Supporting Information

Experimental Details

The cells were designed from Stainless Steel with a 1/1000 inch Stainless Steel window welded by electron beam. The liquids inlet was through ¼ inch Stainless Steel tubing which could be locked by Swagelok caps.

The sample temperature was monitored with a type K thermocouple probe embedded in a tube which extends from the back of the edge of the cell wall close to the sample.

The heated cell and associated plumbing are supported inside a water-cooled jacket which protects the surrounding heat-sensitive plastic scintillators and light pipes of the positron detectors.

Heating or cooling was accomplished by a controlled flow (a silicone oil) system through a fluid jacket wrapped around the the back of the target cell using a circulator and PID control unit.

In contrast to conventional magnetic resonance studies, where bulk polarization is a consequence of differing Boltzmann populations in high magnetic fields, in μSR the muon polarization is intrinsic to the probe, and is a direct consequence of the nuclear weak interaction. In the decay sequence $\pi^+ \rightarrow \mu^+ \rightarrow$ e+, the muon is produced 100% spin polarized from pion decay and the (detected) positron is subsequently emitted preferentially along that spin direction, providing a remarkably sensitive measure of the interactions of the muon spin with its environment.

Calculation of Hyperfine Constants from Spectra for weak signals

When the higher frequency signal (ν_{34}) lacked sufficient signal-to-noise to give reliable results, the muon hyperfine constant was determined from a combination of the lower frequency radical signal (ν_{12}) and the diamagnetic muon Larmor frequency. The Larmor precession frequency of the positive muon is defined by $\omega_\mu = \gamma_\mu B = g_\mu eB/2m_\mu$, expressed in terms of the gyromagnetic ratio $\gamma/2\pi = 135.534$ MHz/T.

The following two equations have been solved for muon hyperfine coupling constant, A_μ.

$$\nu_{12} = \nu_{mid} - \frac{1}{2}A_\mu \qquad \text{S1}$$

$$\nu_{mid} = \frac{1}{2}\left[\left\{A_\mu^2 + \left(\nu_e + \nu_\mu\right)^2\right\}^{1/2} - \nu_e + \nu_\mu\right] \qquad \text{S2}$$

ν_e is the electron Larmor frequency. Proton hyperfine constants A_p were determined from the field positions of resonances in μLCR spectra:

$$B_{LCR} = \frac{1}{2}\left[\frac{A_\mu - A_X}{\gamma_\mu - \gamma_x} - \frac{A_\mu + A_X}{\gamma_e}\right] \qquad \text{S3}$$

where γ_p is the proton gyromagnetic ratio.

The asymmetry parameter, *A(t)*, in a transverse field experiment includes contributions from paramagnetic Mu, as well as free radical and diamagnetic molecules:

$$A(t) = \Sigma_i A_i \exp(-\lambda_i t)\cos(w_i t + \varphi_i) \qquad (1) \qquad \text{S4}$$

where t is time, A_i is the asymmetry of the fraction i in its given environment, λ_i is the relaxation rate of the muon spin in that environment, w_i is the corresponding precession frequency, and φ_i is the initial phase of this fraction. The parameters of interest, A_i, $\lambda_{\mathrm{i},}$, and w_i are extracted from fits of equation S4 to experimental data.