

Electronic Supplementary Information

Remarkable Increase in Basicity Associated with Supramolecular Gelation.

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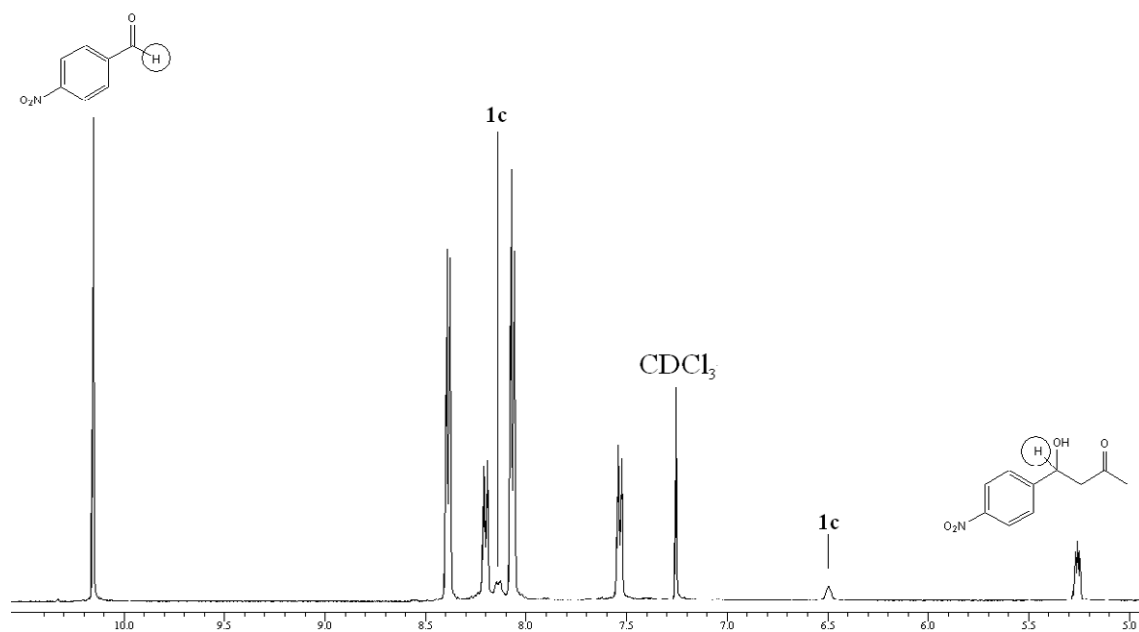
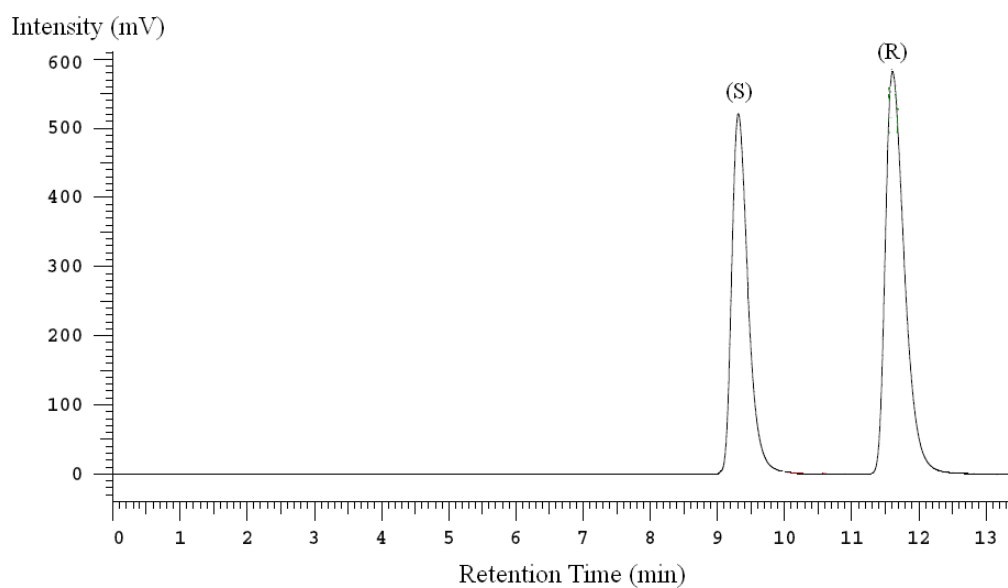


Figure ESI1. Representative ^1H NMR spectrum of the outcome of the aldol reaction catalyzed by **1c**.

a)



b)

