Supporting Information

Multi-component supramolecular gels for the controlled crystallization of drugs: Synergistic and antagonistic effects

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1. Supplementary Figures and Tables

**Figure S1.** X-Ray diffraction patterns (298 K) plotted against the angle 2θ of the crystals of CAF inside the toluene gel of LG2-Lys and hexylamine and the polymorph II (blue lines) of CAF (pattern ICDD 00-051-1953).

**Figure S2.** X-Ray diffraction patterns (298 K) plotted against the angle 2θ of the crystals of CAF inside the toluene gel of DG2-Lys and hexylamine and the polymorph II (blue lines) of CAF (pattern ICDD 00-051-1953).
Figure S3. X-Ray diffraction patterns (298 K) plotted against the angle $2\theta$ of the crystals of CBZ inside the toluene gel of LG2-Lys and hexylamine and the alpha (blue lines) and beta-phases (green lines) of CBZ (patterns calculated from CCDC entries CBMZPN03 and CBMZPN10 for alpha and beta polymorphs respectively).

Figure S4. X-Ray diffraction patterns (298 K) plotted against the angle $2\theta$ of the crystals of CBZ inside the toluene gel of DG2-Lys and hexylamine and the alpha (blue lines) and beta-phases (green lines) of CBZ (patterns calculated from CCDC entries CBMZPN03 and CBMZPN10 for alpha and beta polymorphs respectively).
Figure S5. SEM images of the two types of CBZ crystals obtained inside the organogel generated from LG2-Lys dendron and hexylamine at a concentration of 10 mM on glass surface,

Figure S6. SEM images of the fibers constitutive of the organogel formed by mixing CBZ, LG2-Lys dendron and hexylamine at a concentration of 10 mM in toluene.
**Figure S7.** X-Ray diffraction patterns (298 K) plotted against the angle 2θ of the crystals of CAF inside the toluene gel of LG2-Lys and decylamine and the polymorph II (blue lines) of CAF (pattern ICDD 00-051-1953).

**Figure S8.** X-Ray diffraction patterns (298 K) plotted against the angle 2θ of the crystals of CAF inside the toluene gel of DG2-Lys and decylamine and the polymorph II (blue lines) of CAF (pattern ICDD 00-051-1953).
Figure S9. X-Ray diffraction patterns (298 K) plotted against the angle 2θ of the crystals of CBZ inside the toluene gel of LG2-Lys and decylamine and the beta-phase (green lines) of CBZ (pattern calculated from CDCC entry CBMZPN10).

Figure S10. X-Ray diffraction patterns (298 K) plotted against the angle 2θ of the crystals of CBZ inside the toluene gel of DG2-Lys and decylamine and the alpha (blue lines) and beta-phases (green lines) of CBZ (patterns calculated from CDCC entries CBMZPN03 and CBMZPN10 for alpha and beta polymorphs respectively).
Figure S11. Partial VT $^1$NMR experiments performed with a 10 mM mixture of LG2-Lys dendron, hexylamine and CBZ in toluene-d8.

Figure S12. Partial VT $^1$NMR experiments performed with a 20 mM mixture of LG2-Lys dendron, decylamine and CBZ in toluene-d8.
Figure S13. (a) VT NMR quantification of the mobile components in a 1:1:1 mixture of LG2-Lys, C10NH₂, and CBZ; (b) Van’t Hoff plot of the solubilisation process of the 1:1:1 mixture of LG2-Lys, C10NH₂, and CBZ. All the measurements were performed at 20 mM and in toluene-d8.
Figure S14. Partial $^1$H NMR spectra (300 MHz, 298 K, 2 mM, toluene-d8) of decylamine upon tritrational addition of LG2-Lys dendron.

Figure S15. Isotherms resulting from the titration of decylamine with LG2-Lys dendron. The binding isotherms correspond to a 1:1 complexation model by using the adjacent methylene to the amine group (square) and the aliphatic chain (circle) protons.