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Supporting Information

Enhancement of Guest-Responsivity by Mesocrystallization of Porous Coordination Polymers

Akio Mishima, Tomomi Koshiyama, Jose A. Real and Masaaki Ohba

M. Ohba: ohba@chem.kyushu-univ.jp

1-bulk



1-mesoW5



1-mesoW3 $4 \longrightarrow -\Delta + 200 \times +\Delta + 200 \times 300 \times 300$

Figure S1 Thermal color changes of 1-bulk, 1-mesoW5 and 1-mesoW3.

1-bulk at 300 K



1-mesoW5 at 300 K



1-mesoW3 at 300 K



Figure S2 Responsivity of 1-bulk, 1-mesoW5 and 1-mesoW3 for CS_2 and AcCN.



Figure S3 Reflection spectra in the solid state of 1-bulk (a) and 1-mesoW5 (b): CS_2 clathrate (red line), AcCN clathrate (blue line) and guest-free sample in the HS state (black line).



Figure S4 PXRD patterns of (a) CS_2 clathrate and (b) AcCN clathrate of 1-bulk, 1-mesoW5 and 1-mesoW3



Figure S5 Magnetic behavior of 1-bulk (a) and 1-mesoW5 (b) clathrated AcCN



Figure S6 $\chi_M T$ versus *T* plots for these suspensions; 1-bulk (black), 1-bulk·2H₂O (blue), 1-bulk·4.8H₂O (green). 1-bulk·4.8H₂O prepared under high humidity condition (*P*/*P*₀ > 0.9) at 298 K showed no spin transition.



Figure S7 The photographs of 1-bulk and 1-mesoW5 aqueous suspensions at 300 K and 240 K

1-bulk in H_2O 1.0 mM + AcCN aq. (293K, 6h)



1-mesoW5 in H₂O 1.0 mM + AcCN aq. (293K, 6h)



Figure S8 Photographs of 1-bulk (top) and 1-mesoW5 (down) suspensions controlled varying AcCN concentration