

**Rediscovering Silicones. Molecularly Smooth, Low Surface Energy,
Unfilled, UV/vis-Transparent, Extremely Crosslinked,
Thermally Stable, Hard and Elastic PDMS**

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Solid-State NMR: ^{13}C and ^{29}Si spectra were recorded using a Bruker DSX300 spectrometer. $\text{D}_4^{\text{H}}\text{-D}_4^{\text{V}}\text{-1-1}$: ^{13}C NMR (75 MHz) -0.312 (CH_3), 8.899 (CH_2), 137.378 ($\text{CH}=\text{CH}_2$); ^{29}Si NMR (59 MHz) -35.770 (Si-C-C , $\text{Si-C}=\text{C}$, and Si-C), -21.326 (Si-C-C-Si). $\text{D}_4^{\text{H}}\text{-D}_4^{\text{V}}\text{-2-1}$: ^{13}C NMR (75 MHz) -1.087 (CH_3), 8.341 (CH_2); ^{29}Si NMR (59 MHz) -36.231 (Si-C-C and Si-C), -21.134 (Si-C-C-Si).

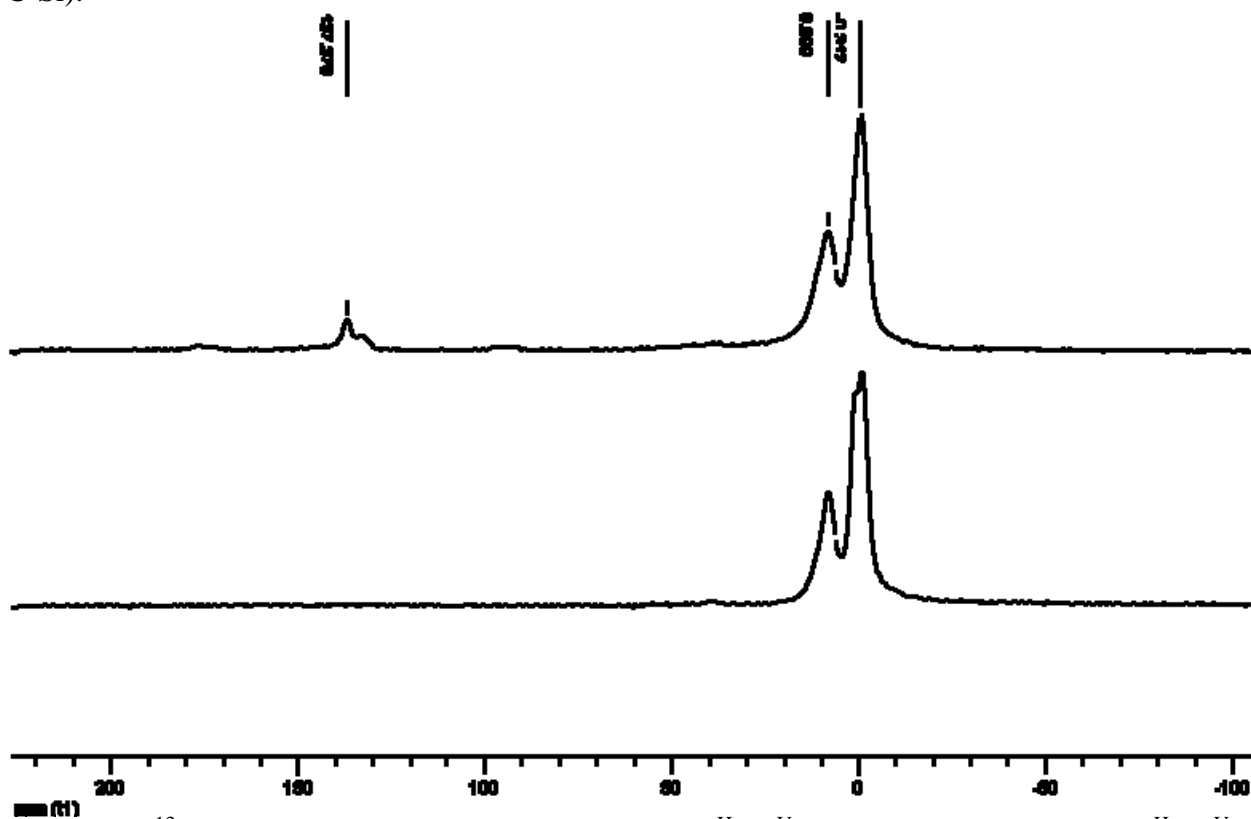


Figure S1. ^{13}C NMR (75 MHz) spectra of silicones: $\text{D}_4^{\text{H}}\text{-D}_4^{\text{V}}$ 1:1 molar ratio (top) and $\text{D}_4^{\text{H}}\text{-D}_4^{\text{V}}$ 2:1 molar ratio (bottom).

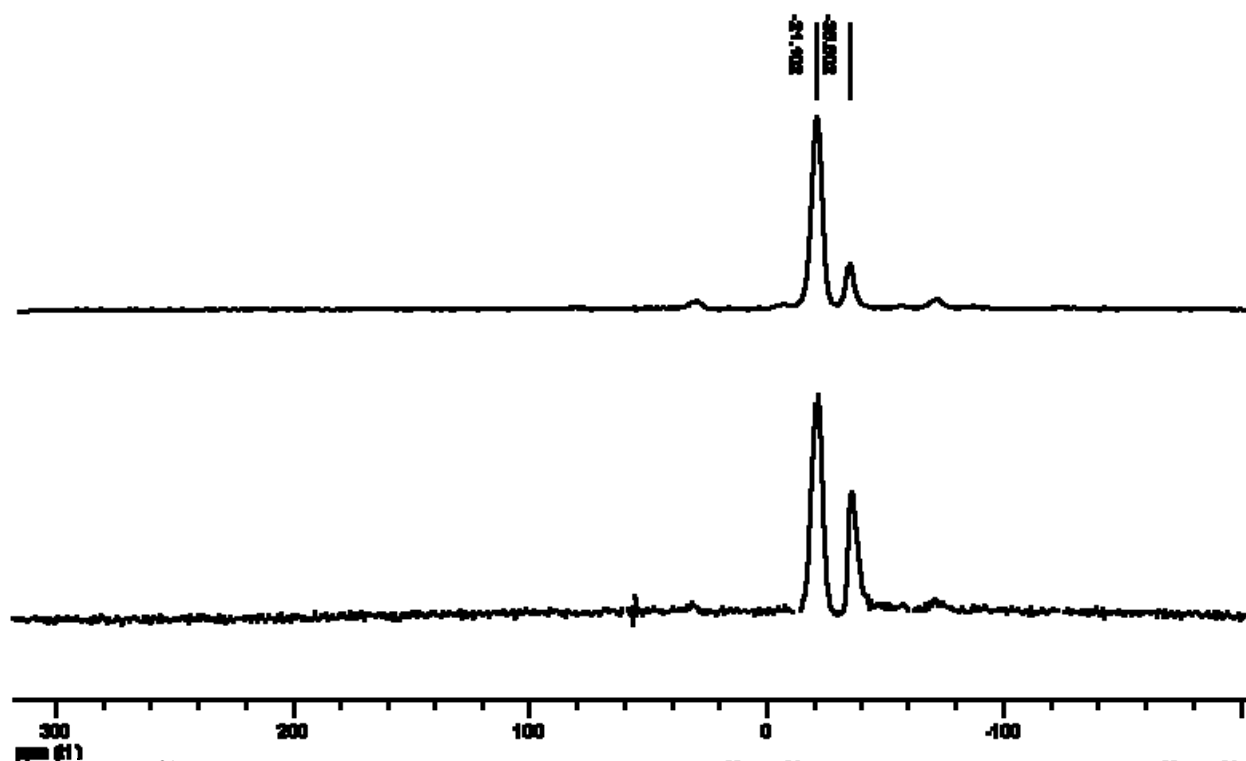


Figure S2. ^{29}Si NMR (59 MHz) spectra of silicones: $\text{D}_4^{\text{H}}\text{-D}_4^{\text{V}}$ 1:1 molar ratio (top) and $\text{D}_4^{\text{H}}\text{-D}_4^{\text{V}}$ 2:1 molar ratio (bottom).

