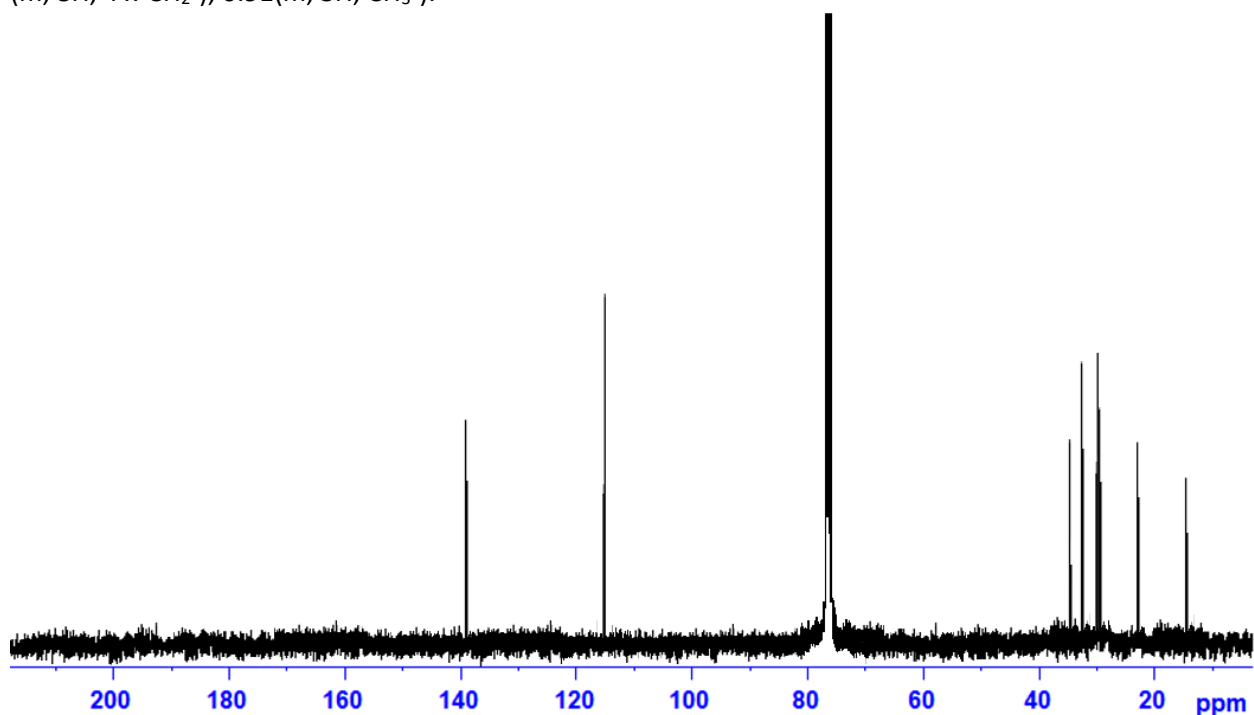


Figure SI#4-4. ^{13}C NMR of 1-Octene (**2**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 139.1 ($-\text{CH}=\text{}$), 115.7 ($=\text{CH}_2$), 33.9, 31.9, 29.6, 29.4, 22.7 ($-\text{CH}_2-$), 14.1 (CH_3 -).

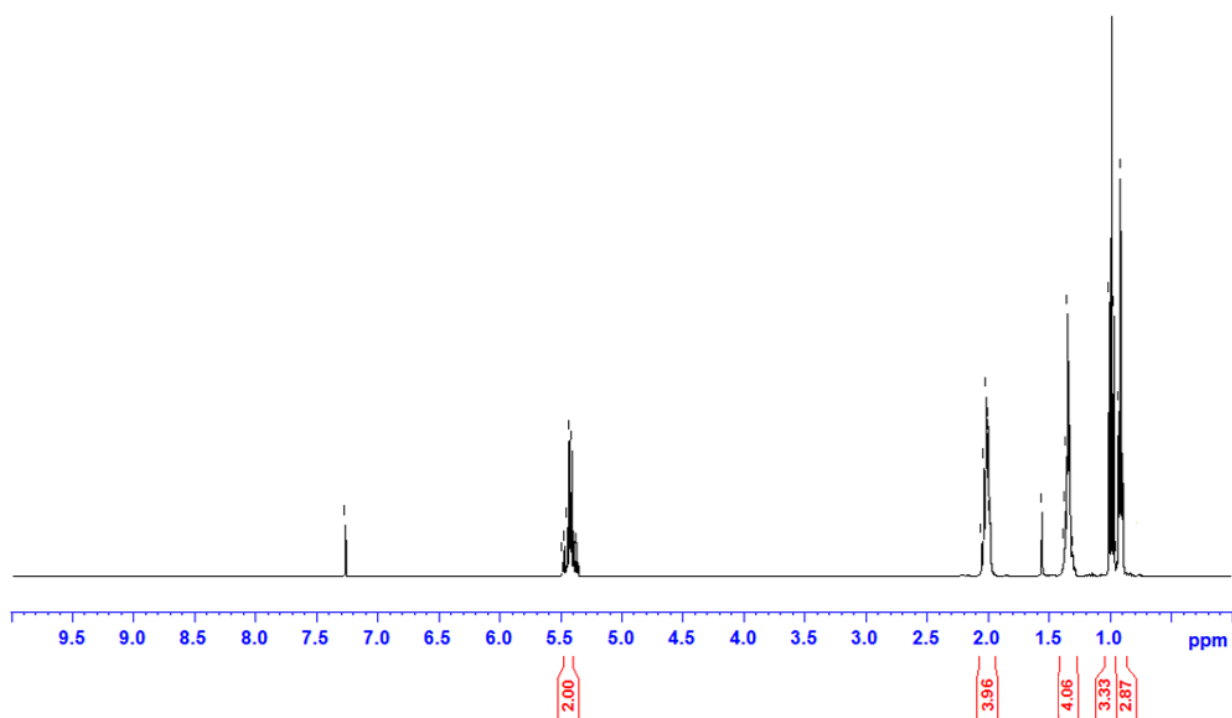


Figure SI#4-5. ^1H NMR of trans-3-octene (**3**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 5.37-5.50 (m, 2H, 2x -CH=), 1.97-2.05 (m, 4H, 2x -CH₂-), 1.30-1.37 (m, 4H, 2x -CH₂-), 1.00(m, 3H, -CH₃) 0.90(m, 3H, -CH₃).

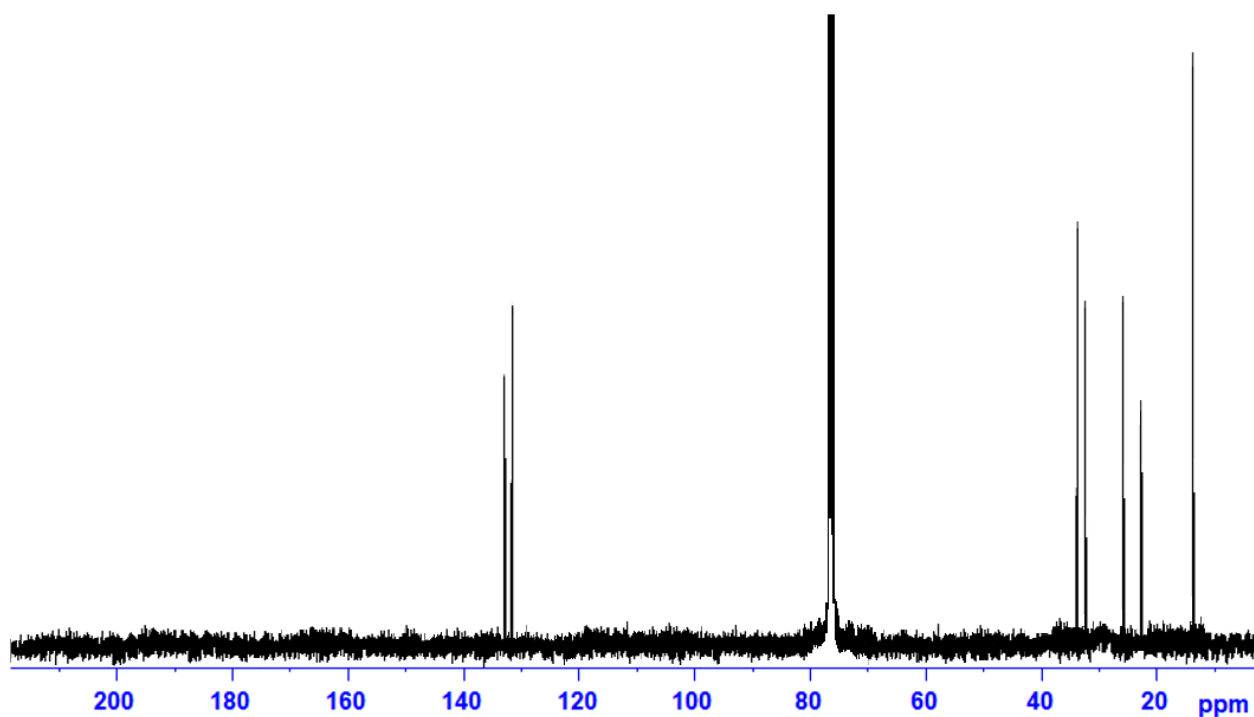


Figure SI#4-6. ^{13}C NMR of trans-3-octene (**3**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 133.7 (-CH=), 131.3 (-CH=), 33.4, 32.1, 26.5, 22.8 (-CH₂-), 14.3 (2 x CH₃-).

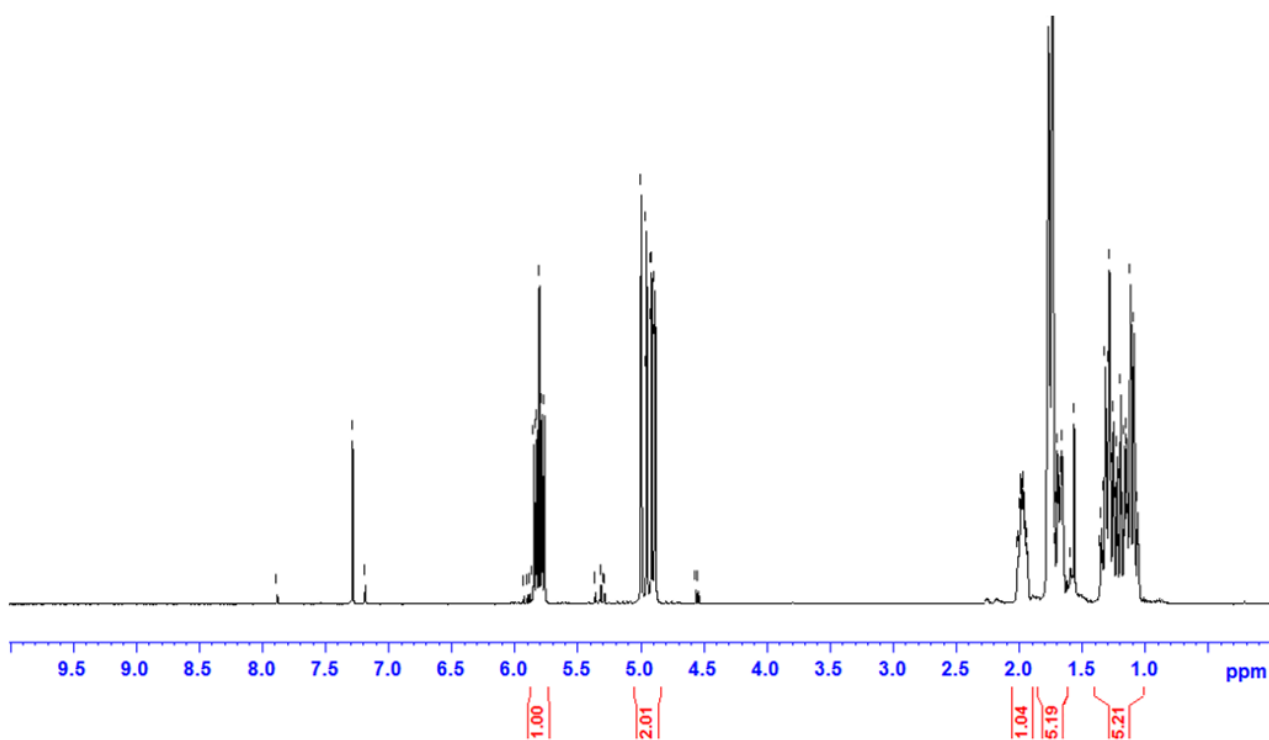


Figure SI#4-7. ^1H NMR of Vinylcyclohexane (**4**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 5.80 (m, 1H, $-\text{CH}=\text{}$), 4.88-5.00 (m, 2H, $=\text{CH}_2$), 1.97 (m, 1H, $-\text{CH}$), 1.09-1.76(m, 10 H, CH_2).

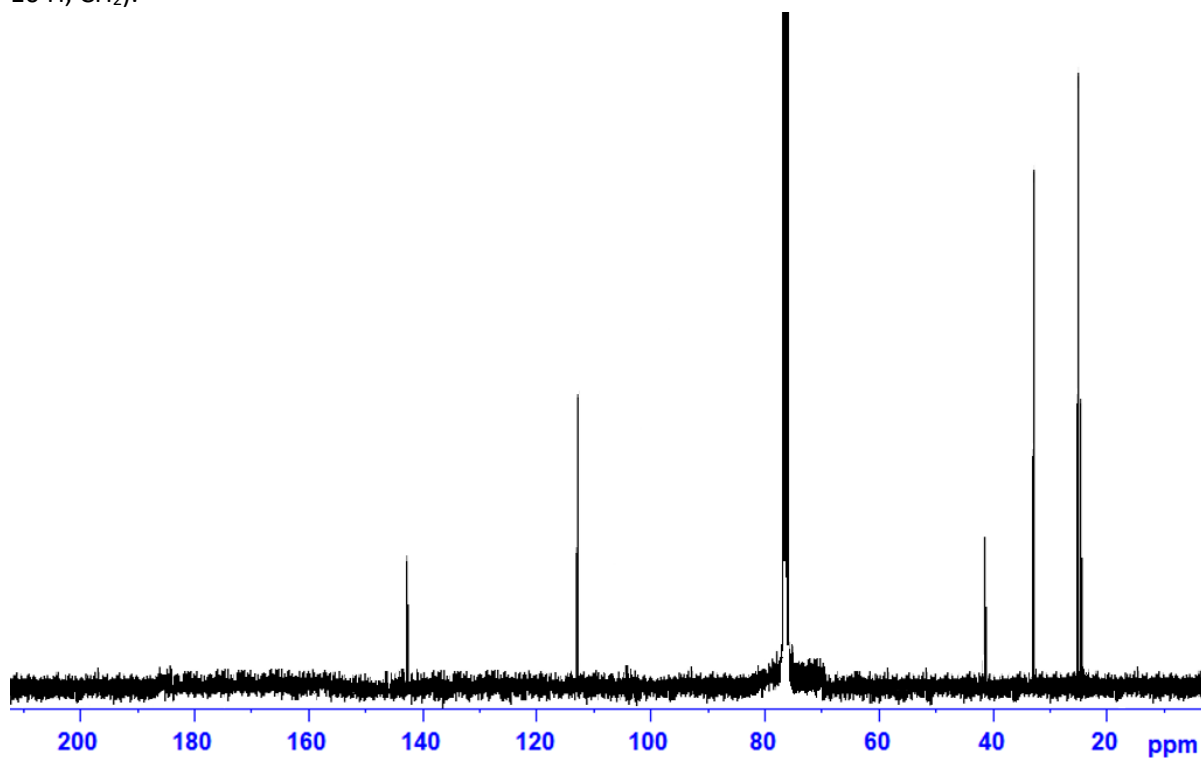


Figure SI#4-8. ^{13}C NMR of Vinylcyclohexane (**4**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 143.5 ($-\text{CH}=\text{}$), 112.7 ($=\text{CH}_2$), 41.8 ($-\text{CH}-$), 33.4 (2 x $-\text{CH}_2-$), 26.1 (2 x $-\text{CH}_2-$), 25.9 ($-\text{CH}_2-$).

