

Experimental part

Well-crystallized kaolinite (KGa-1b; Georgia) was obtained from the Source Clays Repository of the Clay Minerals Society, Purdue University, West Lafayette, IN, USA. The purification of KGa-1b and the preparation of DMSO-K were done according to previously published procedures.⁸

The organic reagents were of reagent grade and used as received from Sigma-Aldrich: EOA (b.p. 170°C), DEOA (b.p. 217°C) and TEOA (b.p. 190-193°C).

X-ray diffraction patterns (XRD) were obtained with a Philips PW 3710 instrument equipped with Ni-filtered and Cu-K radiation ($\lambda = 0.15418$ nm) operating at 45 kV and 40 mA.

Differential thermal analysis (DTA) and thermal gravimetric analysis (TGA) was recorded on a SDT 2960 Simultaneous DSC-TGA instrument under N₂ flow (100 mL/min) with a heating rate of 10°C/min.

Infrared spectra were acquired on a Thermo Nicolet Nexus 670 FT-IR *E.S.P.* spectrometer under dry air using 128 scans with a resolution of 4 cm⁻¹. The spectra were recorded using KBr pellets. Before the recording of the spectrum, a flow of hot air was circulated in the room containing the pellet during 30 minutes to remove the water molecules adsorbed physically on the sample.

Solid-state ¹³C NMR CP/MAS spectra were collected on a Bruker AVANCE 500 NMR spectrometer operating at 125.77 MHz for ¹³C. Approximately 50 mg of sample were packed in 4 mm O.D. Zirconia rotors which were spun at the magic angle at speeds between 10 and 14 kHz. A cross polarization sequence was used employing a 75% - 100% linear ramped pulse for the 2 msec contact pulse in the ¹³C channel. Proton 90° pulses of 3.25 µsec were used. TPPM proton decoupling was used during the 25 msec acquisition with 5.6 µsec pulses and an 84 kHz decoupling field. The recycle delay was 2 seconds. The spectral width was set at 80 kHz and 4 K data points were collected for each free induction decay. Data collection times ranged from 1 to 2.5 hours. The ¹³C chemical shifts were referenced to TMS

at 0 ppm using the high frequency signal of adamantane at 38.4 ppm as a secondary standard. ^{27}Al MAS NMR spectra were obtained on a Bruker AVANCE-500 instrument operating at 130.32 MHz at a spinning rate of 14.0 kHz.

Elemental analysis (CHN) was obtained from Guelph Chemical Laboratories Ltd:

EOA-K: C: 5.46%; H: 2.47%; N: 3.10%

DEOA-K: C: 5.39%; H: 2.52%; N: 1.82%

TEOA-K: C: 7.11%; H: 2.23%; N: 0.86%

These values correspond to stoichiometries respectively of 0.7, 0.4 and 0.3, compared respectively to values of 0.9, 0.5 and 0.5 obtained from TGA. The stoichiometry obtained from EA is systematically lower than the one obtained from the TGA data. The discrepancy is due to the difficulty in combusting all the carbonaceous material during the elemental analysis, as was observed before for other thermally robust organokaolinite derivatives.^{5c} Carbon-mineral nanocomposites are formed during the thermolysis process, by the trapping of carbonaceous materials in the interlayers of kaolinite. This can be seen by a continuous slight decrease of weight up to 1100⁰C, above the transition temperature from metakaolinite to mullite.

