Figure 1s

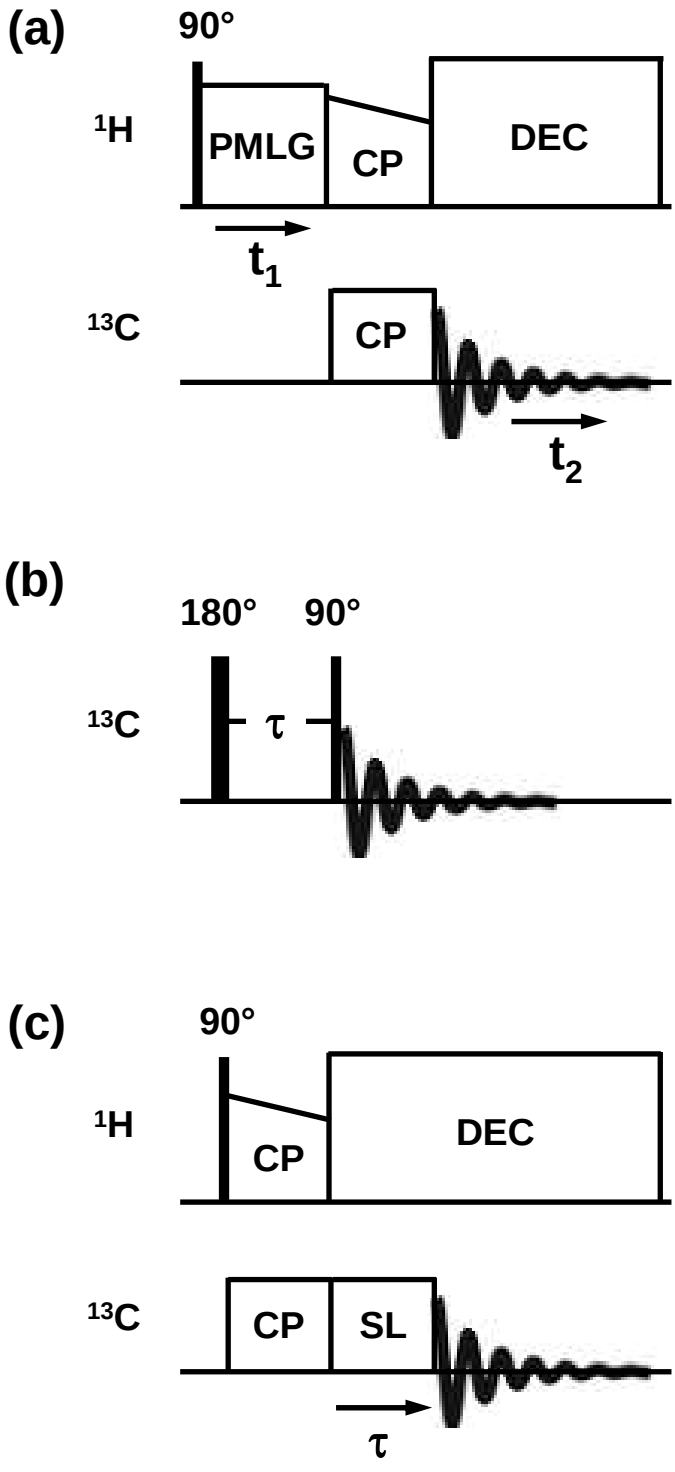

Figure 3s. (a) Pulse schemes for 2D ^{1}H-^{13}C HETCOR using PMLG ^{1}H-^{1}H decoupling during t_1 and polarization transfer using ramp-field on ^{1}H. (b) Inversion recovery pulse scheme for T_1 Measurement. (c) Pulse scheme using variable ^{13}C spin-lock period following CP from ^{1}H to follow $T_{1\rho}$ magnetization decay in carbons.