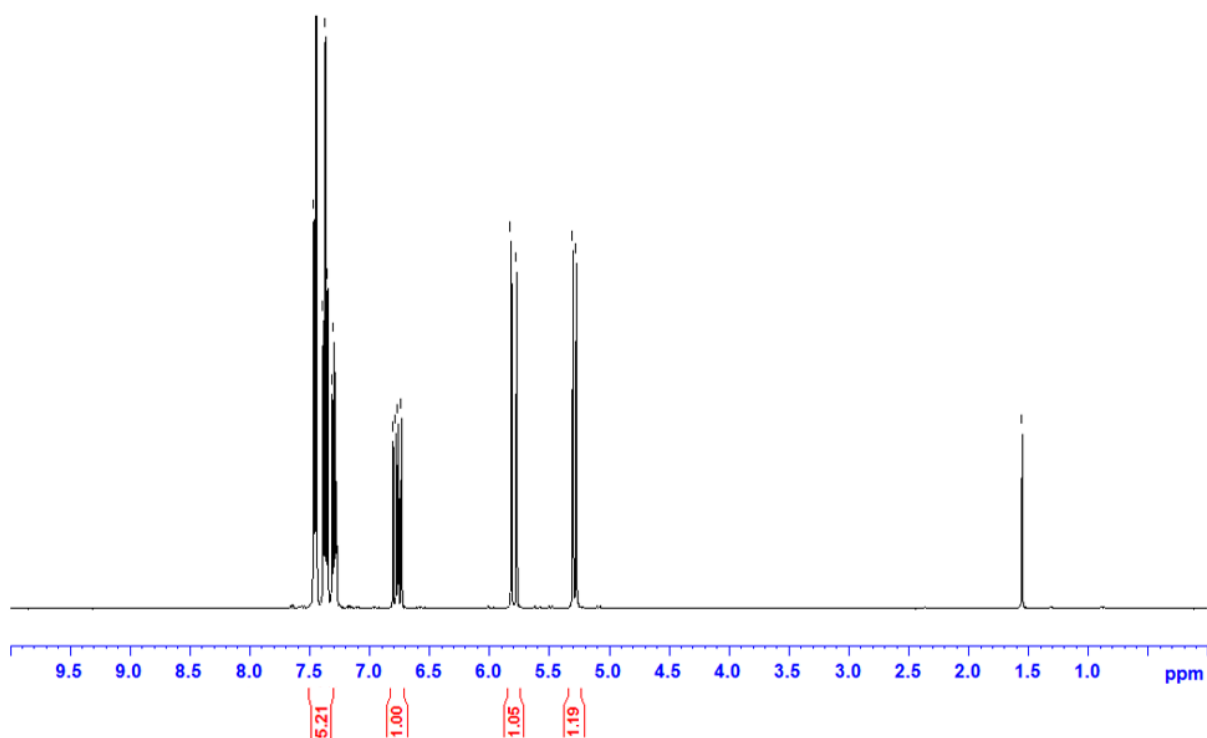


Figure SI#4-9. ^1H NMR of Styrene (**5**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 7.27-7.46 (m, 5H, CH Ar), 6.77 (m, 1H, -CH=), 5.79 (d, J = 16, 1H, =CH₂), 5.28 (d, J = 10, 1H, =CH₂).

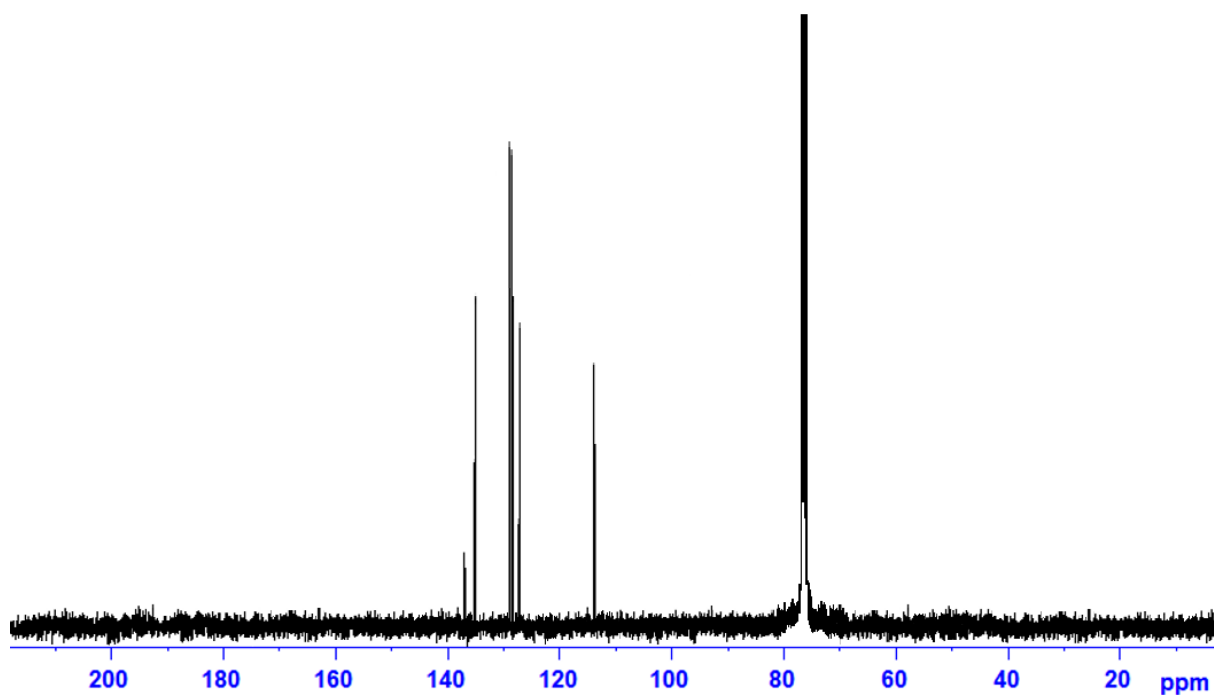


Figure SI#4-10. ^{13}C NMR of styrene (**5**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ =137.9 (-C-), 136.1 (-CH=), 128.6 (2 x -CH-), 128.5 (2 x -CH-), 127.9 (-CH-), 114.7 (=CH₂).

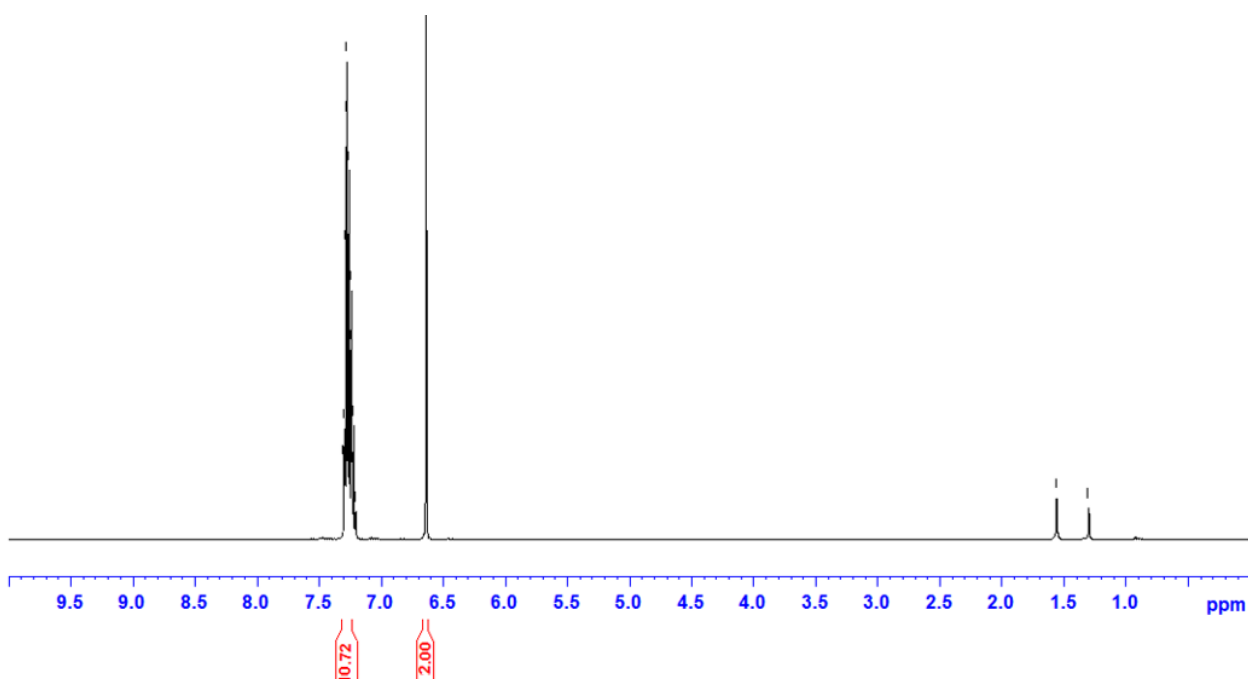


Figure SI#4-11. ^1H NMR of cis-stilbene (**6**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 7.20-7.30 (m, 10H, CH Ar), 6.64 (s, 2H, 2x -CH=).

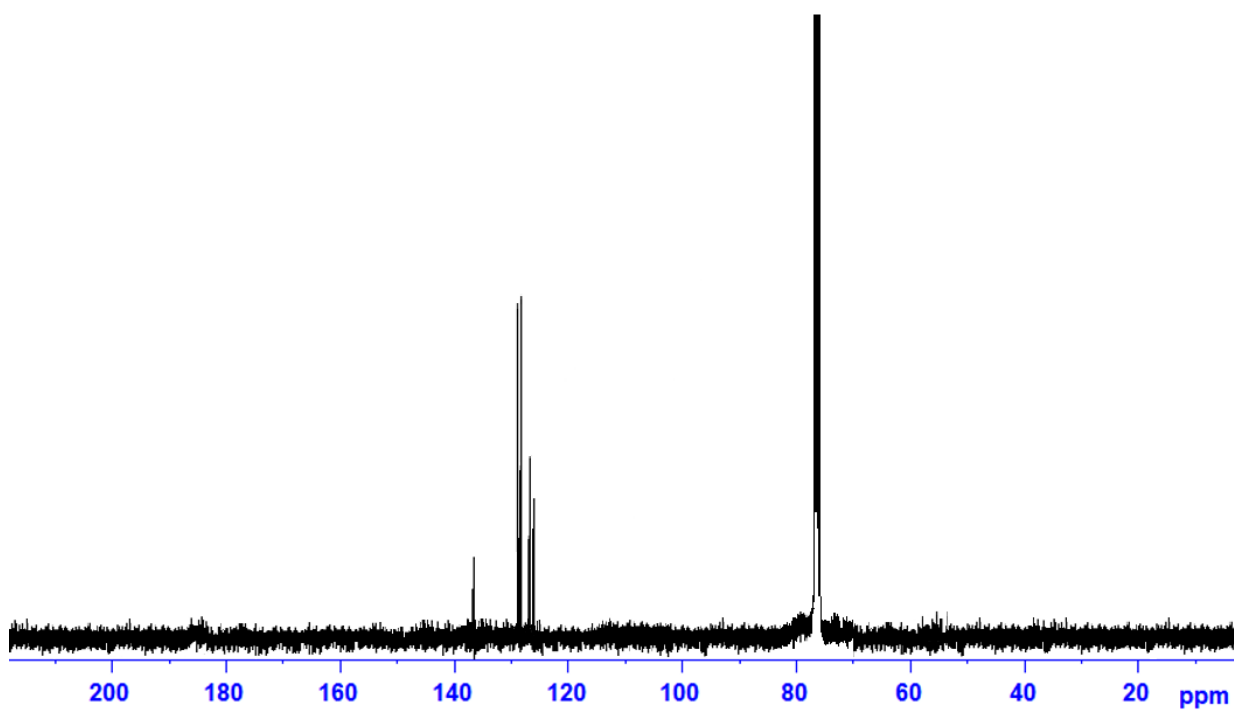


Figure SI#4-12. ^{13}C NMR of cis-stilbene (**6**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 137.5 (2 x -C-), 128.6 (4 x -CH-), 128.5 (4 x -CH-), 127.9 (2 x -CH-), 127.4 (2 x -CH=).

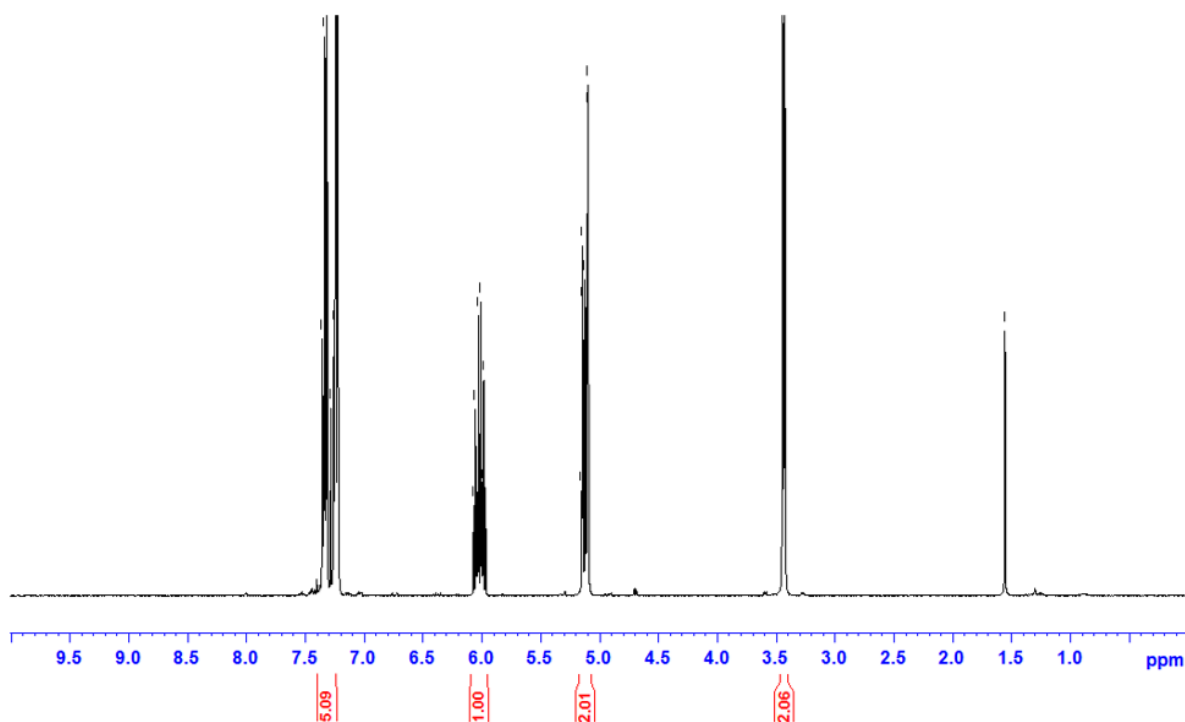


Figure SI#4-13. ^1H NMR of allylbenzene (**7**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 7.22-7.35 (m, 5H, CH Ar), 6.01 (m, 1H, -CH=), 5.12 (m, 2H, =CH₂), 3.43 (s, 2H, -CH₂-).

