

## Supporting Information

### Interaction of electrolyte molecules with carbon materials of well-defined porosity:

#### Characterization by solid-state NMR spectroscopy

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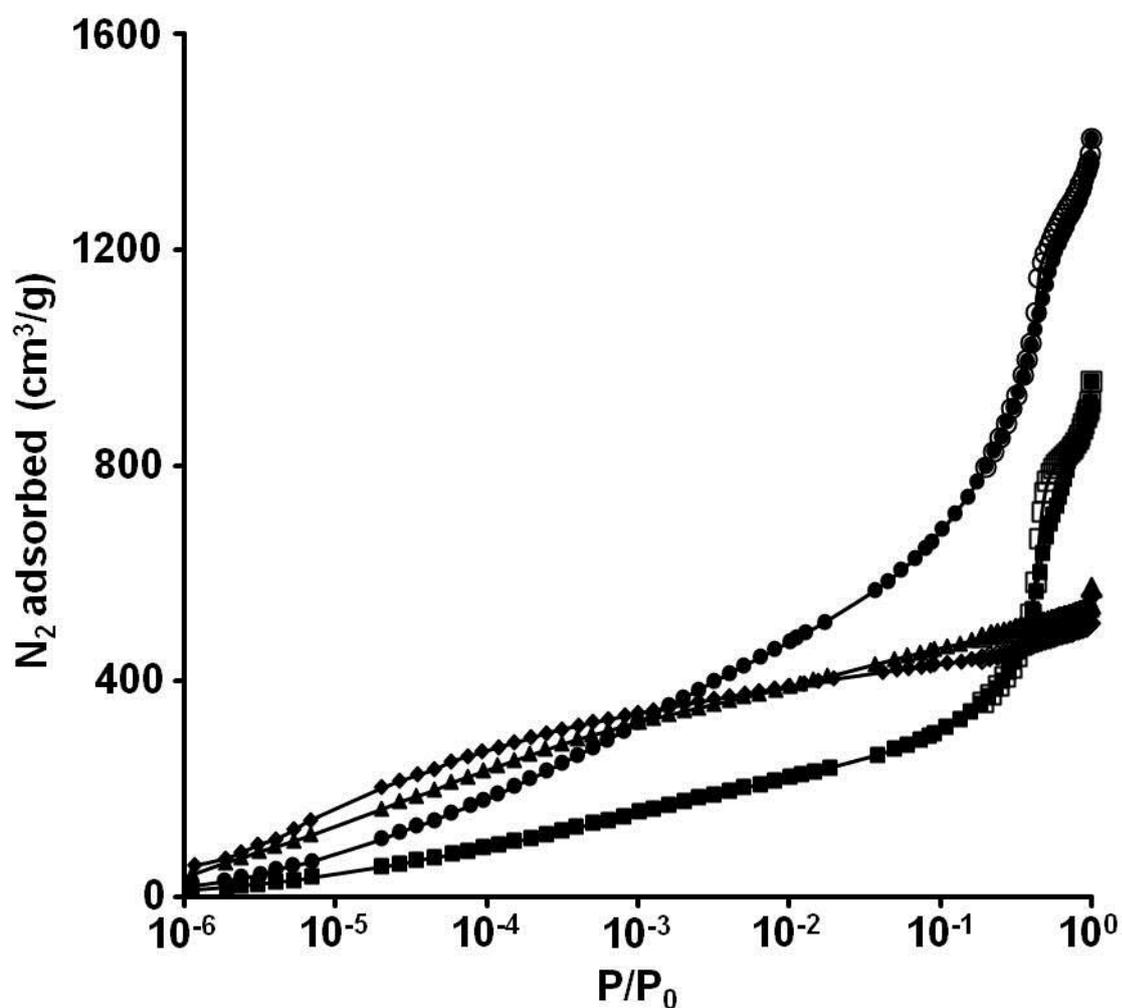


Figure S1: Logarithmically plotted nitrogen physisorption isotherms of the different materials (circles: OM-CDC, squares: CMK-3, triangles: TiC-CDC-1000, diamonds: TiC-CDC-600).