Differential Scanning Calorimetry: A TA DSC Q200 was used. The sample was heated to 150 °C and then cooled to -150 °C and finally heated again to 150 °C (10 °C/min heating and cooling).

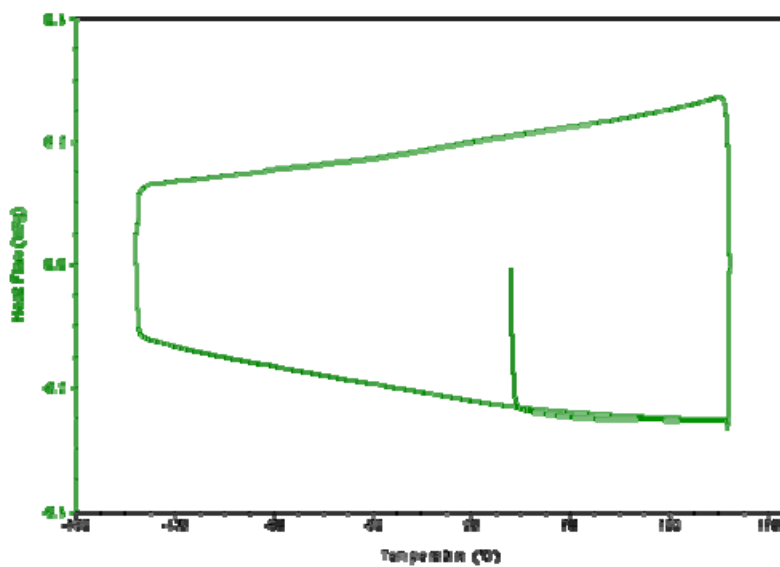


Figure S3. DSC data for $\text{D}_4^{\text{H}}\text{-D}_4^{\text{V}}$ (2:1 molar ratio).

Attenuated total reflection infrared (ATR-IR) spectroscopy: An ATR-IR spectrum of D_4^H - D_4^V 2:1 molar ratio was recorded using a Perkin Elmer 100 FT-IR spectrometer. Assignments: 2884-2960 cm^{-1} , CH_3 stretching; 2158 cm^{-1} , Si-H stretching; 1408 cm^{-1} , CH_2 deformation; 1260 cm^{-1} , Si- CH_3 stretching; 1026 cm^{-1} , Si-O stretching; 908 cm^{-1} , Si-H bending; 756 cm^{-1} , CH_3 rocking and Si-C stretching.

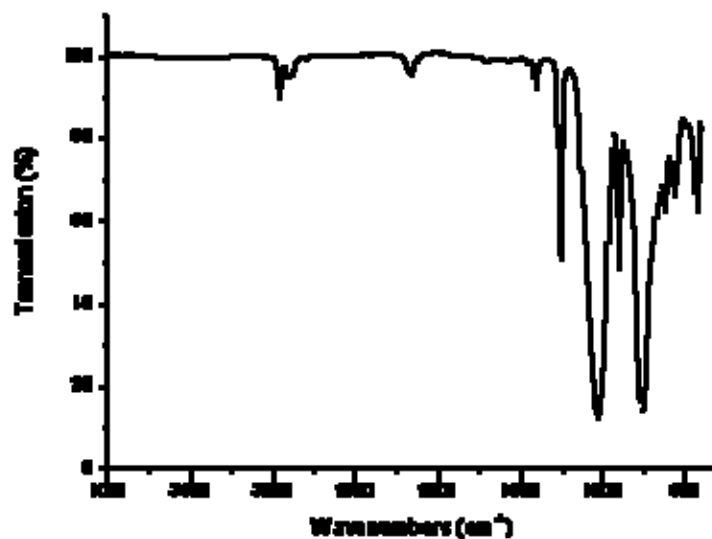


Figure S4. Infrared spectrum of D_4^H - D_4^V (2:1 molar ratio).