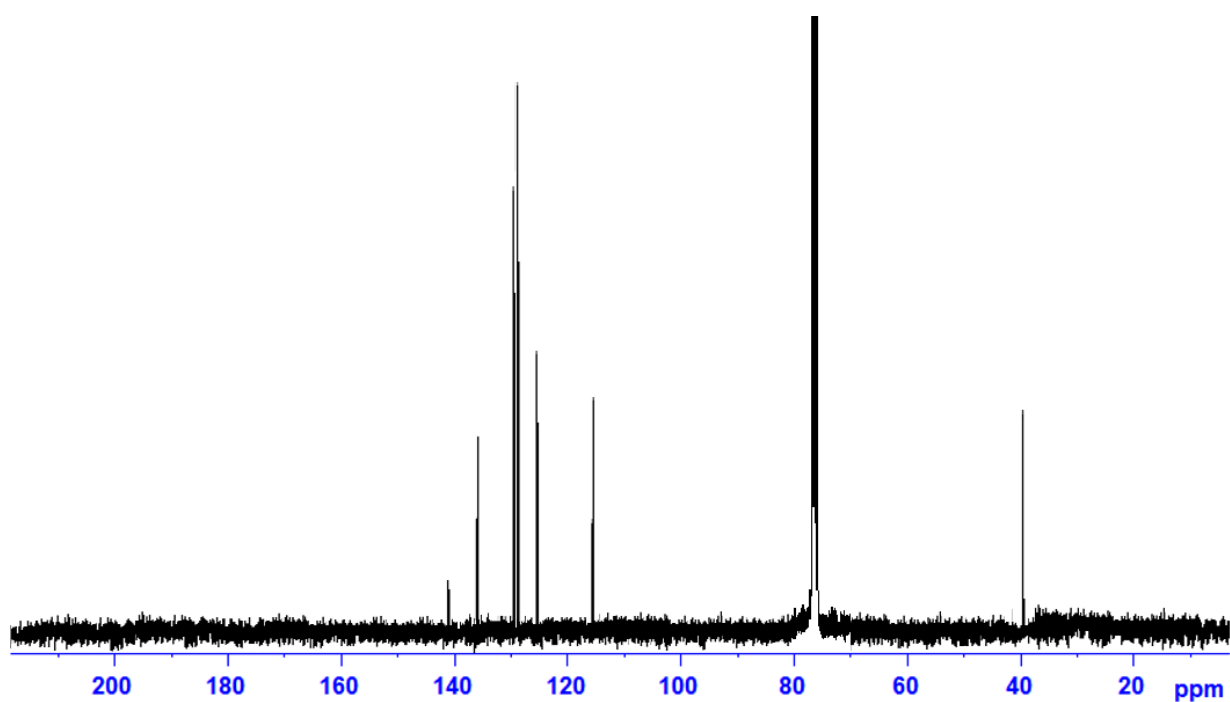


Figure SI#4-14. ^{13}C NMR of allylbenzene (**7**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 141.4 (-C-), 136.5 (-CH=), 129.0 (2 x -CH-), 128.6 (2 x -CH-), 125.7 (-CH-), 115.9 (=CH₂), 39.5 (-CH₂-).

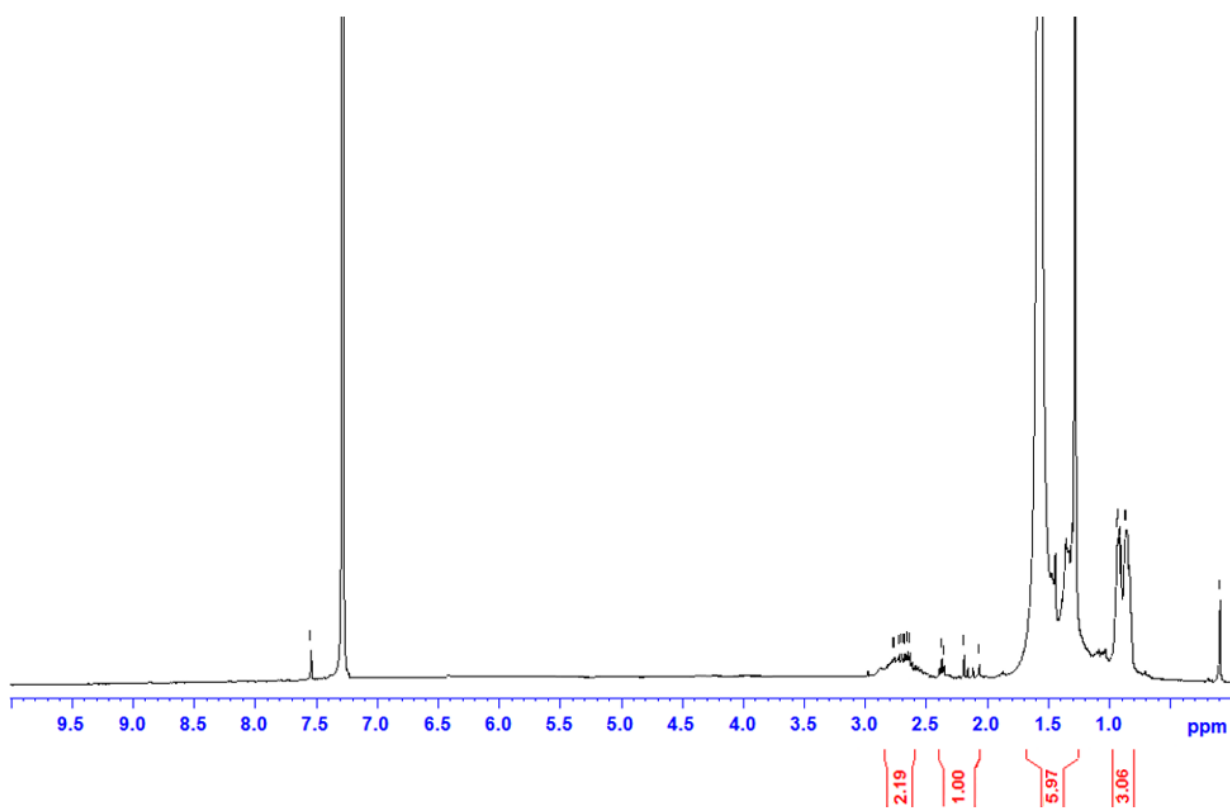


Figure SI#4-15. ^1H NMR of 1,2-epoxyhexane (**8**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 2.63-2.78 (m, 2H, O-CH), 2.15-2.37 (m, 1H, O- CH_2), 1.20-1.65 (m, 6H, 3 x- CH_2), 0.85-0.94 (m, 3H, CH_3 -).

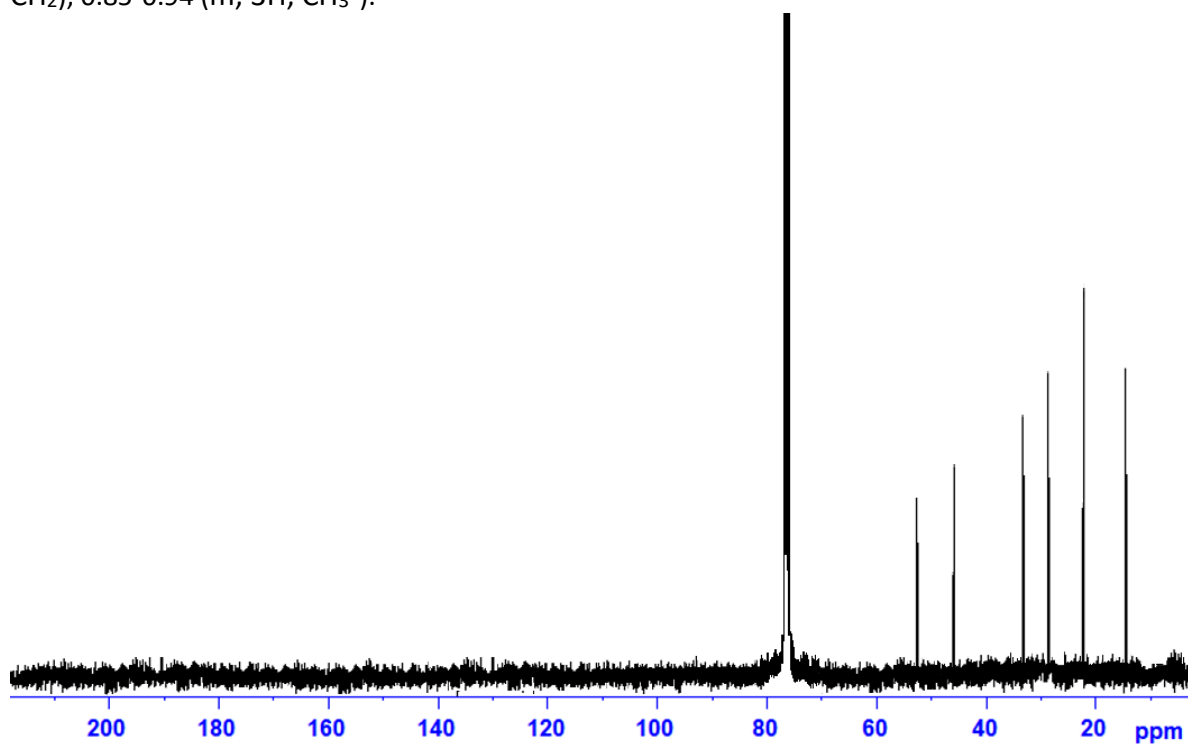


Figure SI#4-16. ^{13}C NMR of 1,2-epoxyhexane (**8**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 51.8 (O-CH), 46.4 (O- CH_2), 32.5, 28.4, 22.8 ($-\text{CH}_2-$), 14.1 (CH_3-).

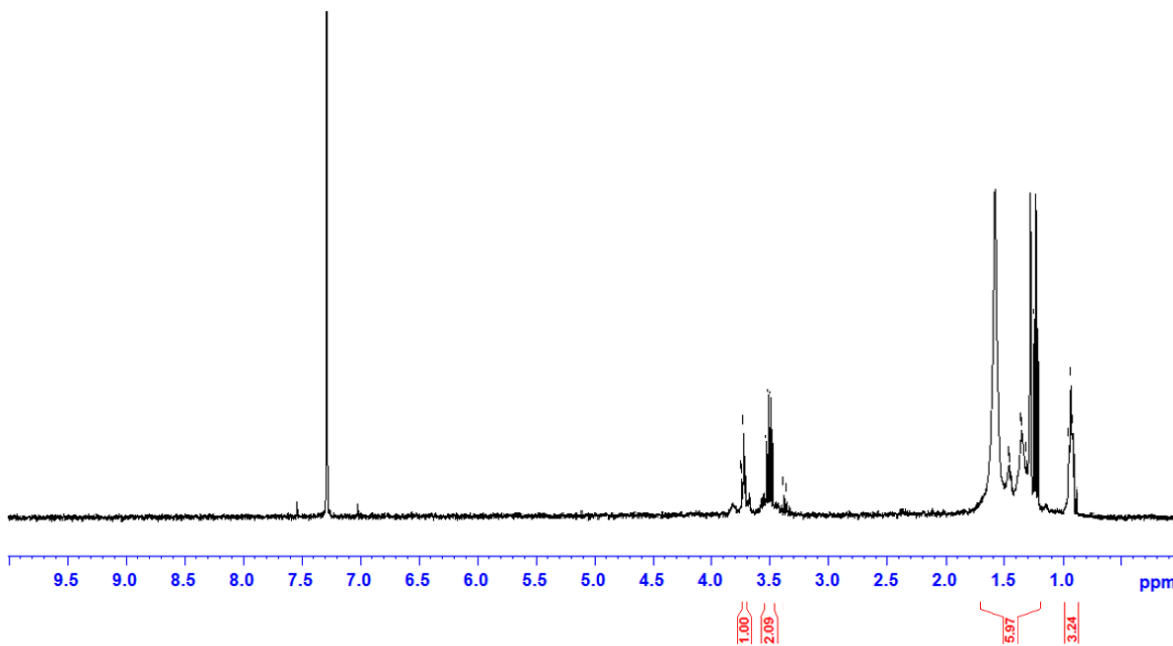


Figure SI#4-17. ^1H NMR of hexane-1,2-diol (**9**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 3.70 – 3.74 (m, 1H, CH-OH), 3.47-3.53 (m, 2H, CH_2 -OH), 1.34 – 1.65 (m, 6H, 3 \times - CH_2 -), 0.91 (m, 3H, CH_3 -).

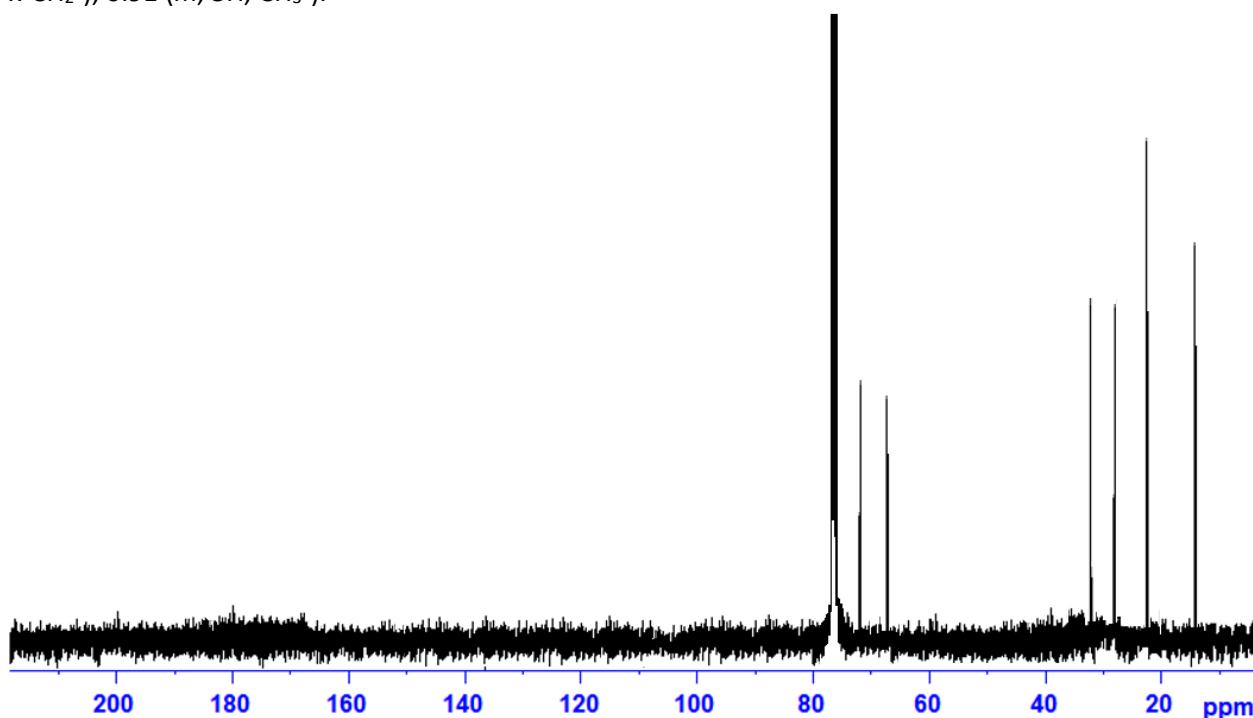


Figure SI#4-18. ^{13}C NMR of hexane-1,2-diol (**9**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 72.3 (CH-OH), 66.7 (CH_2 -OH), 32.7, 27.7, 22.7 ($-\text{CH}_2-$), 14.0 (CH_3-).

