Supporting Information

Stimuli-Responsive Hybrid Materials: Breathing in Magnetic Layered Double Hydroxides Induced by a Thermoresponsive Molecule

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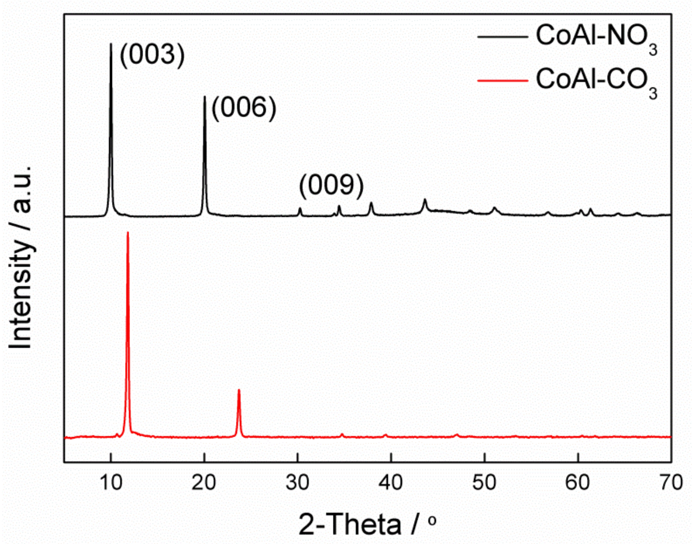
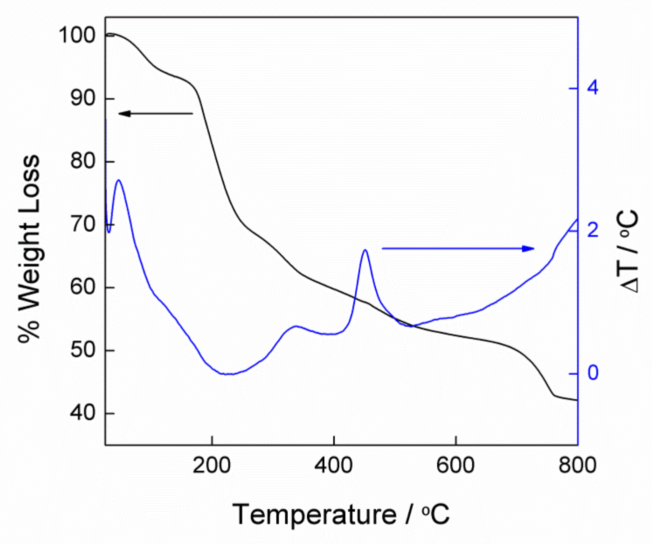
Keywords: photochemistry, azo compounds, magnetic properties, layered compounds, host-guest systems

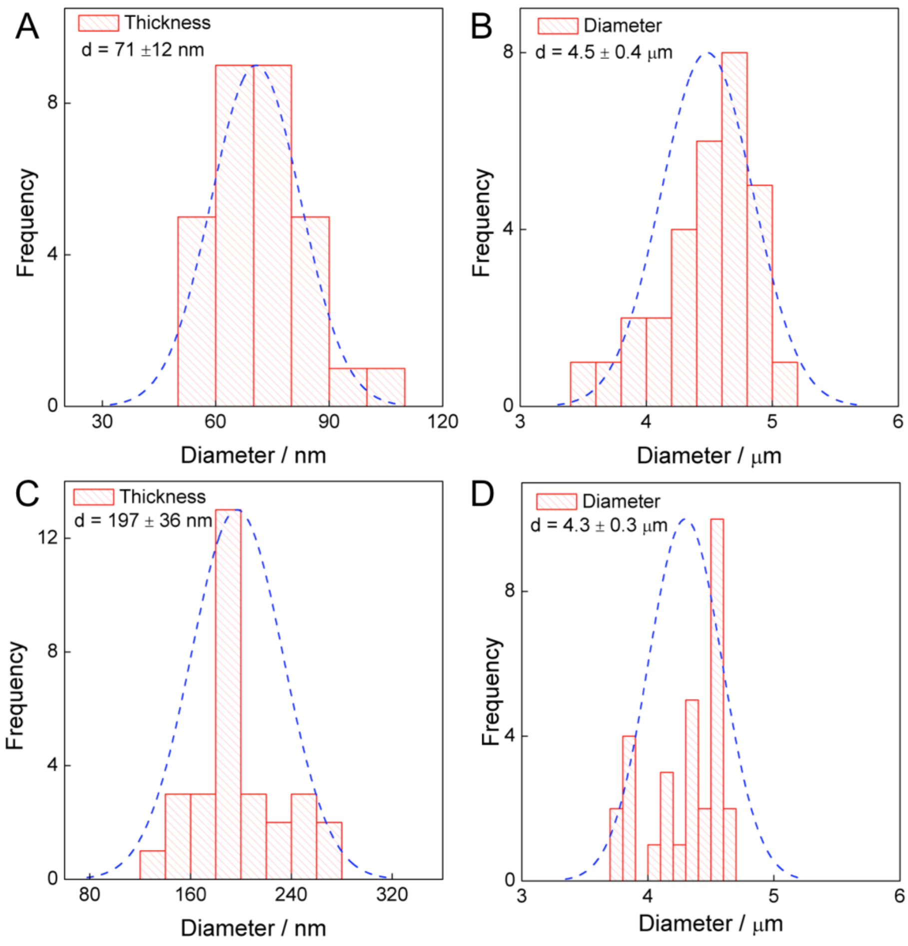
SI 1. X-ray powder diffraction patterns of CoAl–CO_3 and CoAl–NO_3.



