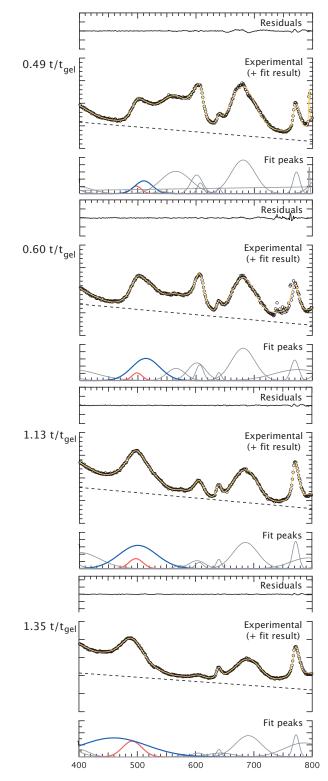
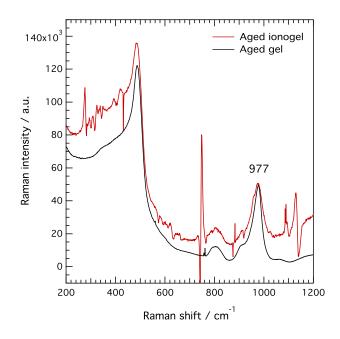
**Figure S1.** Experimentally recorded Raman spectra during the sol-gel reaction in the absence of the protic ionic liquid, for reaction times equal to 0.49, 0.60, 1.13, and 1.35  $t/t_{gel}$ , from top to bottom. For each plot the fitting components, the fit results, and the fit residuals are shown. The red and blue curves found below 530 cm<sup>-1</sup> are assigned to the D<sub>1</sub> and R network modes.



**Figure S2.** Raman spectra recorded for the aged gel (black) and the aged ionogel (red), *i.e.*, after 7 months from preparation. For the ionogel, the spectrum shown is indeed a difference spectrum between the ionogel and the pure ionic liquid, to reveal the spectral features due to silica.



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