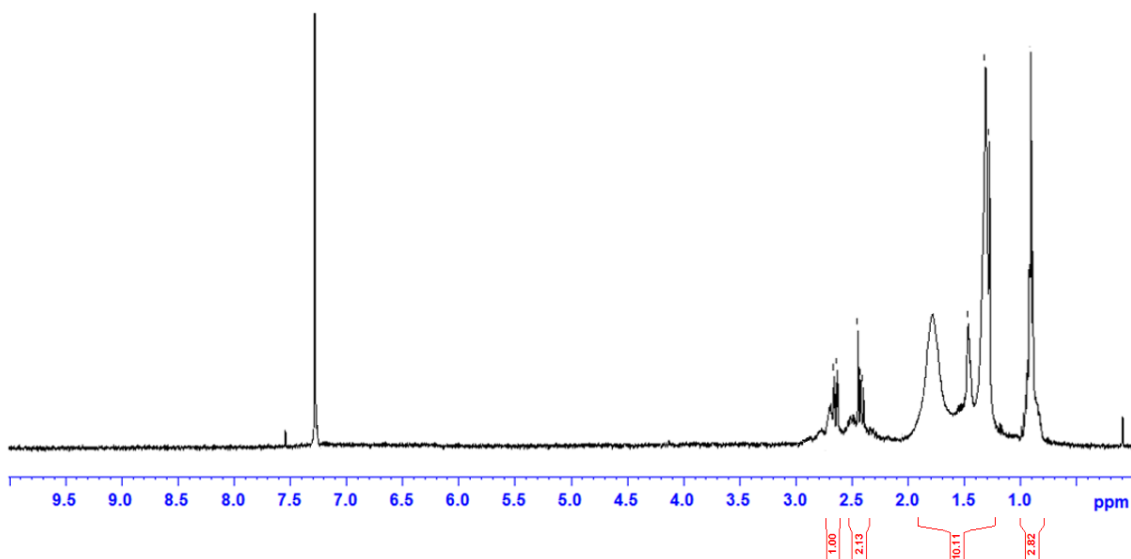


Figure SI#4-19. ^1H NMR of 1,2-Epoxyoctane (**10**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 2.61-2.74 (m, 1H, CH-O), 2.38-2.44 (m, 2H, CH_2 -OH), 1.27 - 1.90 (m, 10H, 5 x CH_2 -), 0.85-0.90(m, 3H, CH_3 -).

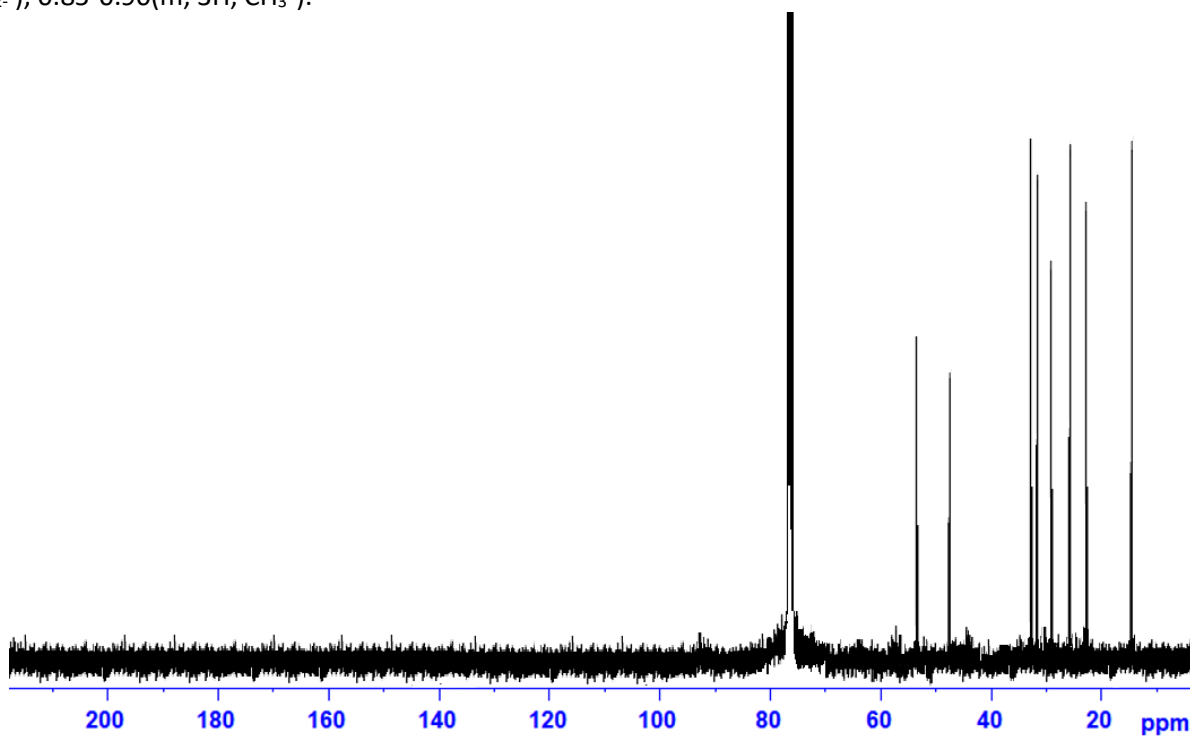


Figure SI#4-20. ^{13}C NMR of 1,2-Epoxyoctane (**10**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 52.3 (O-CH), 47.0 (O- CH_2), 32.5, 31.7, 29.1, 25.9, 22.5 ($-\text{CH}_2-$), 14.0 (CH_3-).

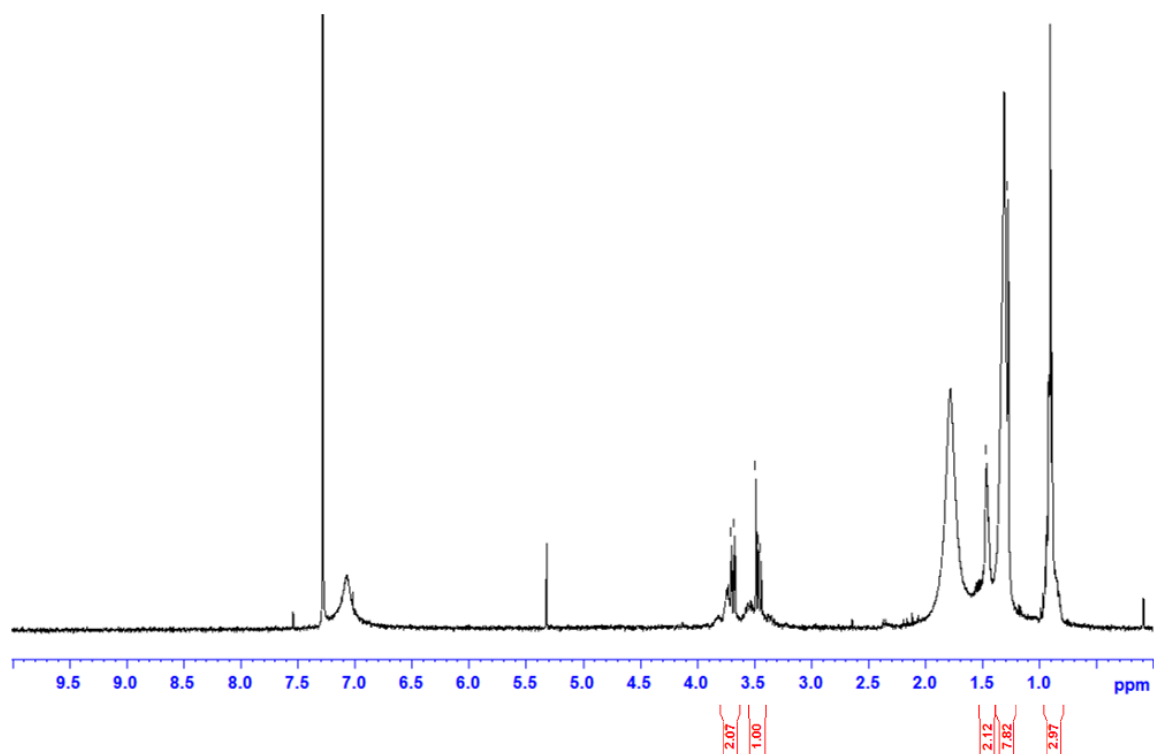


Figure SI#4-21. ^1H NMR of octane-1,2-diol (**11**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 3.62 – 3.70 (m, 2H, $\text{CH}_2\text{-OH}$), 3.43-3.48 (m, 1H, CH-OH), 1.45-1.50 (m, 2H, $-\text{CH}_2-$) 1.25-1.35 (m, 8H, $-\text{CH}_2-$), 0.90 (m, 3H, CH_3-).

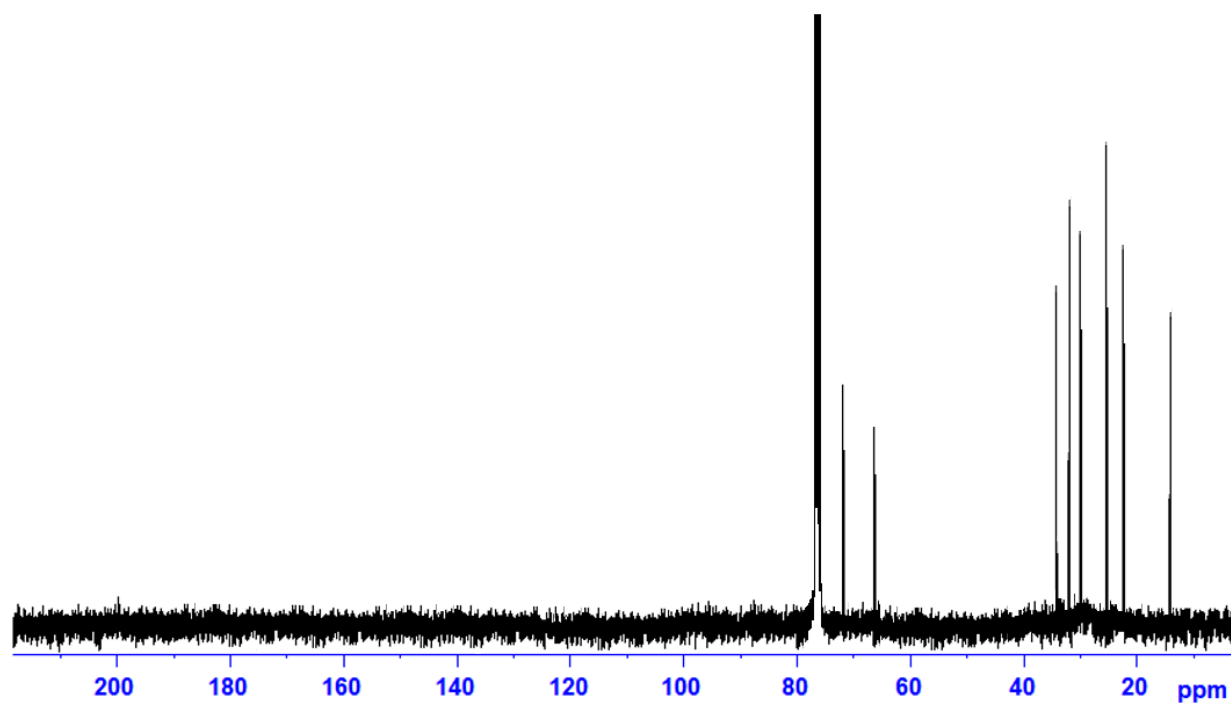


Figure SI#4-22. ^{13}C NMR of octane-1,2-diol (**11**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 72.4 (CH-OH), 66.8 ($\text{CH}_2\text{-OH}$), 33.1, 31.7, 29.3, 25.5, 22.6 ($-\text{CH}_2-$), 14.1 (CH_3-).

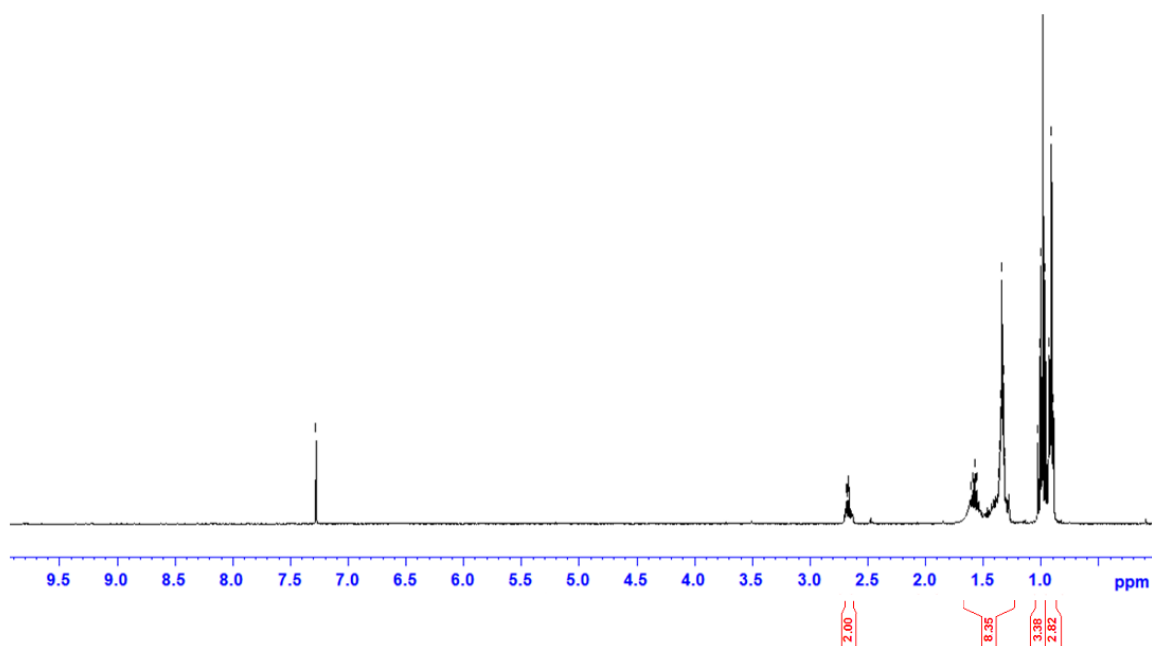


Figure SI#4-23. ^1H NMR of trans-3,4-epoxyoctane (**12**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz) δ =2.60-2.73 (m, 2H, 2 x CH-O), 1.28-1.62 (m, 8H, 4 x CH_2 -), 0.98 (m, 3H, CH_3 -), 0.88 (m, 3H, CH_3 -).