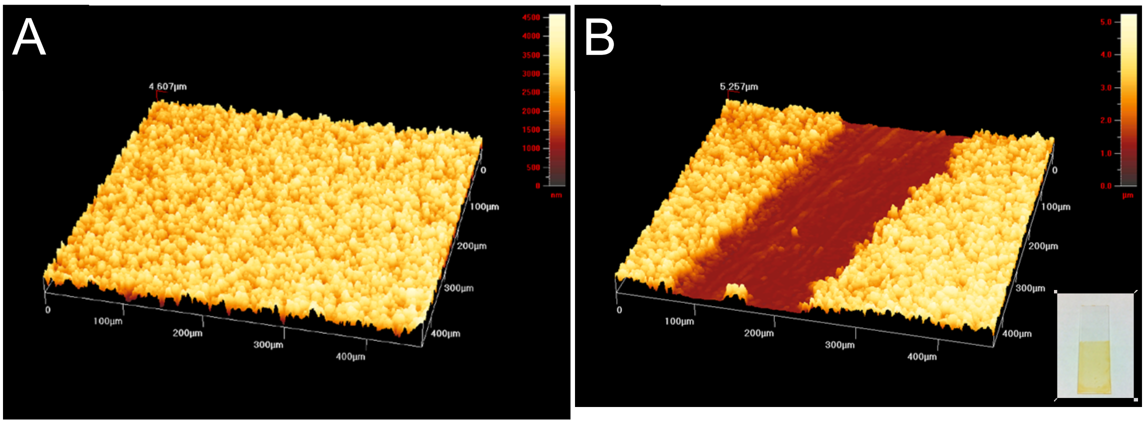
SI 2. Thermogravimetric (TGA/DTA) analysis. Thermal decomposition in air of CoAl–AO5 collected at a scan rate of 10 ºC·min^{-1}.



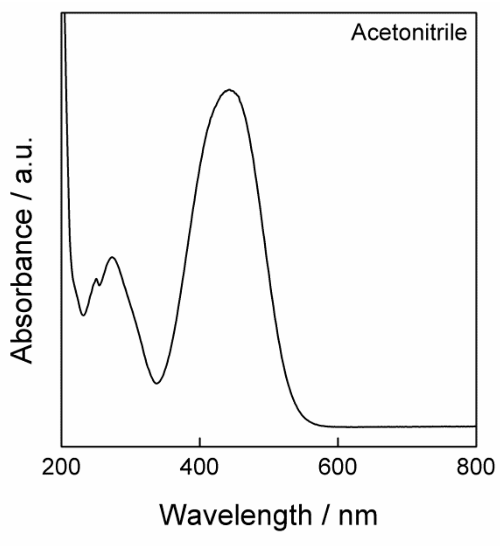
SI 3. Particle size distribution highlighting the average values for the diameter and thickness of CoAl–NO_3 (A and B) and CoAl–AO5 (C and D), respectively.