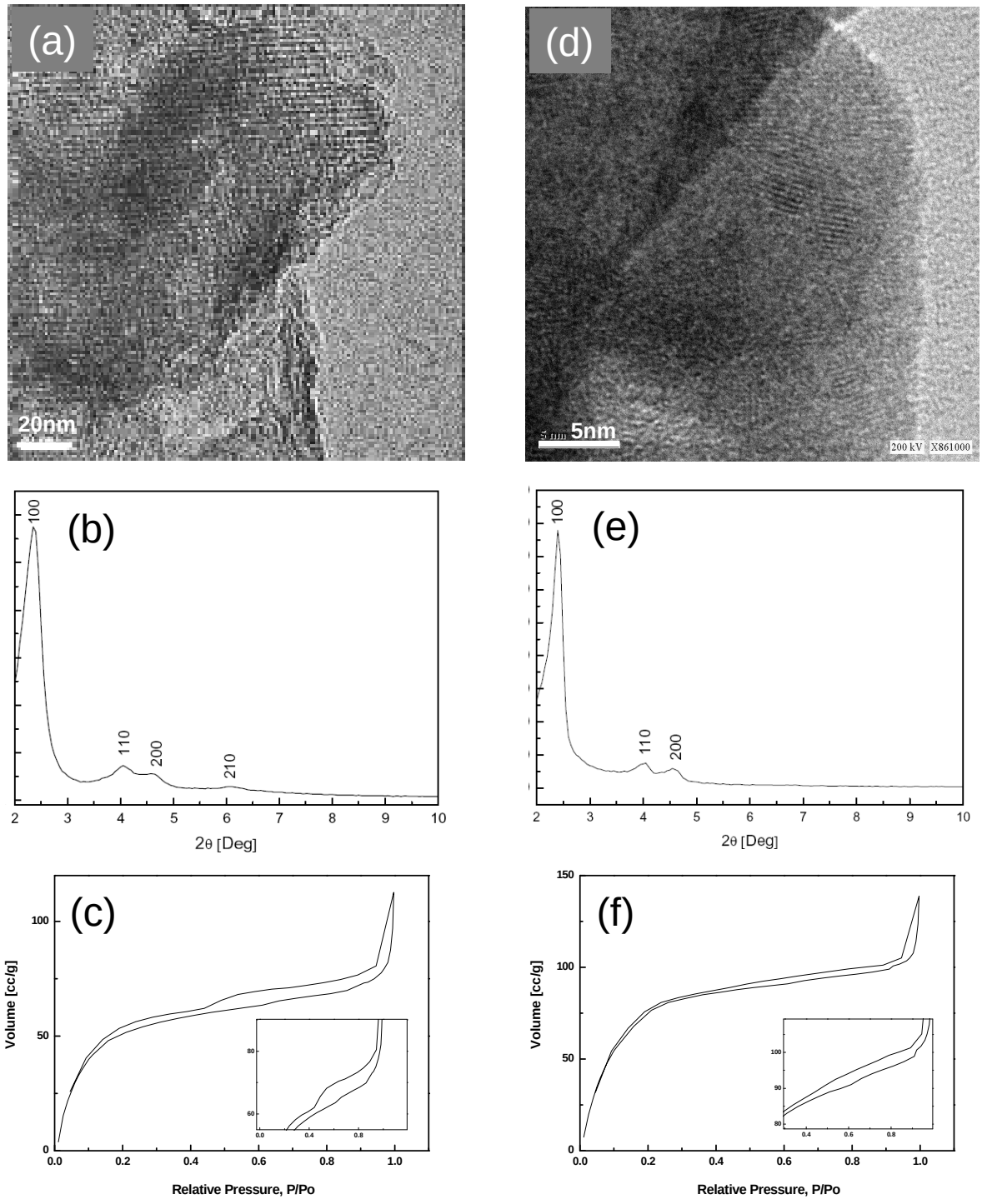

Fig. 1s. HR-TEM micrograph, low angle XRD diffractogram and BET isotherm of ap-MCM41 (a-c) and o-MCM41 (d-f) are shown. Insets in (c) and (f) show hysteresis regions in the $N_{2(g)}$ adsorption-desorption cycle for the two materials.