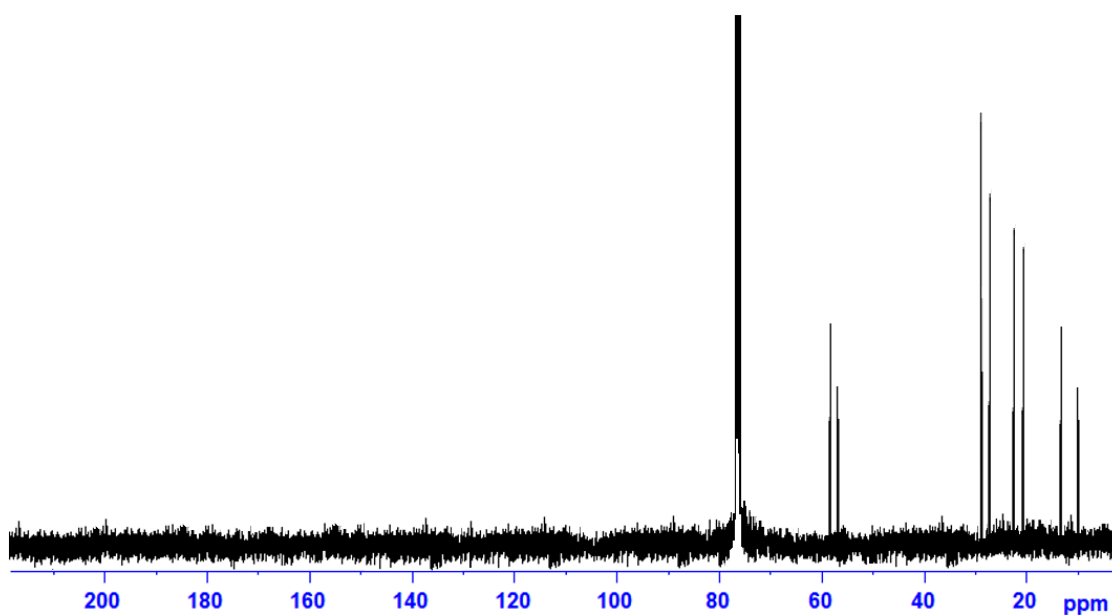


Figure SI#4-24. ^{13}C NMR of trans-3,4-epoxyoctane (**12**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 58.3 (O-CH), 57.3 (O-CH), 28.7, 27.3, 22.6, 21.0, (CH_2 -), 13.9 (CH_3 -), 10.5 (CH_3 -).

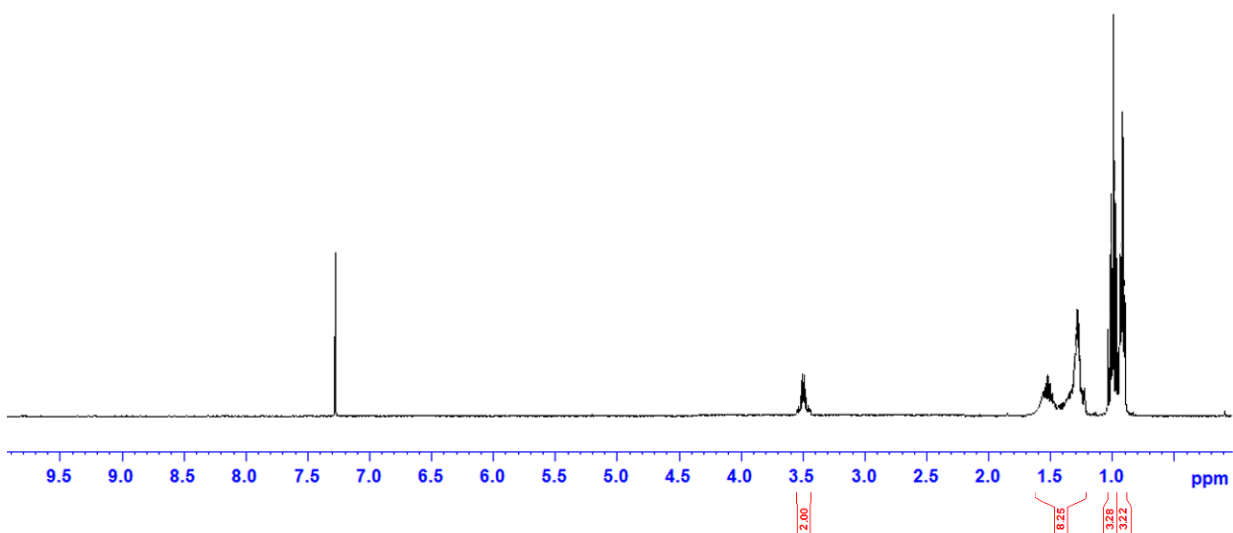


Figure SI#4-25. ^1H NMR of octane-3,4-diol (**13**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 3.43-3.55 (m, 2H, 2 x CH-OH), 1.21-1.55 (m, 8H, 4 x CH_2 -), 1.08 (m, 3H, CH_3 -), 0.83-0.95 (m, 3H, CH_3 -).

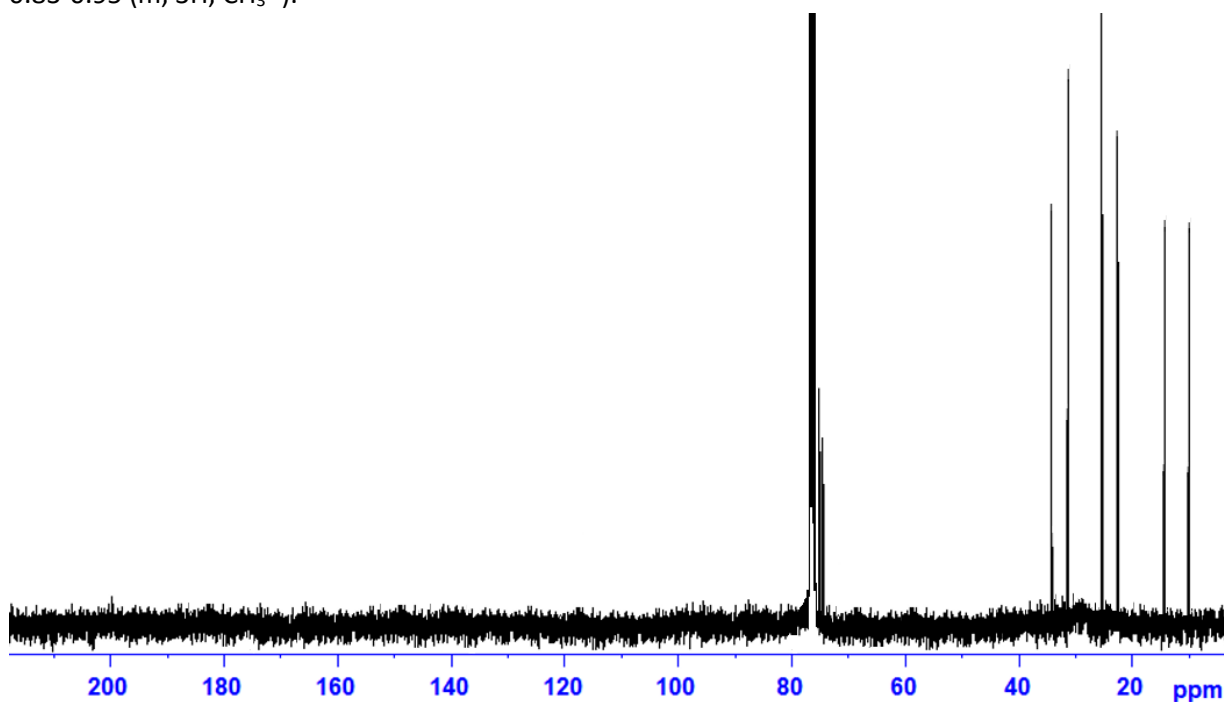


Figure SI#4-26. ^{13}C NMR of octane-3,4-diol (**13**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 76.2 (CH-OH), 75.3 (CH-OH), 33.8, 31.7, 25.2, 22.5 (CH_2 -), 13.9 (CH_3 -), 10.5 (CH_3 -).

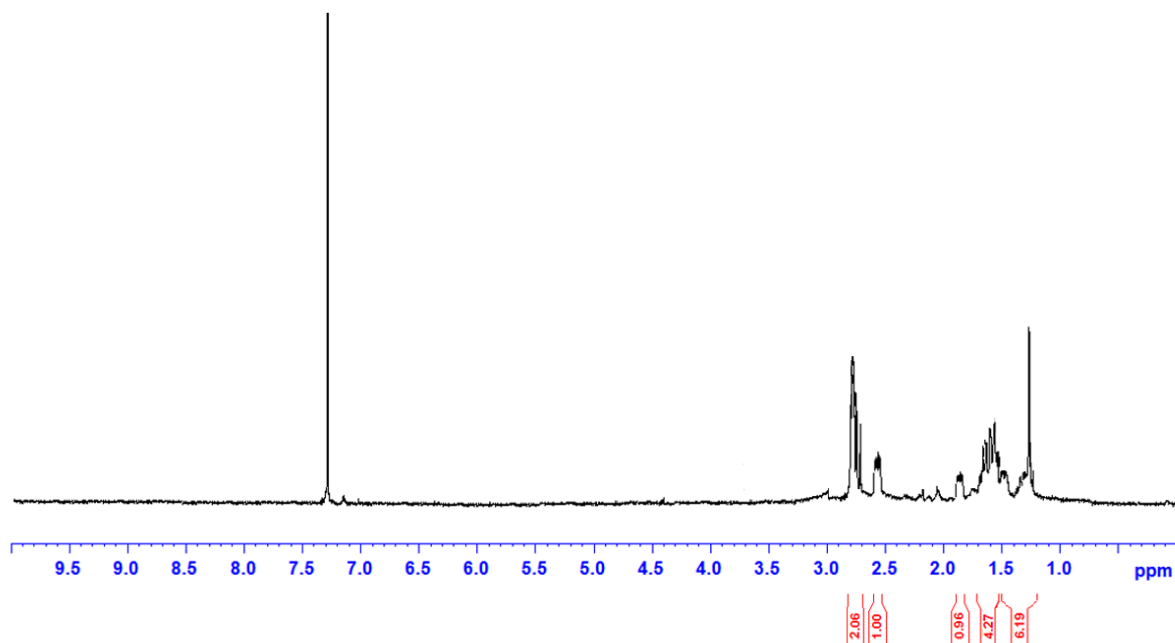


Figure SI#4-27. ^1H NMR of 2-cyclohexyloxirane (**14**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 2.60–2.72 (m, 2H, $\text{CH}_2\text{-O}$), 2.51–2.54 (m, 1H, CH-O), 1.84-1.89 (m, 1H, CH), 1.53-1.72 (m, 4H, 2 x CH_2), 1.20-1.52 (m, 6H, 3 x CH_2).

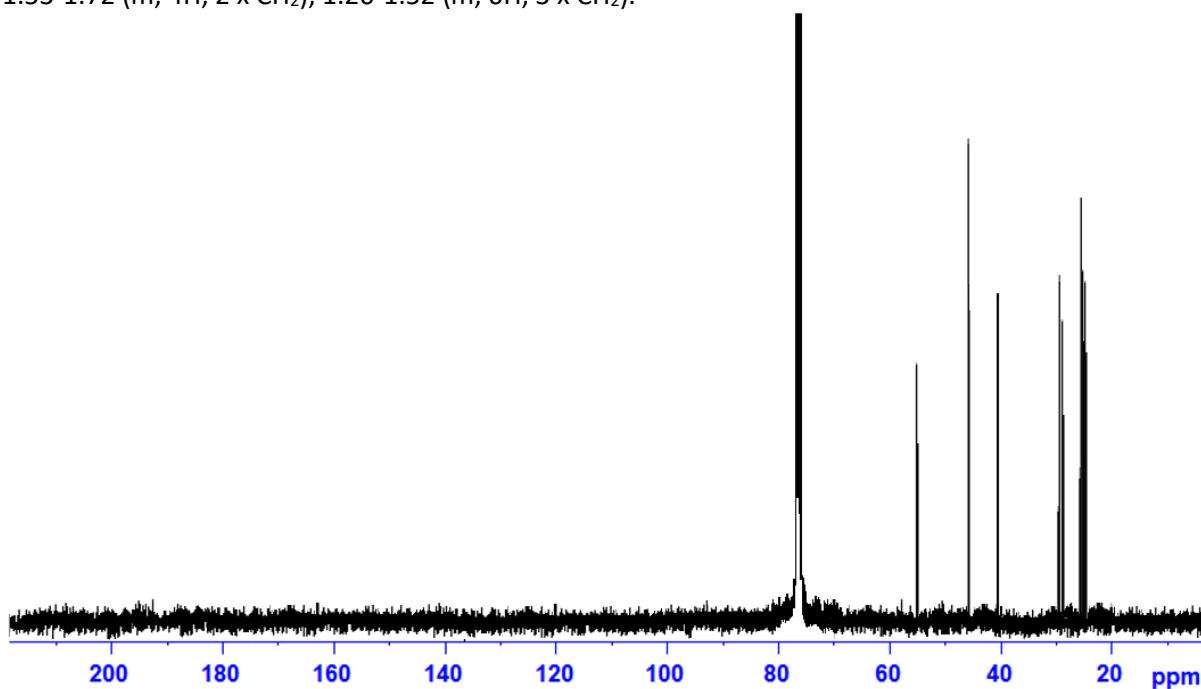


Figure SI#4-28. ^{13}C NMR of 2-cyclohexyloxirane (**14**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 56.6 (O-CH), 46.0 (O- CH_2), 40.4 (-CH-), 29.7, 28.8, 26.3, 25.7, 25.5 (- CH_2 -).