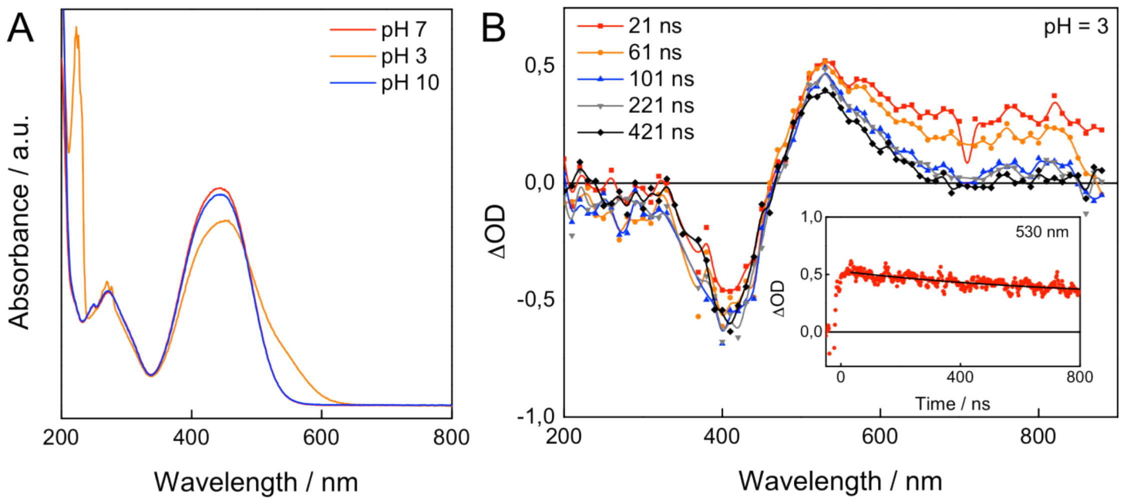
SI 4. (A) Surface and (B) profilometric characterization of the CoAl–AO5 films prepared using acetyl acetonate, α-terpineol and carboxymethyl cellulose. The inset presents a digital photograph of the as-obtained film on suprasil quartz substrate.



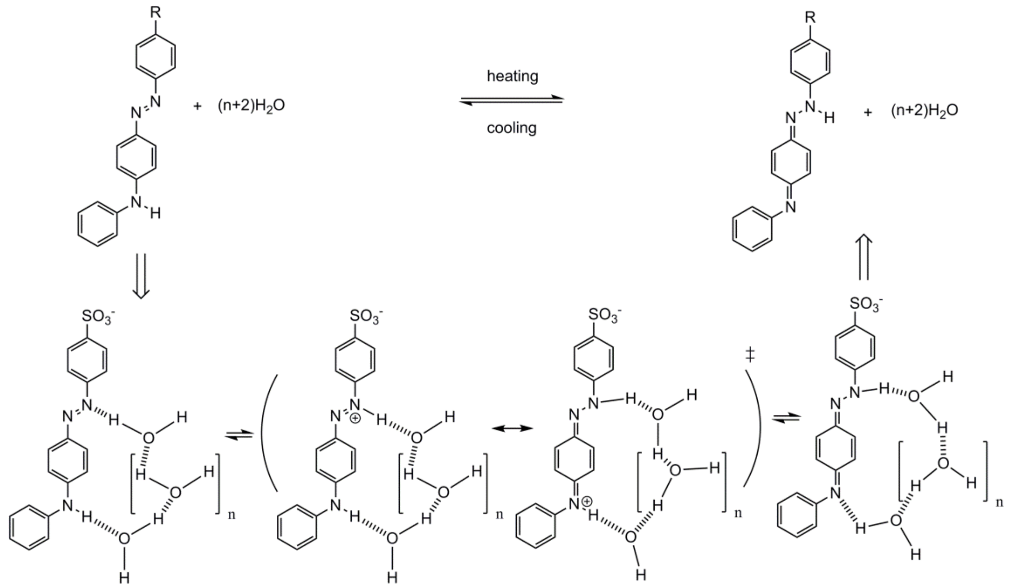
SI 5. UV-Vis absorption spectra of a solution of AO5 in acetonitrile.



