Significance: The aldol reaction using L-proline-based supramolecular hydrogel as an efficient heterogeneous organocatalyst was described. Thus, 4-nitrobenzaldehyde (0.16 mmol) and cyclohexanone (155 mL) in 1 mL of toluene was added on the top of the hydrogel at 5 °C over 24 hours to give the aldol product (98% yield, anti/syn = 92:8, 88% ee). After decantation of the organic layer, the hydrogel did not seem macroscopically affected and was reused for two additional runs without any loss of catalytic activity (2nd: >99% yield, anti/syn = 93:7, 87% ee, 3rd: >99% yield, anti/syn = 92:8, 90% ee, respectively).

Comment: The morphology of the gel was observed by scanning electron microscopy (SEM), revealing the presence of a network of ribbons of several μm in length and less than 300 nm in width. X-ray powder diffraction of the xerogel was consistent with a lamellar structure in which compound 1 formed a bilayer with intercalation of the alkyl tails. The temperature (filtering, washing with toluene, and re-gelating) and pH (acidification, washing with toluene, neutralizing and re-gelating) as well as the responsiveness of the hydrogel allowed for its complete recovery and regeneration.