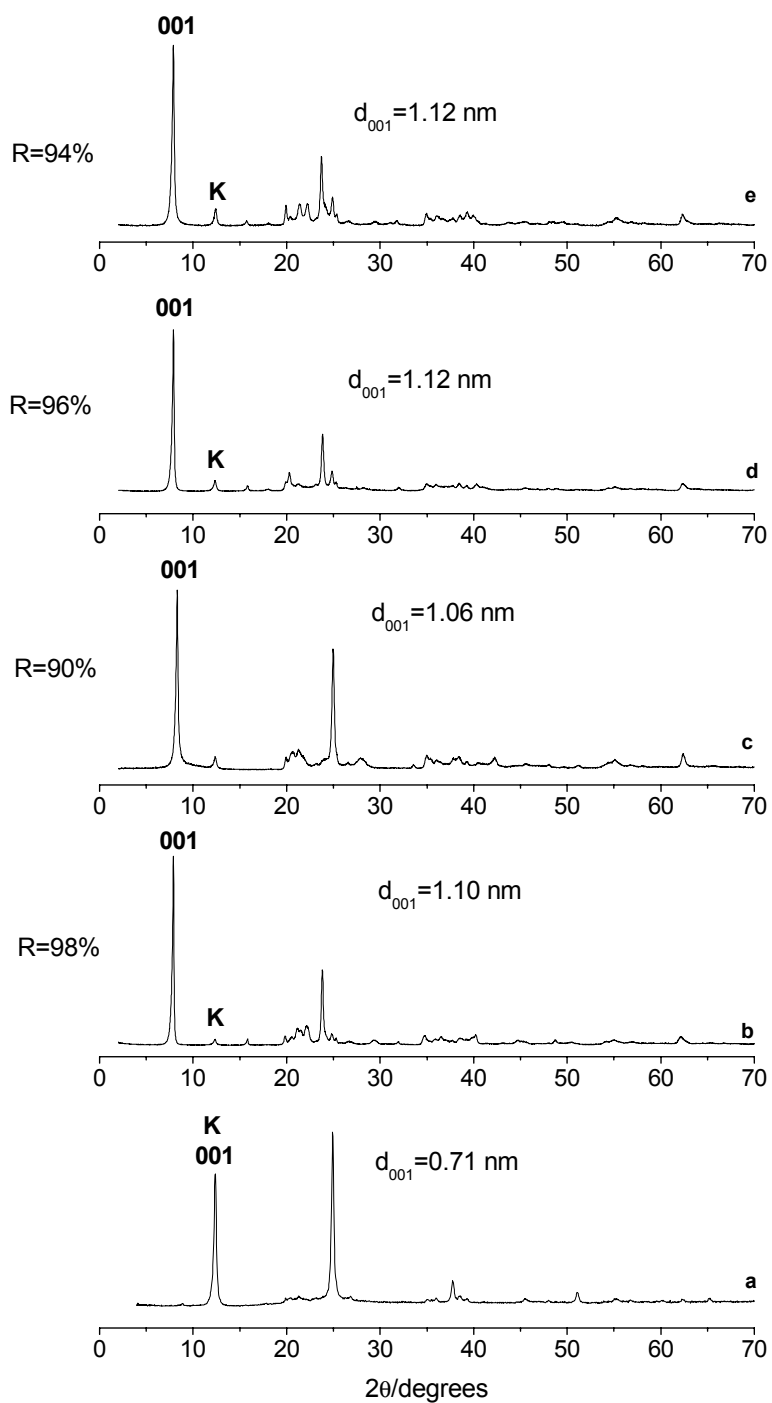


Fig. S1 Oriented XRD patterns (2° - 70° 2θ) of (a) kaolinite, (b) kaolinite intercalated with DMSO and after reaction with (c) EOA, (d) DEOA, (e) TEOA.