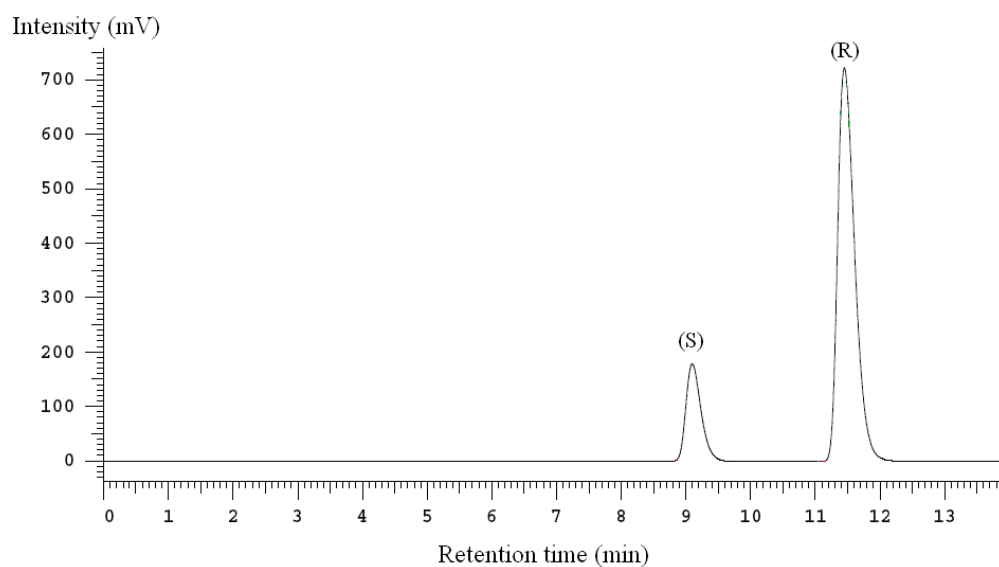


Figure ESI2. **a)** Chromatogram of racemic mixture of aldol and **b)** mixture enriched in the enantiomer R.

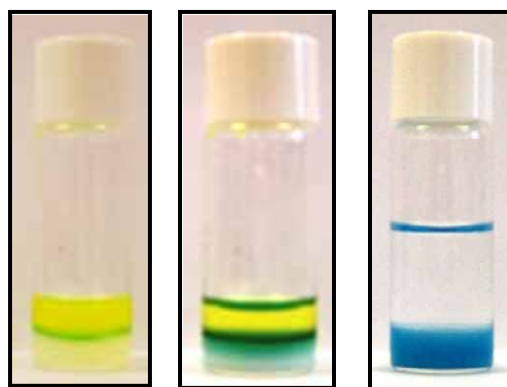


Figure ESI3. Diffusion of a solution of Bromothymol Blue (**I**₃, conc.= 3×10^{-4} mM, yellow) through a gel of compound **1c** in acetonitrile (left to right). The gel phase takes a blue colour typical from the basic form of the dye as diffusion progresses. Final addition of extra solvent allows the observation of a strong blue gel.

pKa	Base	I ₁	I ₂	I ₃	I ₄
24.3	DBU				
19.5	Pyrrolidine				
16.9	Benzylamine				
	1c (solution)				
	2c (solution)				
12.5	Pyridine				

Figure ESI4. pKa scale for **1c**, **2** and other common organic bases in CH₃CN and their solutions with dyes (**I**₁: Congo Red; **I**₂: 4-nitrophenol; **I**₃: Bromothymol Blue; **I**₄: Phenol Red; [**I**_n] = 3×10^{-4} mM; [Base] = 3.6 mM in all the cases but **1c gel** (1.8 mM).

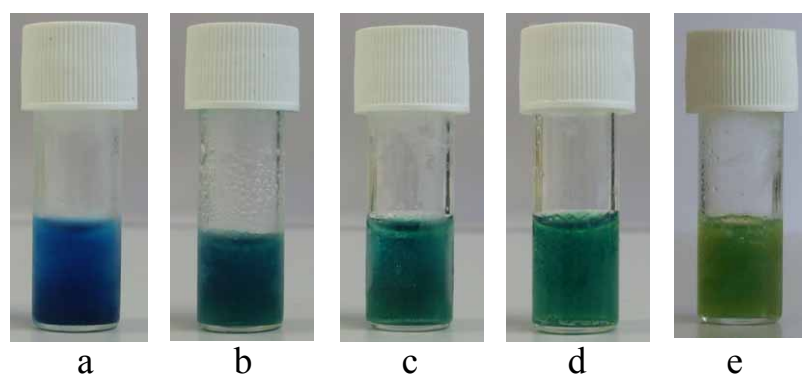


Figure ESI5. Two-component gels of **1c** and **3** in CH₃CN in the presence of **I**₃. [**1c**] = 15 mM in all cases; a) [**3**] = 0 mM; b) [**3**] = 3.75 mM; c) [**3**] = 7.50 mM; d) [**3**] = 11.25 mM and e) [**3**] = 15.00 mM.