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Supplementary Information





Hydrogen-treated DND E_{ZETA} =+45 ±5 mV



Deuterium-treated DND









Deuterium-treated DND_{OX} $E_{ZETA}=+38 \pm 1 \text{ mV}$



 $DND_{OX}E_{ZETA}$ =-57 ±5 mV



Tritium-treated DND_{OX}

Figure S1: DLS hydrodynamic diameters and E_{ZETA} of the different DND investigated in this study. Zeta potential were measured at the pH of the native suspension measured at 6.5, without any adjustment. For hydrogen and deuterium-treated samples, at least 10 measurements were performed and error bars represent the standard deviation. For tritium-treated sample, the use of another DLS set-up, dedicated to radioactive nanoparticles, did not allow us to measure E_{ZETA} .

NMR experiments



Figure S2: Experimental CPMAS 13C{1H} NMR spectra of A-DND-D acquired at variable contact time (Tcp). The peak at 44 ppm is better excited at short CP time (80us) than the 34 ppm peak. For longer contact time, the decrease results from the relaxation of the proton in the rotating-frame (T1rho).



Figure S3: ¹H-¹³C 2D HETCOR spectrum of deuterium-treated (left) DND and (right) DND_{OX}.

Direct acquisition of ¹H spectra was not possible because of absorbed water on surface as well as other impurities. Thus, Two-dimensional heteronuclear correlations (HETCOR) approach was used which guaranty the observation of surface proton sites. HETCOR of deuterium-treated samples shown in Figure S5 reveal that almost all carbon peaks correlate to aliphatic protons around 1.8 ppm, in contrast to what observed (3.8 ppm) in Fang et al. paper (reference 38). Indeed, only a small contribution of carbon correlated to protons around 3.8 ppm is visible for deuterium-treated DND sample.



Figure S4: Experimental and simulated direct acquisition ¹³C MAS NMR spectra of deuterium-treated DND. Insert show the CPMAS spectra acquired at short (80us) and long (2ms) contact time.

Inset in Figure S4 shows how the 44 ppm component is better excited at short CP contact time that the broad 34 ppm component. An attempt to deconvolute the spectra using the main peaks observed in CPMAS with two narrow components is displayed in Figure S2. To limit uncertainties (especially between the two contributions underlying the peak at 34 ppm), all spectra were fitted simultaneously using the same NMR parameters for each peak (Table S1) and varying populations. It was found that two broad components were necessary to describe correctly the tail at higher chemical shifts ascribed to hydrogenated species. The values of the population of various species are rather similar for all samples and are compatible with the DND diameter.

NMR parameters		Population ±2%				
$\begin{array}{c} \delta \text{ (ppm)} \\ \pm 1 \end{array}$	FWHM (ppm) ±1	A-DND-H	A-DND-D	A-DND _{Ox}	A-DND _{Ox} - D	Site
28	2	13	16	20	18	Undetermined
34	4	3	4	2	1	Bulk
34	9	49	48	55	53	Sub-surface / Inner layer
40	19	35	32	33	28	Surface
64	26					

Table S1: ¹³C NMR parameters and population (Figures S4).



Figure S5: Experimental and simulated spectra of (direct acquisition) ¹³C MAS NMR of studied DND.



Figure S6: Experimental and simulated spinning sideband intensities of deuterium-treated DND.