Figure 2s

Table 1s: Low Angle X-Ray Diffraction of Modified Mesoporous Silica

Materials	**2θ**			***d-spacing* (Å)**			**a_o (Å)**
	100	*110*	*200*	*100*	*110*	*200*	
ap-MCM41	2.3	4	4.6	37.5	22.1	19.2	44
o-MCM41	2.4	4	4.5	36.8	22.1	19.6	43

Table 2s: $N_{2(g)}$ adsorption and unit cell data from BET isotherms of Modified Mesoporous Silica

Materials	BET surface area/m^2g^{-1}	BJH pore volume/ccg^{-1}	Pore diameter/Å	$a_o(=2d/\sqrt{3})$/Å	Wall thickness/Å
ap-MCM41	500	0.23	35	44	9
o-MCM41	520	0.25	35	43	8

Tables 1s

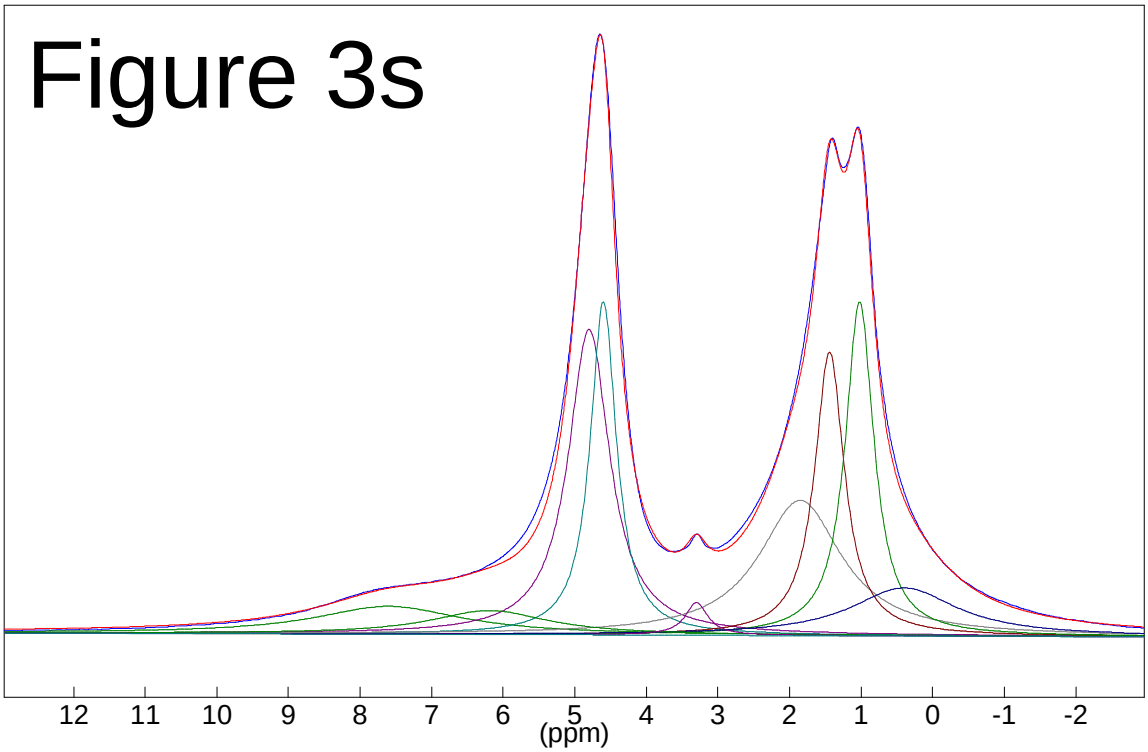

Figure 1s. ^{1}H 90°-acquire spectrum of o-MCM41 with deconvolutio n of the overlapping spectral lines. Deconvolution here and Fig. 2s was carried out using the DMFIT software (ref 25 in text).