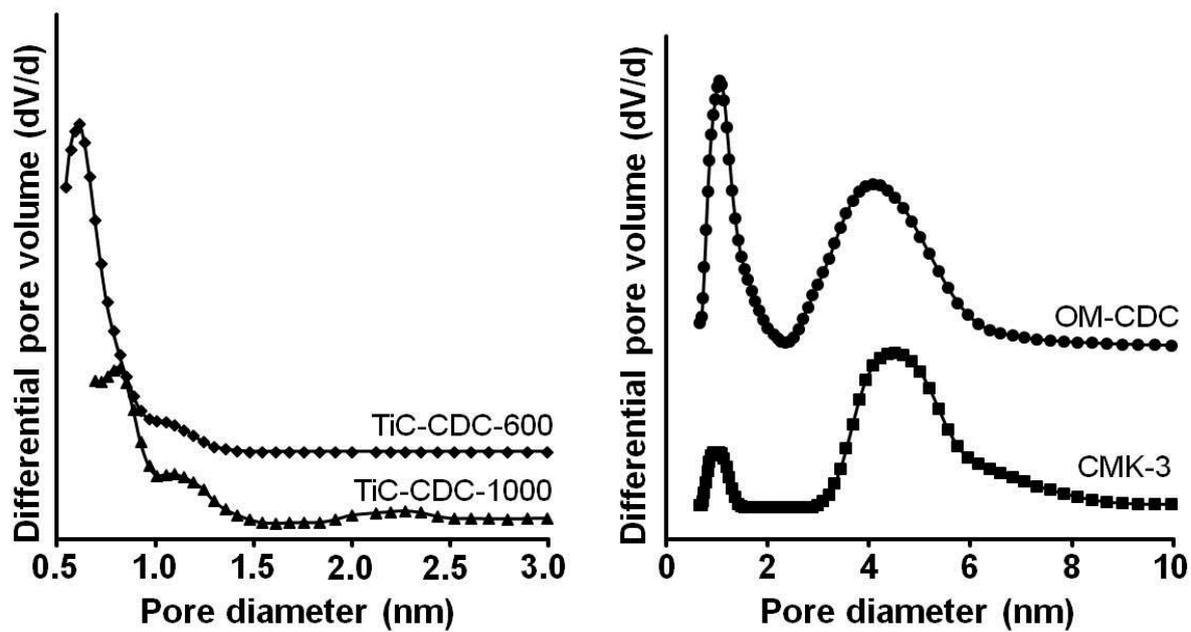


Figure S2: QSDFT PSD of the different samples showing the distribution of pore sizes.