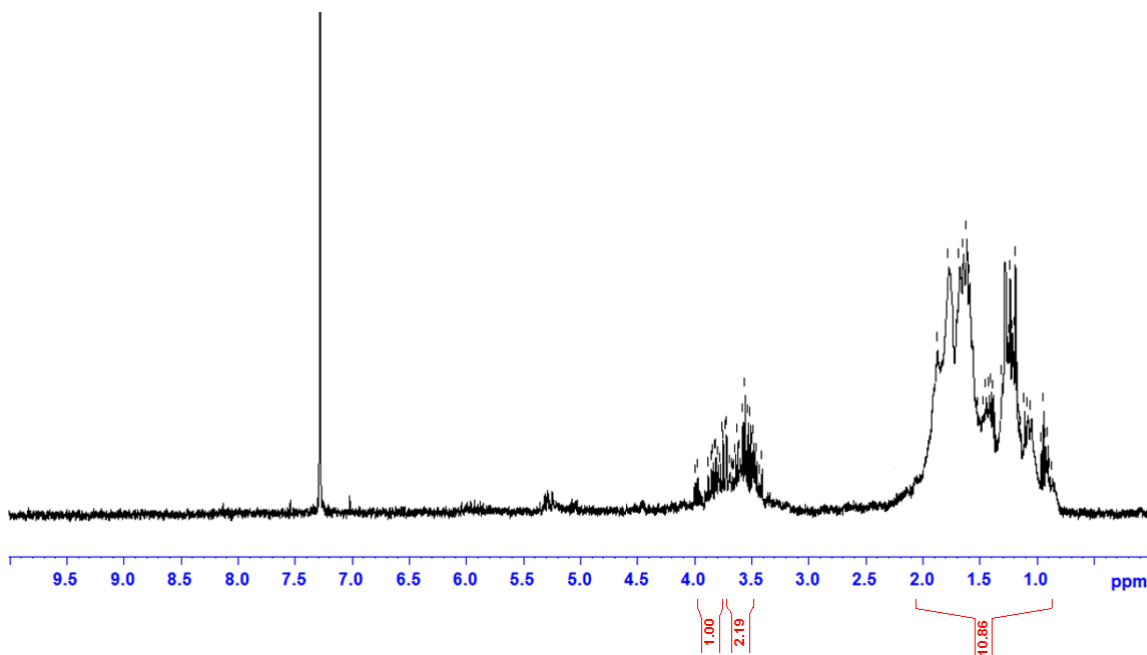


Figure SI#4-29. ^1H NMR of 1-cyclohexylethane-1,2-diol (**15**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 3.76-3.93 (m, 1H, CH-OH), 3.47-3.75 (m, 2H, CH_2 -OH), 0.85-1.88 (m, 11H).

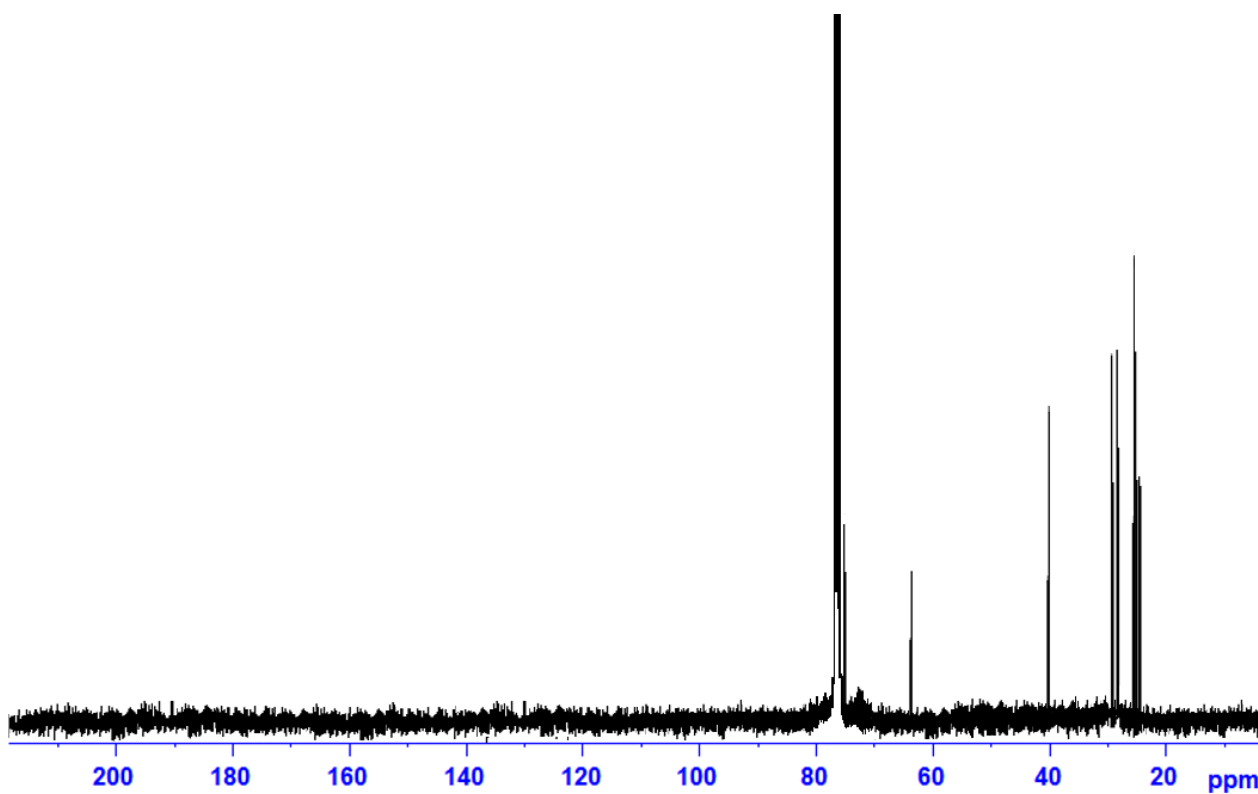


Figure SI#4-30. ^{13}C NMR of 1-cyclohexylethane-1,2-diol (**15**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 76.5 (CH-OH), 64.7 (CH_2 -OH), 40.7 (-CH-), 29.0, 28.6, 26.3, 26.1, 26.0 ($-\text{CH}_2-$).

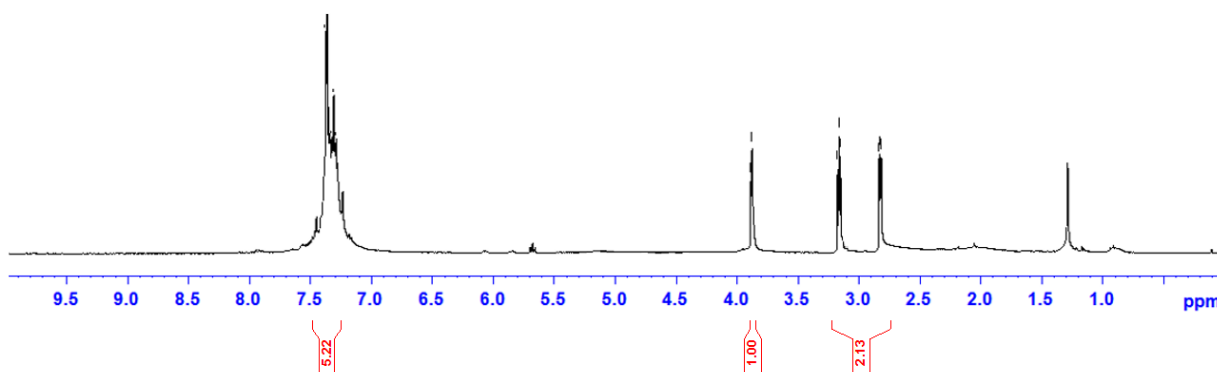


Figure SI#4-31. ^1H NMR of 2-phenyloxirane (**16**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): $\delta = 7.29\text{--}7.38$ (m, 5H, CH-Ar), 3.88 (m, 1H, CH-O), 2.82-3.16(dd, 2H, $J = 4.0$ Hz, $\text{CH}_2\text{-O}$).

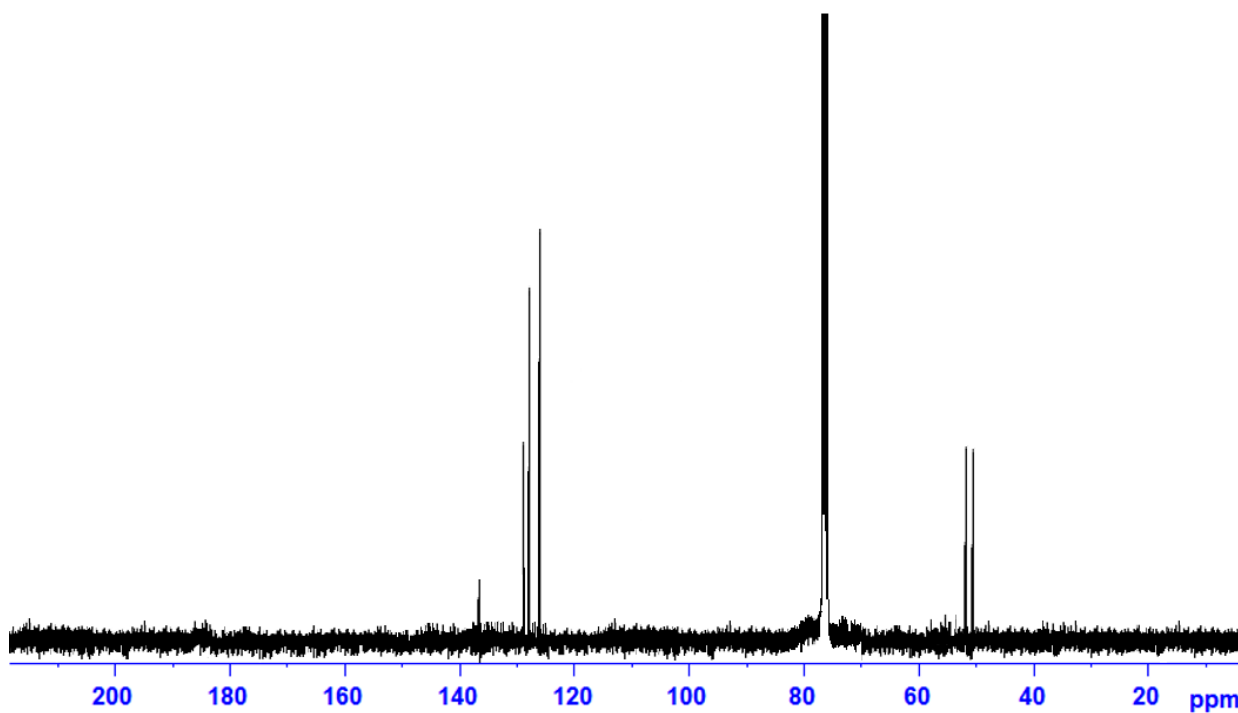


Figure SI#4-32. ^{13}C NMR of 2-phenyloxirane (**16**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): $\delta = 137.5$ (-C-), 128.4 (-CH-), 128.1 (2 x -CH-), 125.4 (2 x -CH-), 52.2 (O-CH), 51.1 (O- CH_2).

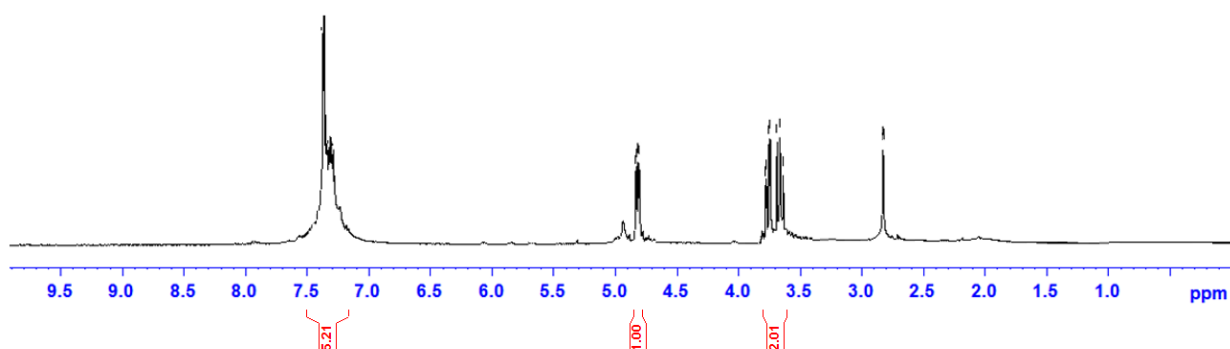


Figure SI#4-33. ^1H NMR of 1-phenylethane-1,2-diol (**17**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 7.29 – 7.38 (m, 5H, CH-Ar), 4.80-4.85 (m, 1H, CH-OH), 3.62-3.79 (m, 2H, CH_2 -OH).

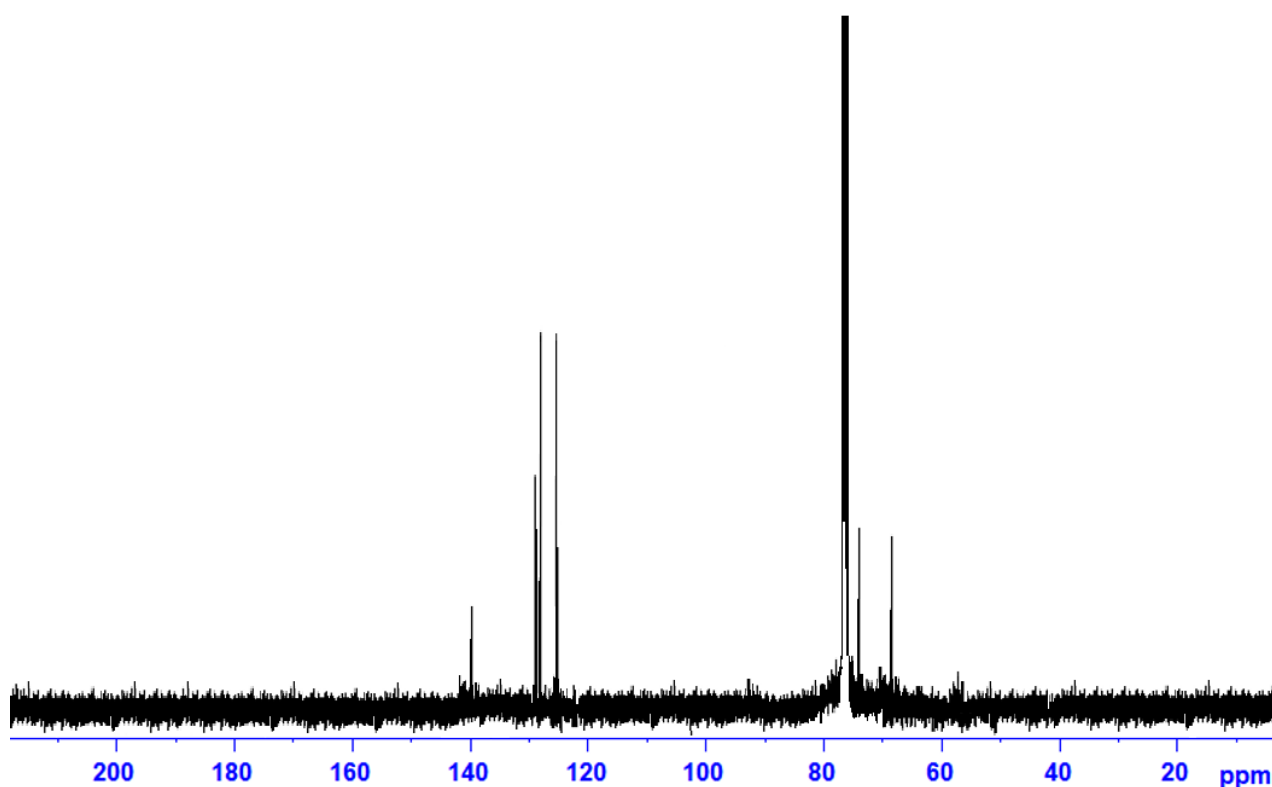


Figure SI#4-34. ^{13}C NMR of 1-phenylethane-1,2-diol (**17**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 140.4 (-C-), 128.5 (-CH-), 128.0 (2 x -CH-), 126.1 (2 x -CH-), 74.7 (CH-OH), 68.71 (CH_2 -OH).