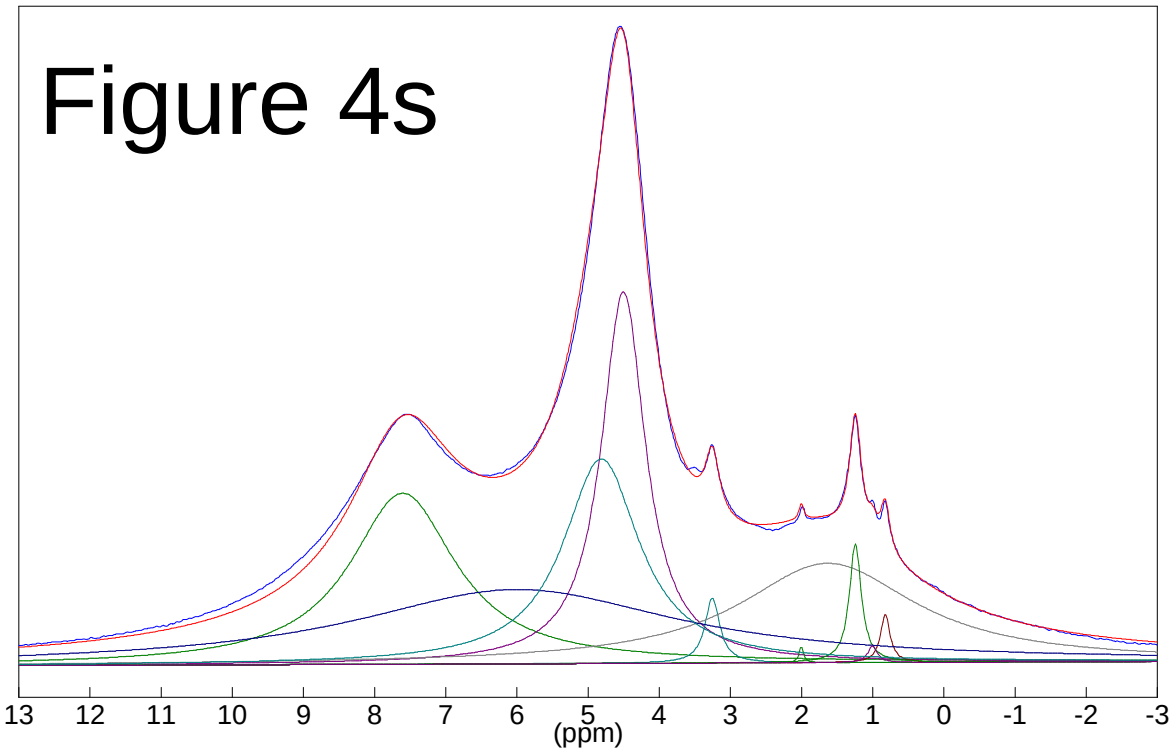

Figure 2s. ^{1}H 90°-acquire spectrum of ap-MCM41 with deconvoluti on of the overlapping spectral lines. Table 2s lists the parameters derived from this deconvolution.

Table 3s. Spectral parameters of the proton species in ^{1}H spectrum of o-MCM41

Chemical identity	Measure δ Chemical Shift	Relative intensity %	Linewidth (ppm)
H on C1 and C8	0.4	8.1	1.8
H on C2 – C7 ‡	1.0	15	0.48
H on C2 - C7 ‡	1.35	13.3	0.5
Silanol (N.E.) *	1.8	17.7	1.4
Silanol (W.E.) *	3.2	1.2	0.38
H_2O	4.5	13.7	0.44
H_2O	4.7	20.0	0.7
acidic	6.1	4.5	2.0
acidic	7.5	6.5	2.5

‡Protons show up as two lines, internal assignment not known
*** N.E. – non exchangeable , W.E. water exchangeable**

Table 4s. Spectral parameters of the proton species in ^{1}H spectrum of ap-MCM41

Chemical identity	Measure δ Chemical Shift	Relative intensity %	Linewidth (ppm)
H on C1	0.8	0.5	0.2
other	1.0	0.2	0.2
H on C2	1.2	1.5	1.5
Silanol (N.E.) *	1.7	18.6	3.0
H on C3	3.2	1.0	0.3
H_2O	4.5	17.3	0.8
H_2O	4.8	16.5	1.3
acidic	6.0	25.3	5.5
acidic	7.6	19	1.8