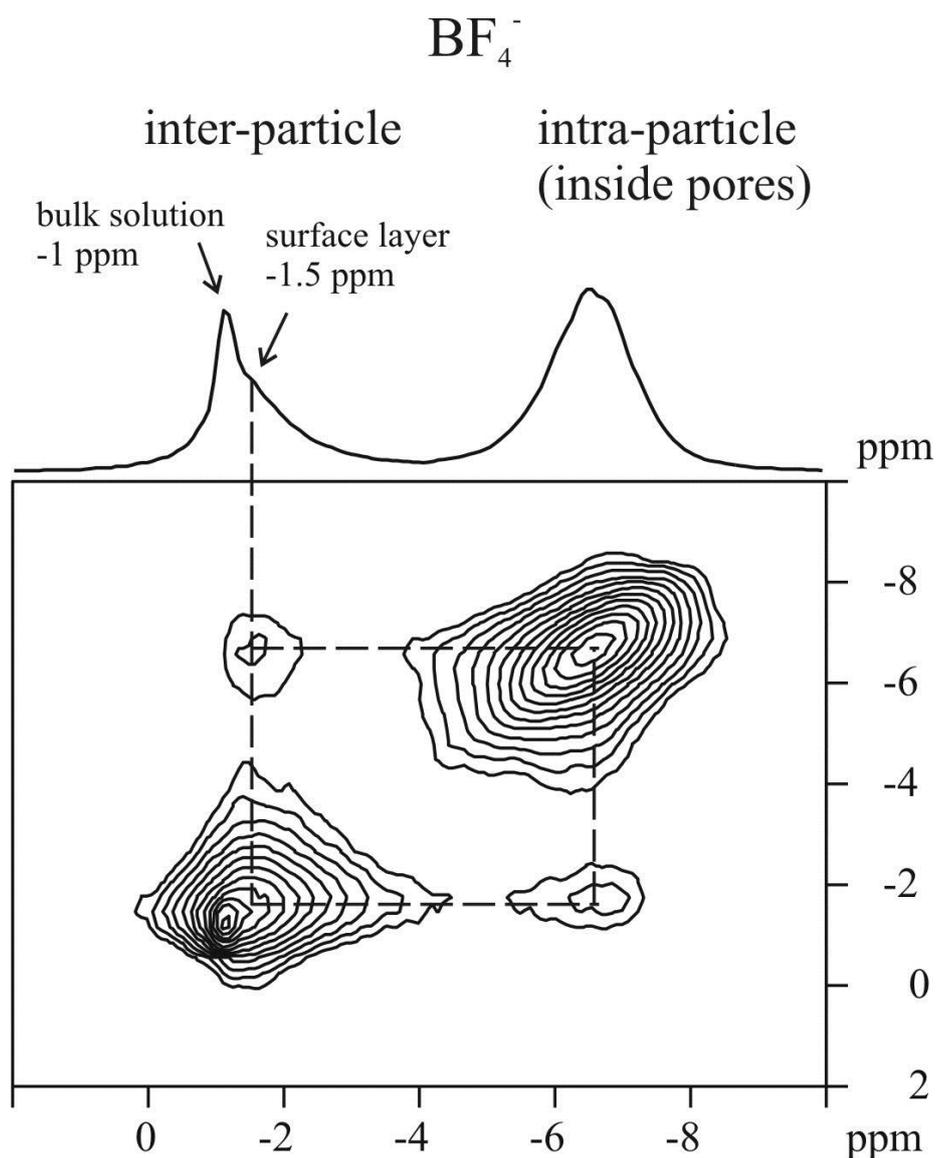


Figure S3: Two-dimensional  $^{11}\text{B}$ - $^{11}\text{B}$  exchange spectroscopy (2D EXSY) of TiC-CDC-1000 loaded with 1M TEABF<sub>4</sub> in AN measured at a mixing time of 5 ms. In this sample, the loading exceeded the available pore volume. This results in the additional presence of a relatively broad signal at -1.5 ppm and a narrow signal at -1 ppm. Since these signals only occur for loadings exceeding the internal pore volume and based on the chemical shift, both signals are ascribed to  $\text{BF}_4^-$  located outside the pores, i.e. in the inter-particle space. The species giving rise to the broad signal at -1.5 ppm exchange with the species adsorbed inside the pores even at a mixing time of only 5 ms as can be seen from the corresponding cross peaks. The broad signal at -1.5 ppm is, therefore, ascribed to  $\text{BF}_4^-$  outside the particles - but in close contact with the outer surface, i.e., they represent the outer surface layer covering the particles. The narrow signal at -1 ppm is ascribed to  $\text{BF}_4^-$  in the inter-particle space without surface contact (bulk solution, see Table 2).

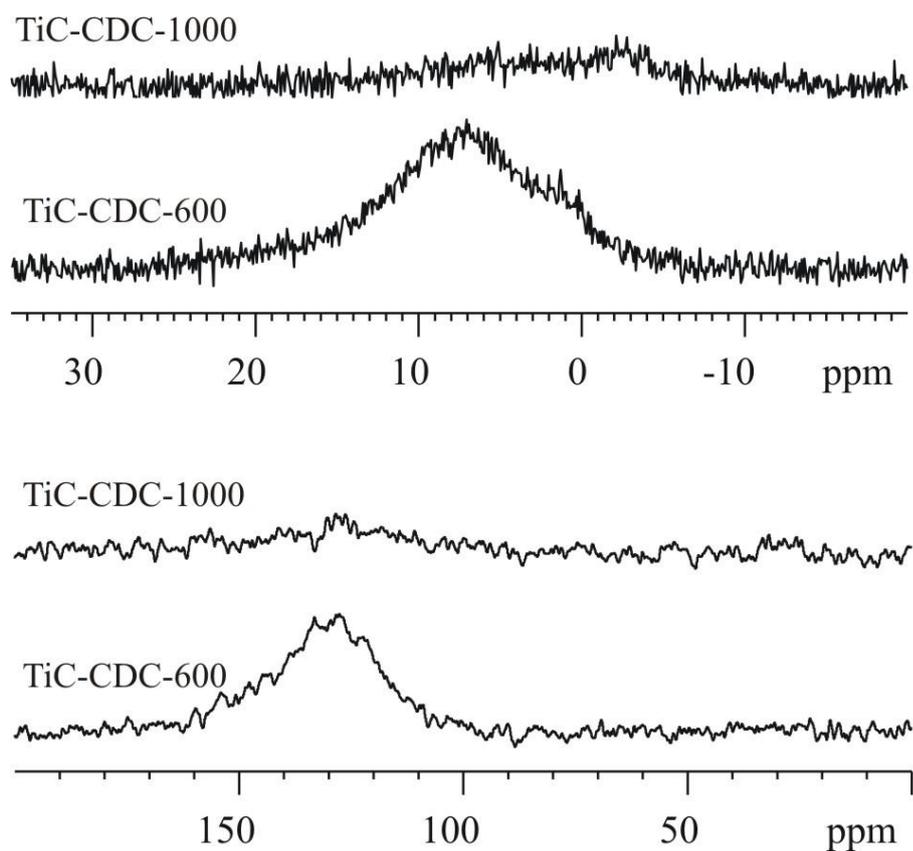


Figure S4:  $^1\text{H}$  MAS NMR spectra (top) and  $^{13}\text{C}$  CP MAS NMR spectra (bottom) of activated, unloaded TiC-CDC-600 and TiC-CDC-1000. Note the presence of a high  $^1\text{H}$  concentration in TiC-CDC-600 as reflected by a broad, intense  $^1\text{H}$  NMR signal at ca. 8 ppm and a pronounced cross polarization effect from  $^1\text{H}$  to  $^{13}\text{C}$  nuclei – in contrast to TiC-CDC-1000.