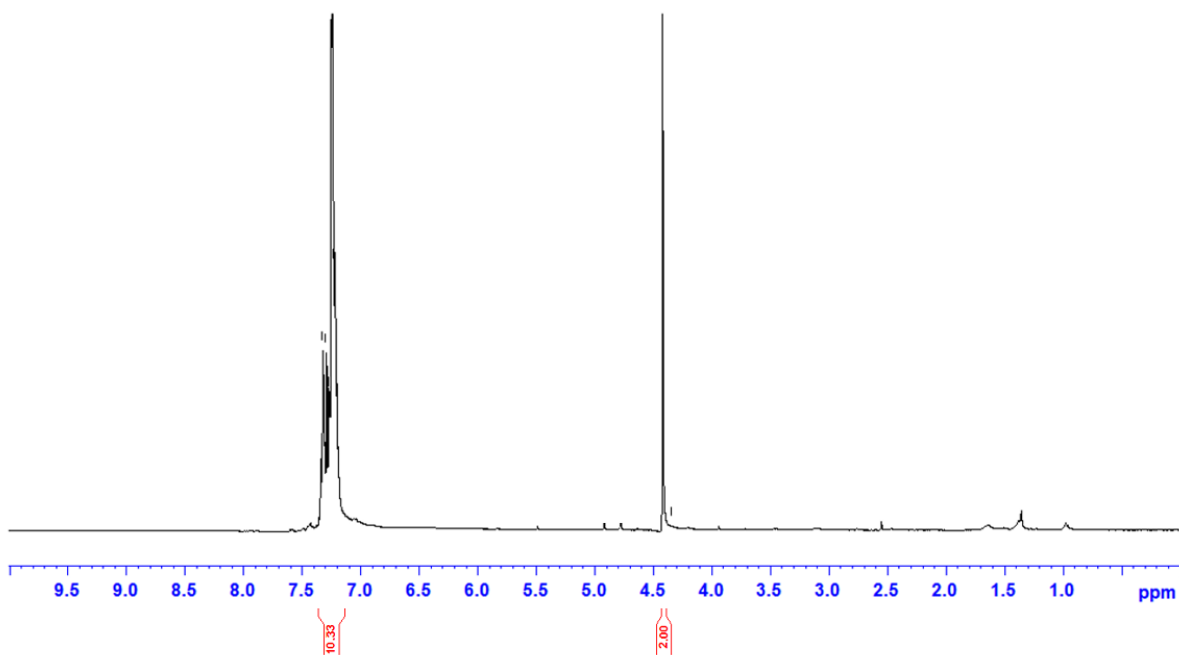


Figure SI#4-35. ^1H NMR of 2,3-Diphenyloxirane (**18**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 7.18-7.32 (m, 10H, CH-AR), 4.40 (s, 2H, 2 x CH-O).

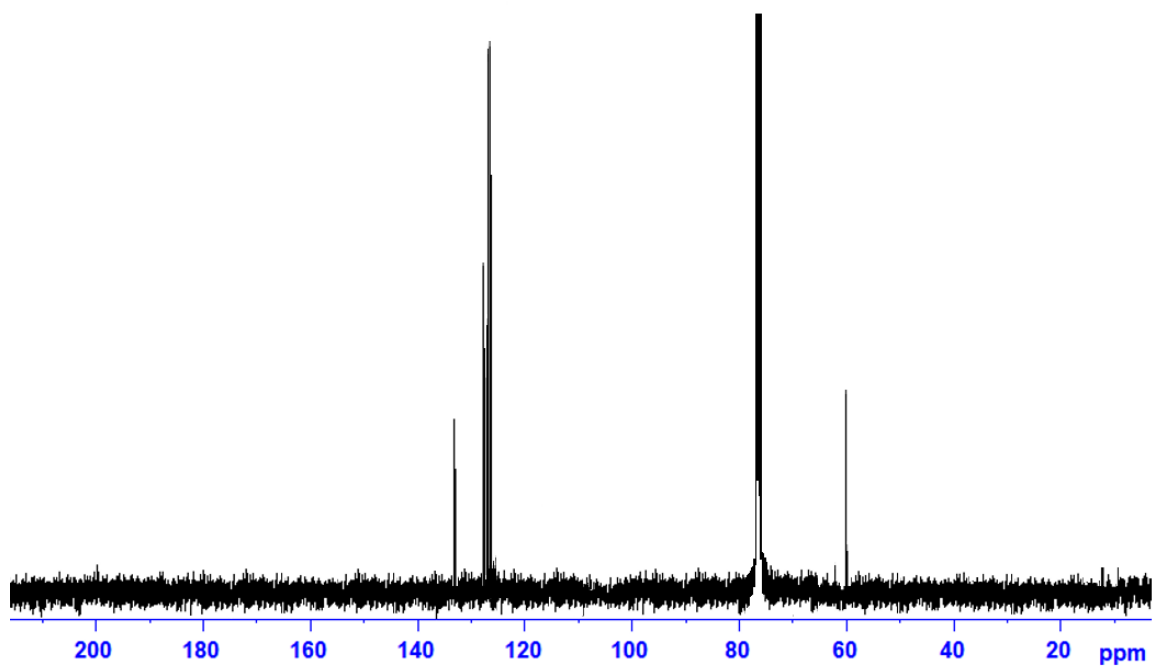


Figure SI#4-36. ^{13}C NMR of 2,3-Diphenyloxirane (**18**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 134.5 (2 x -C-), 127.9 (2 x -CH-), 127.6 (4 x -CH-), 127.0 (4 x -CH-), 59.9 (O-CH).

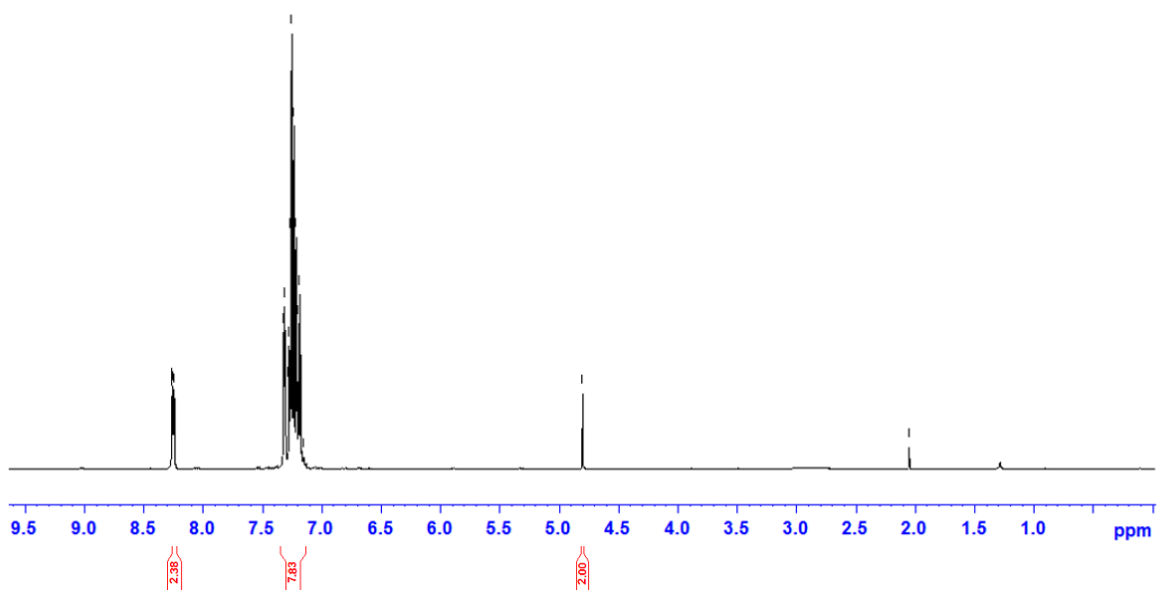


Figure SI#4-37. ^1H NMR of 1,2-diphenylethane-1,2-diol (**19**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): $\delta = 8.21\text{-}8.27$ (m, 2H, CH-Ar), $7.15\text{-}7.35$ (m, 8H, CH-Ar), 4.78 (s, 2H, 2 x CH-OH).

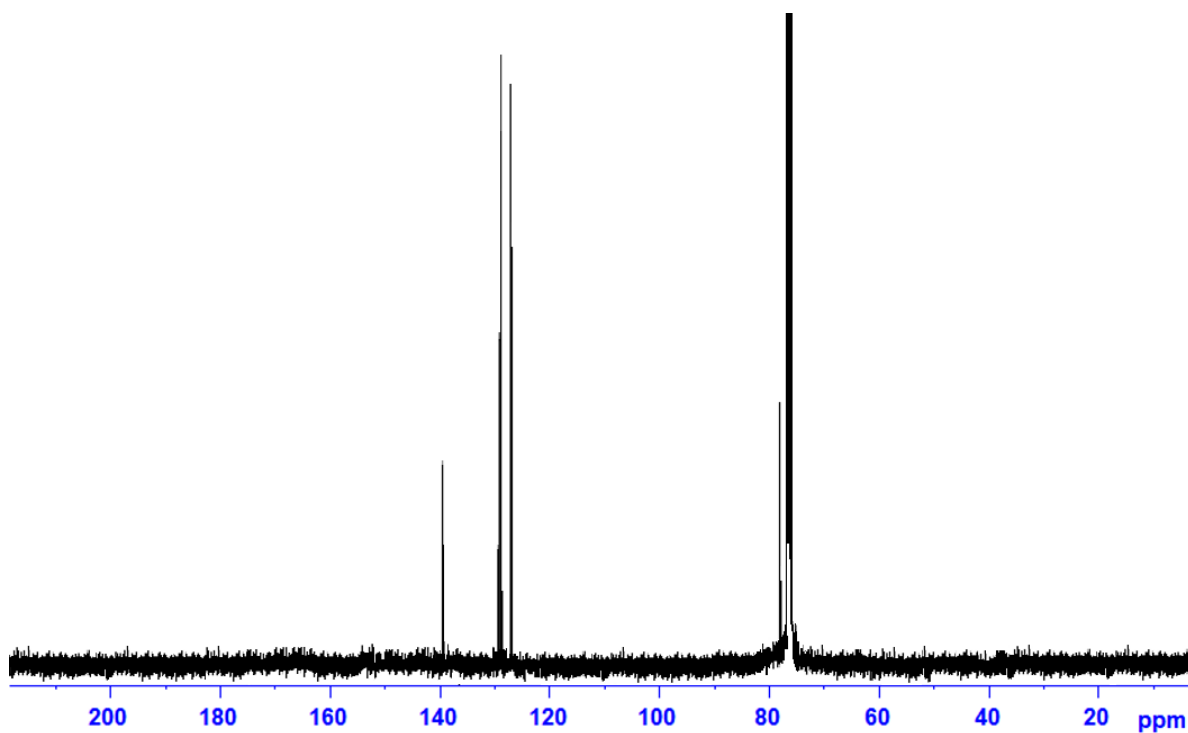


Figure SI#4-38 ^{13}C NMR of 1,2-diphenylethane-1,2-diol (**19**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): $\delta = 139.8$ (2 x -C-), 128.2 (2 x -CH-), 128.1 (4 x -CH-), 127.0 (4 x -CH-), 78.1 (2 x CH-OH).

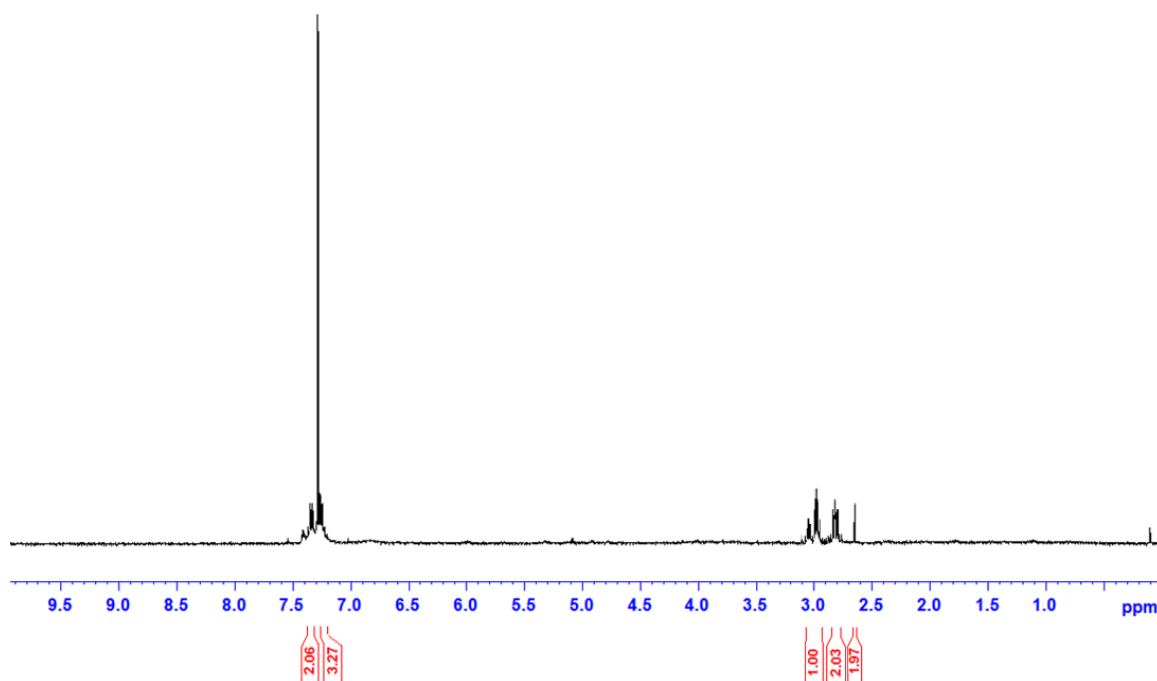


Figure SI#4-39. ^1H NMR of 2-benzyloxirane (**20**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 7.2–7.38 (m, 5H, CH-Ar), 2.93–3.07 (m, 1H, CH-O), 2.79–2.81 (m, 2H, CH_2 -), 2.64 (m, 2H, CH_2 -O).