*** N.E. – non exchangeable , W.E. water exchangeable**

Figure 5s

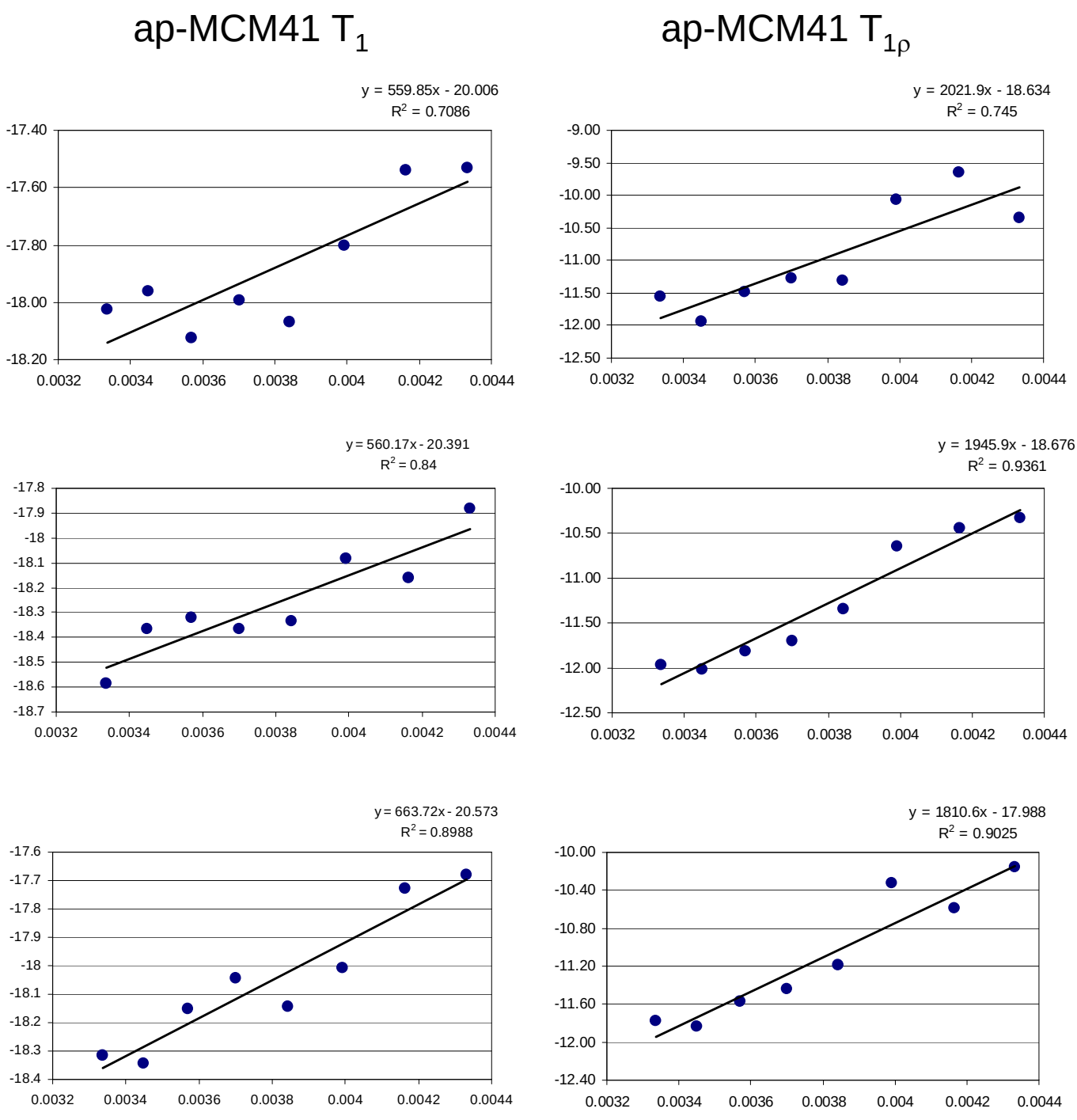

Figure 4s. Data from T_1 (left) and $T_{1\rho}$ (right) measurements versus temperature converted to Ln(τ) versus reciprocal of the temperature showing Arrhenius dependence of correlation time on temperature. Correlation time (τ) is extracted from the relaxation times using Eq. 1 and 2 separately and the corresponding *K* and *K'*.