Experimental Details

The pressure cell assembly consists of three parts – body, window and retaining flange – machined from titanium alloy (Ti 6AL-4V). The window design is as previously described\(^9\) but the body of the cell is smaller (see Figure S1), so that the sample volume has been reduced from 112 ml to 30 ml, with consequent improvement in temperature control and homogeneity, and heating and cooling times. The disadvantage of a smaller cell is that it is only suitable for relatively high density samples (≥ 0.5 g cm\(^{-3}\)), to ensure complete muon stops in the sample before they hit the end of the cell.

**Figure S1.** Schematic drawing of pressure cell assembly. Muons enter the target from the left side of the figure while all electrical and plumbing connections exit to the right.

The sample temperature was monitored with a type K thermocouple probe embedded in a tube which extends from the back of the cell to the centre of the sample volume. Temperature control, pressure measurement and plumbing are largely as described previously\(^9\) except that the sample in/out fittings are at the back of the cell and the ¼ inch high pressure tubing has been replaced with 1/16 inch diameter
standard flexible steel capillary tubing commonly used in HPLC apparatus. Also, the heated pressure 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. The whole assembly (pressure cell inside jacket) extends into the HELIOS solenoid bore such that the pressure cell is positioned at the center of the magnet. For safety reasons the apparatus is limited to 400°C and 400 bar.

**Calculation of Hyperfine Constants from Spectra**

Muon hyperfine constants were extracted from TF-μSR spectra, usually from the difference of a pair of muon precession frequencies. In a few cases, where the higher frequency signal lacked sufficient signal-to-noise to give reliable results, the muon hyperfine constant was determined from a combination of the lower frequency (stronger) radical signal and the diamagnetic muon Larmor frequency, as described previously.\textsuperscript{10} The muon Larmor frequency $\nu_\mu$ is directly proportional to the applied magnetic field $B_0$:

$$\nu_\mu = \gamma_\mu B_0 \quad (S1)$$

where $\gamma_\mu = 13.554 \text{ MHz/kG}$ is the muon magnetogyric ratio, and the two muon precession frequencies of the radical are given by\textsuperscript{14}

$$\nu_R = \nu_{\text{mid}} \pm \frac{1}{2} A_\mu, \quad (S2)$$

where

$$\nu_{\text{mid}} = \frac{1}{2} \left[ A_\mu^2 + \left( \nu_e + \nu_\mu \right) \right]^{1/2} - \nu_e + \nu_\mu, \quad (S3)$$

$A_\mu$ is the muon hyperfine frequency, and $\nu_e$ is the electron Larmor frequency. Proton hyperfine constants $A_p$ were determined from the field positions of resonances in μLCR spectra:\textsuperscript{15}

$$B_{\text{LCR}} = \frac{1}{2} \left[ \frac{A_\mu - A_p}{\gamma_\mu - \gamma_p} - \frac{A_\mu + A_p}{\gamma_e} \right] \quad (S4)$$

where $\gamma_p$ is the proton magnetogyric ratio.
The resonance fields have statistical fit errors of 2-6 G, which translates into an uncertainty in $A_p$ of up to 0.1 MHz. In addition, the systematic error in extrapolating $A_\mu$ is estimated to be 0.1 MHz, resulting in an overall uncertainty in the $A_p$ values listed in Tables 1 and 2 of no more than 0.2 MHz.