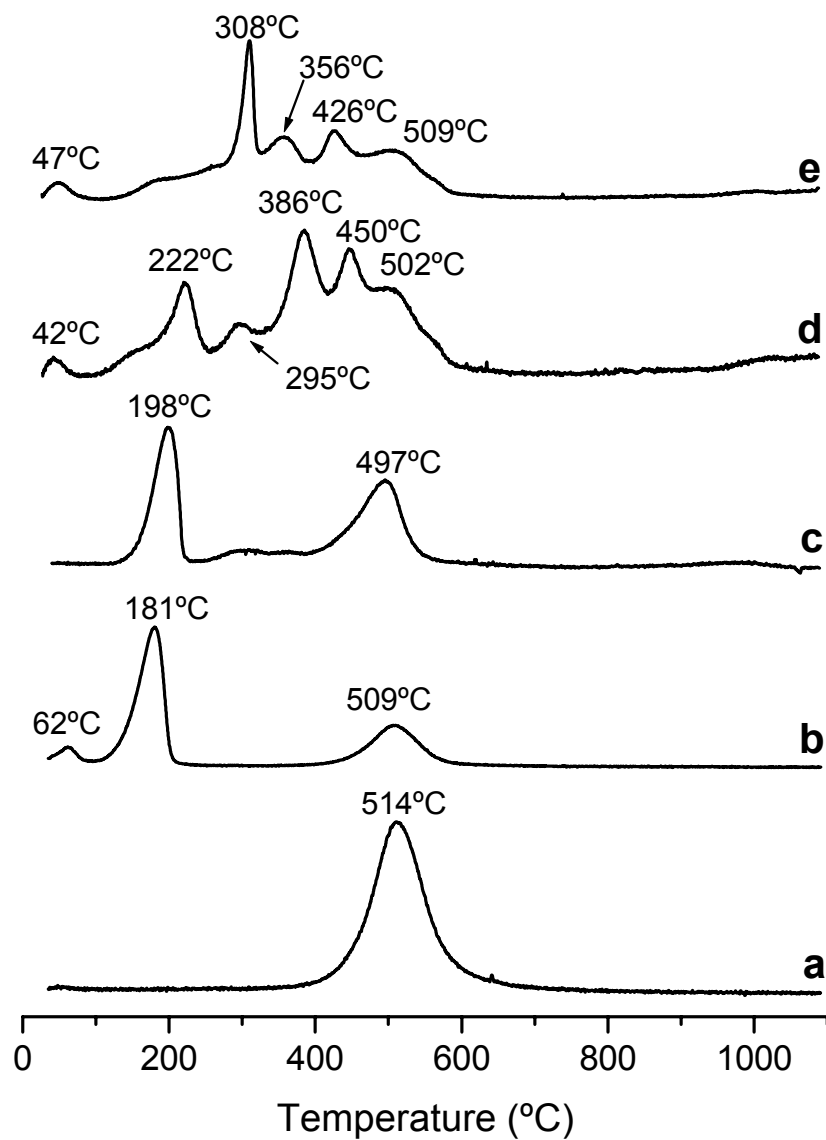


Fig. S3 DTG traces (RT-1100^oC) of (a) kaolinite, (b) DMSO-K, (c) EOA-K; (d) DEOA-K; (e) TEOA-K. The maxima are indicated.