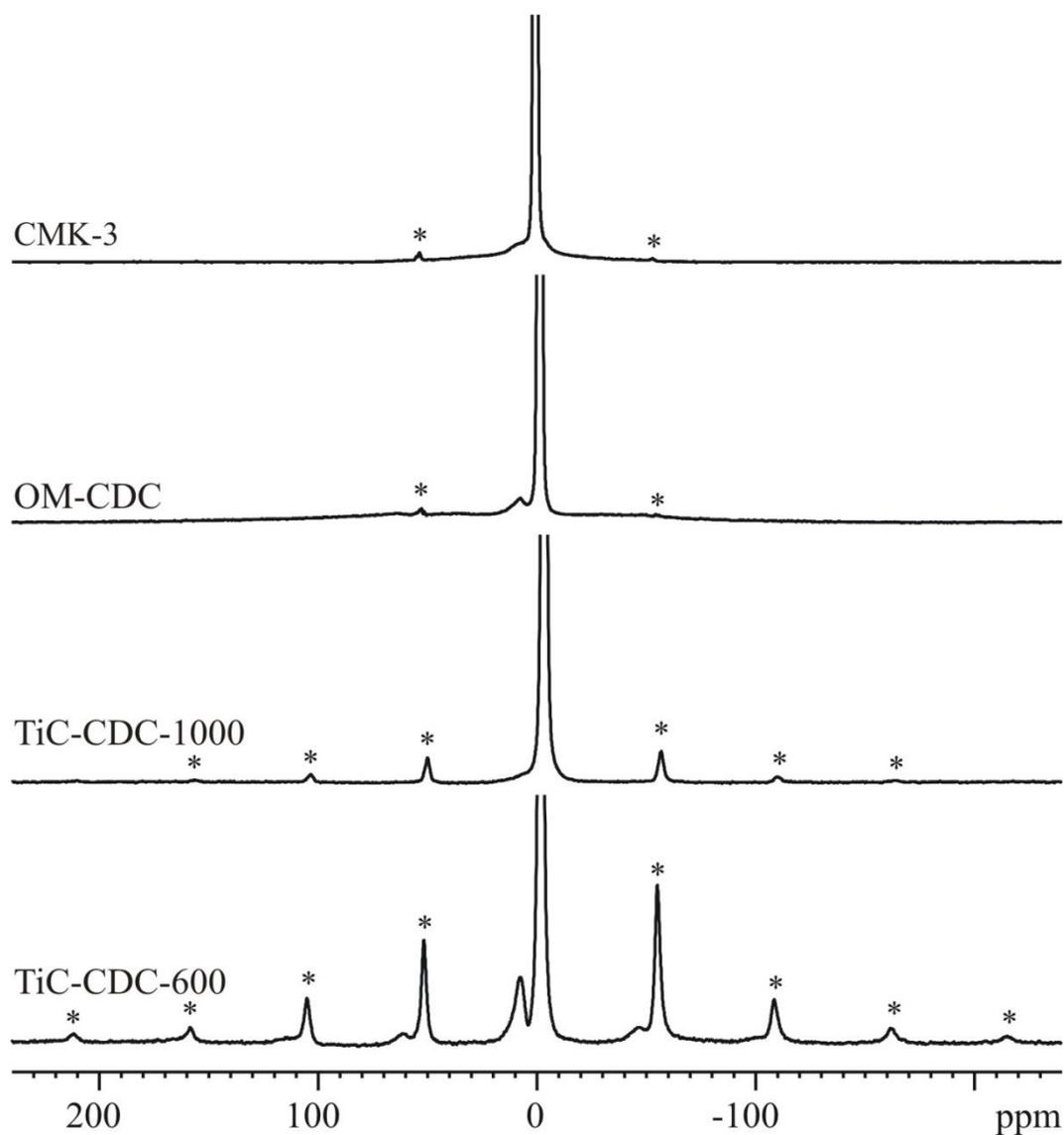


Figure S5:  $^1\text{H}$  MAS NMR spectra of CMK-3, OM-CDC, TiC-CDC-1000, and TiC-CDC-600 loaded with 1M TEABF<sub>4</sub> in AN. Note the presence of spinning sidebands (\*) especially in TiC-CDC-600 and TiC-CDC-1000 indicating an increasing degree of immobilization of the adsorbed molecules for narrow pore diameters. The spectra are identically scaled and the central lines of the electrolyte signals were cut in order to better visualize the spinning sidebands. The  $^1\text{H}$  MAS NMR signal at ca. 8 ppm is due to  $^1\text{H}$  nuclei located at the porous carbon material (see Figure S3). This signal exhibits a comparable intensity for CMK-3, OM-CDC, and TiC-CDC-1000.