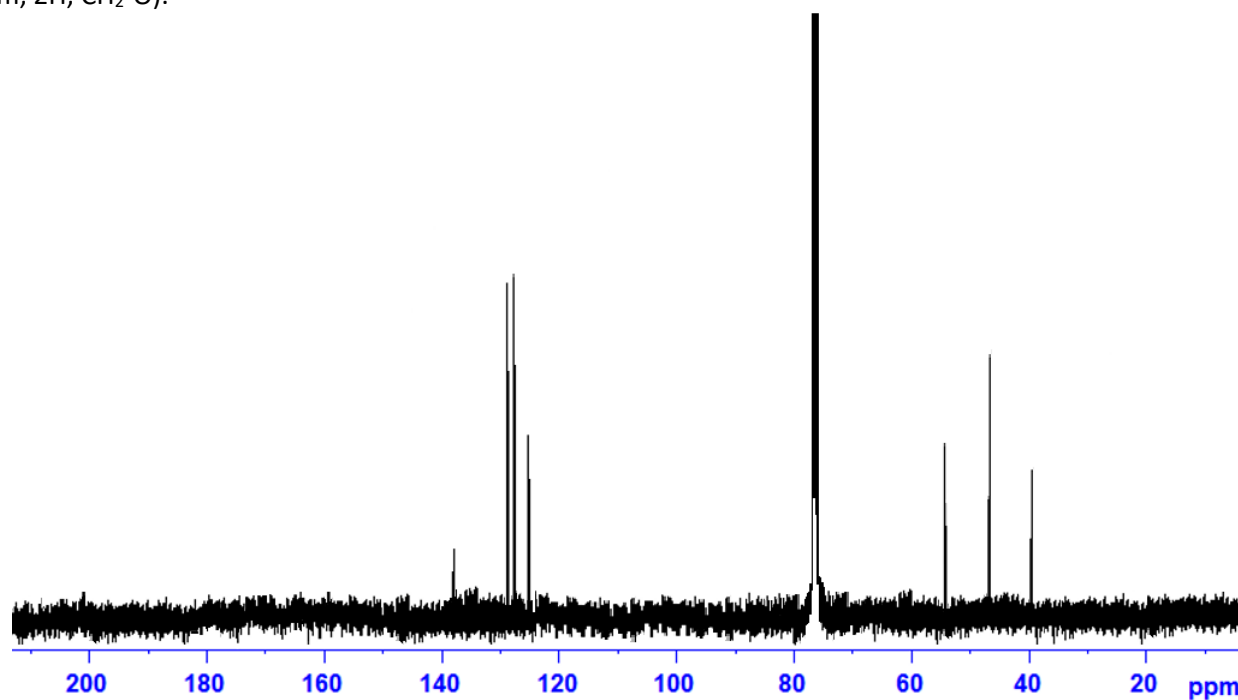


Figure SI#4-40. ^{13}C NMR of 2-benzyloxirane (**20**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 138.0 (-C-), 128.8 (2 x -CH-), 128.1 (2 x -CH-), 126.0 (-CH-), 54.5 (O-CH), 47.5 (O- CH_2), 39.6 (- CH_2 -).

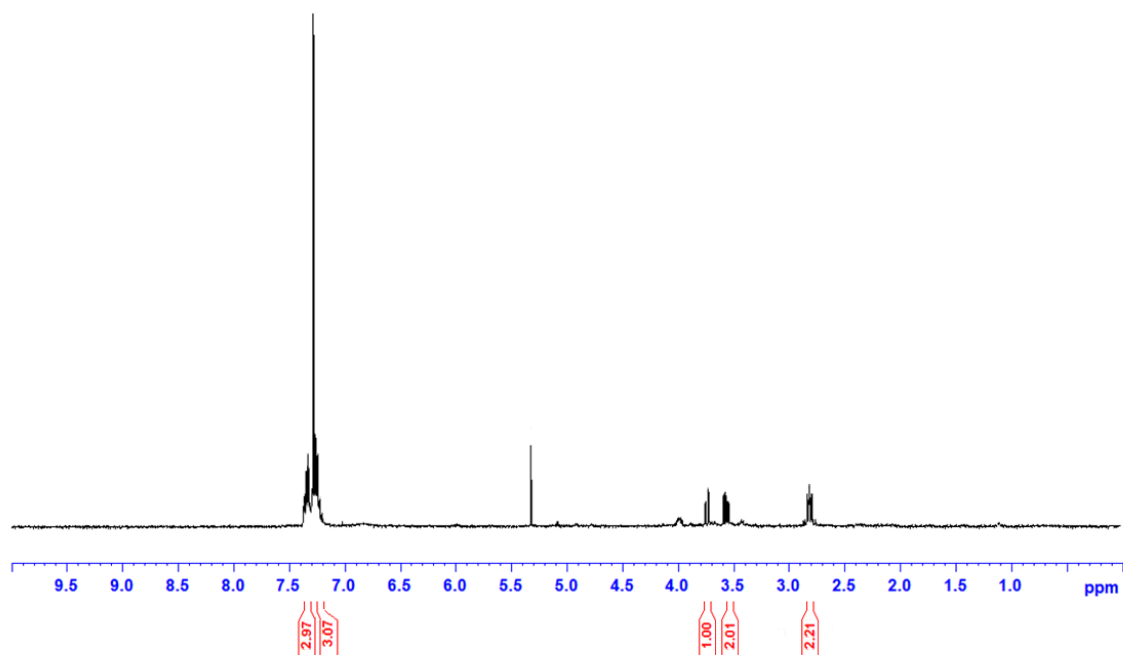


Figure SI#4-41. ^1H NMR of 3-phenylpropane-1,2-diol (**21**), (CDCl_3 , 400 MHz).

^1H NMR (CDCl_3 , 400 MHz): δ = 7.23- 7.38 (m, 5H, CH-Ar), 3.70-3.75 (m, 1H, CH-OH), 3.54-3.58 (m, 2H, CH_2 -OH), 2.80-2.83 (m, 2H, CH_2 -).

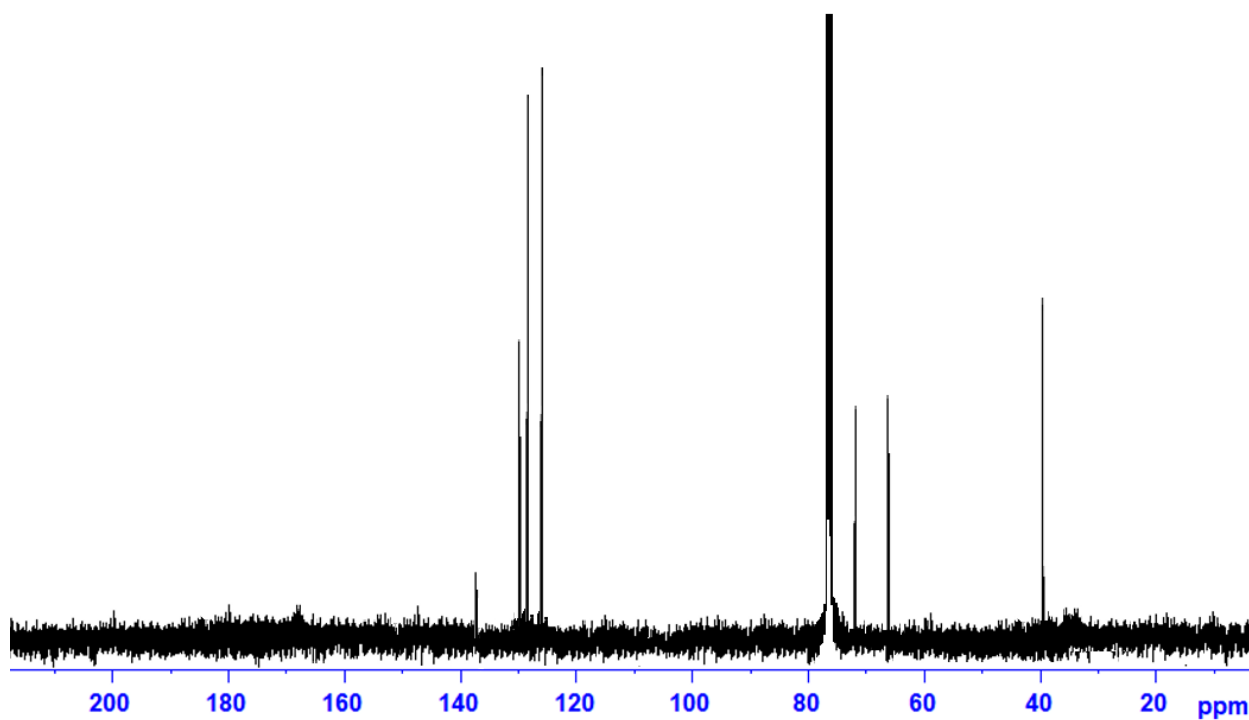


Figure SI#4-42. ^{13}C NMR of 3-phenylpropane-1,2-diol (**21**), (CDCl_3 , 100 MHz).

^{13}C NMR (CDCl_3 , 100 MHz): δ = 137.7 (-C-), 129.6 (-CH-), 128.6 (2 x -CH-), 126.6 (2 x -CH-), 73.0 (CH-OH), 66.0 (CH_2 -OH), 39.8 (- CH_2 -).

SI#5 Far IR of IVD and VG compared to the reference MTO-1,2-cyclohexanediamine complex

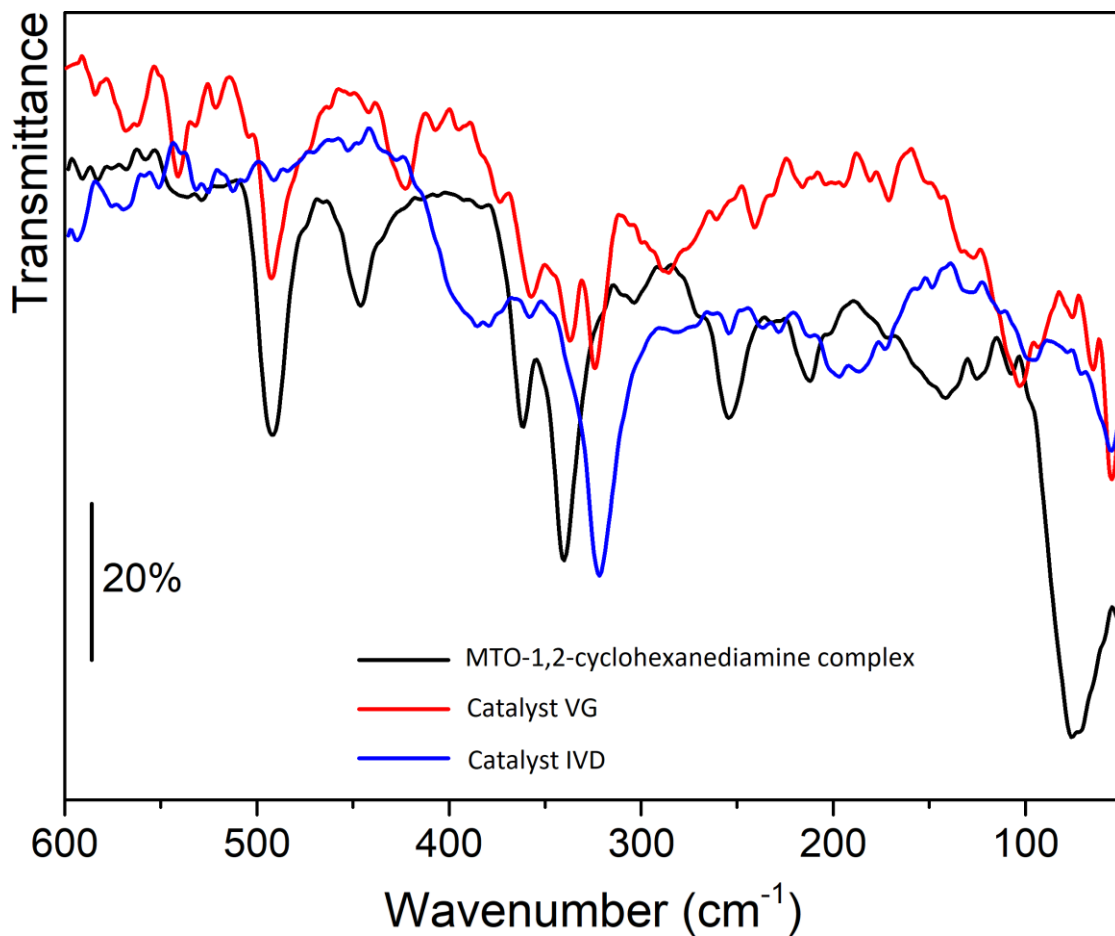


Figure SI#5-1. Far IR of IVD (blue) and VG (red) compared to the reference MTO-1,2-cyclohexanediamine complex (black)

SI#6 Transmission electron microscopy (TEM) of IV D and V H.

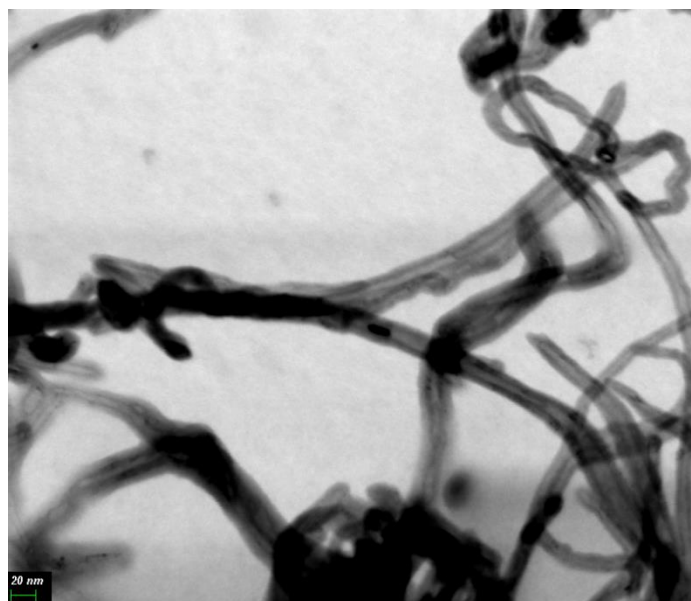


Figure SI#6-1. Transmission electron microscopy (TEM) of V F

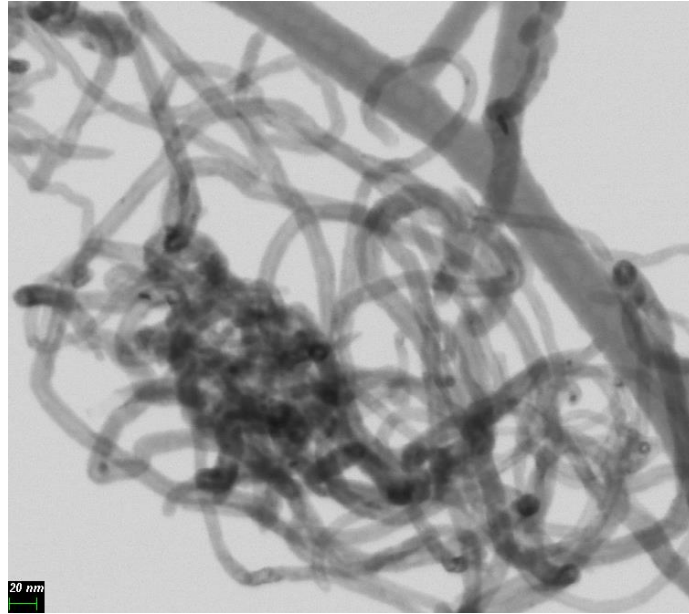


Figure SI#6-2. Transmission electron microscopy (TEM) of **V H**