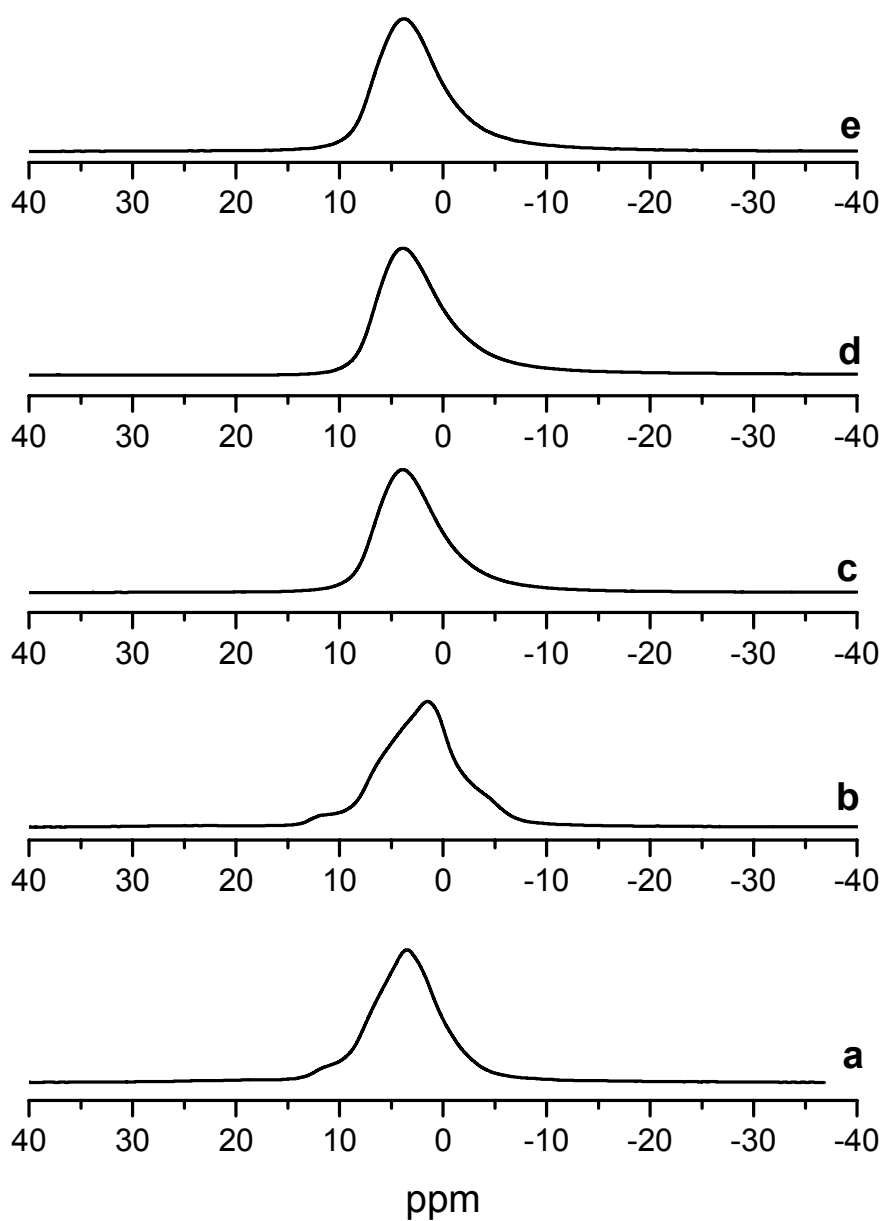


Fig. S5 ^{27}Al MAS NMR spectra of (a) kaolinite, (b) Kaolinite intercalated with DMSO, and after reaction with (c) EOA, (d) DEOA, (e) TEOA.

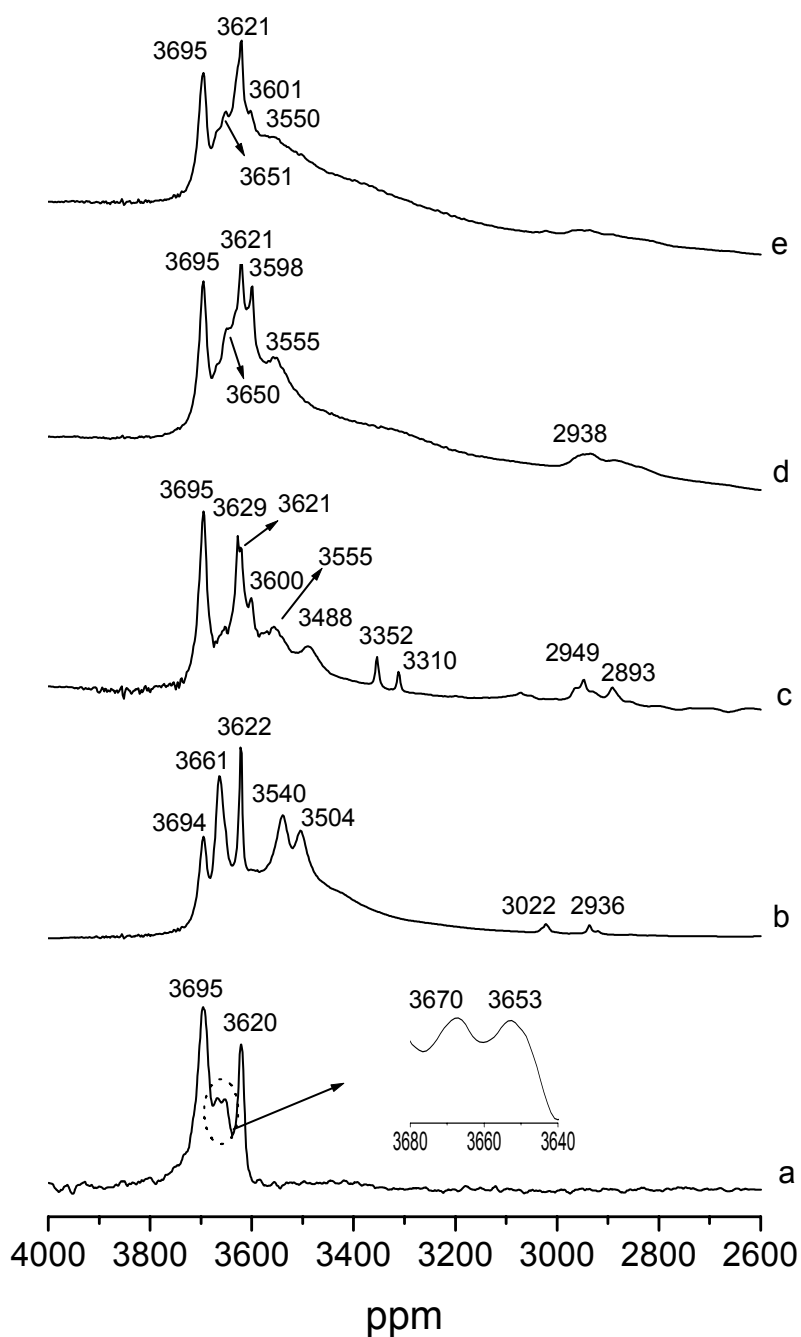


Fig. S6 FTIR spectra of (a) kaolinite, (b) kaolinite intercalated with DMSO and after reaction with (c) EOA, (d) DEOA, (e) TEOA.