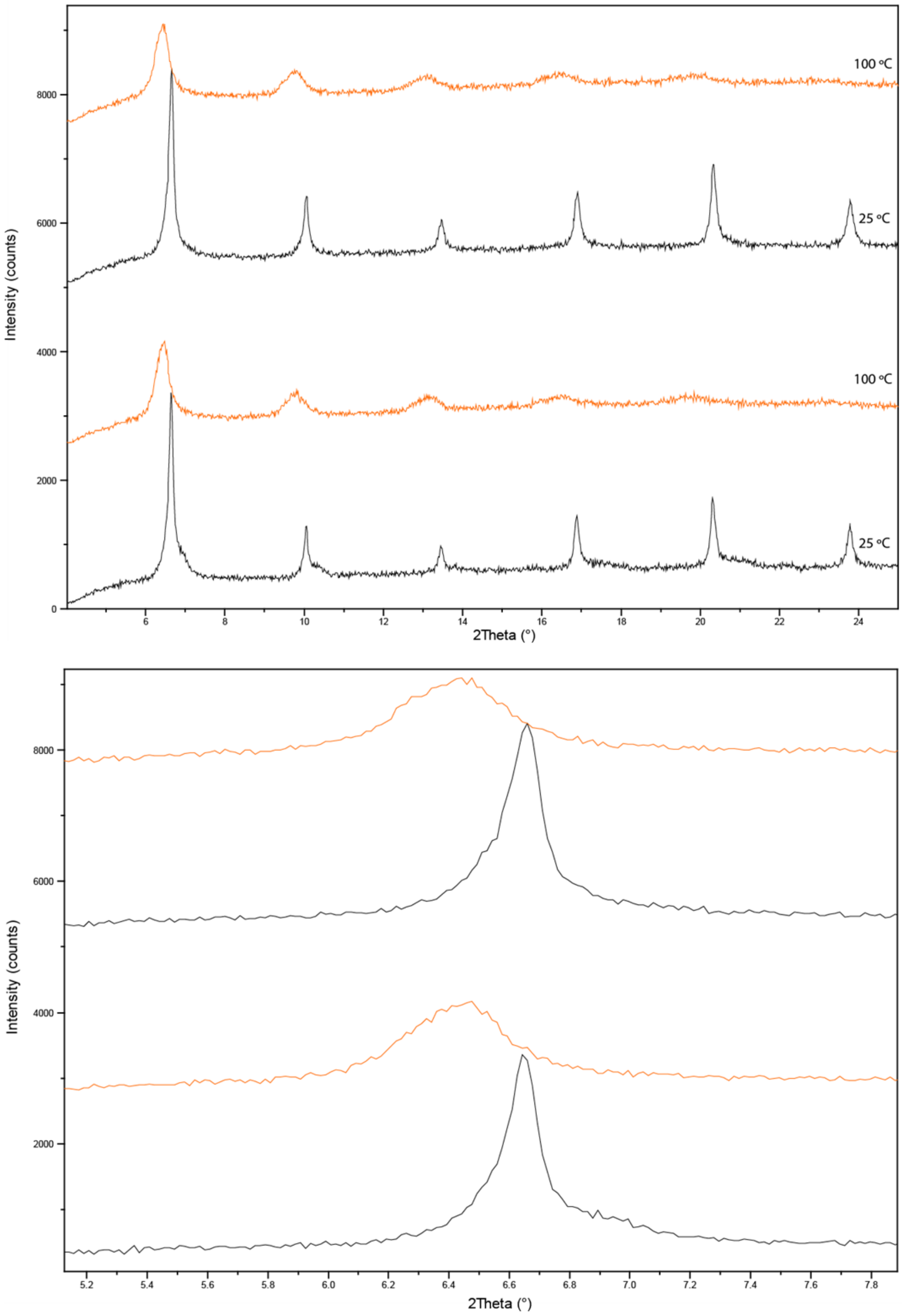
SI 6. (A) UV-Vis absorption spectra of aqueous solutions of AO5 at different pH, namely, 3 (ocre), 7 (red) and 10 (blue). (B) Laser flash photolysis absorption spectra of an acidic solution of AO5 (pH = 3), depicted spectral traces are from 21 to 421 ns after the laser pulse (λ_{exc} = 355 nm). The inset represents the decay spectra at 530 nm, highlighting the presence of long live transient ascribed to the formation of the *cis* isomer.



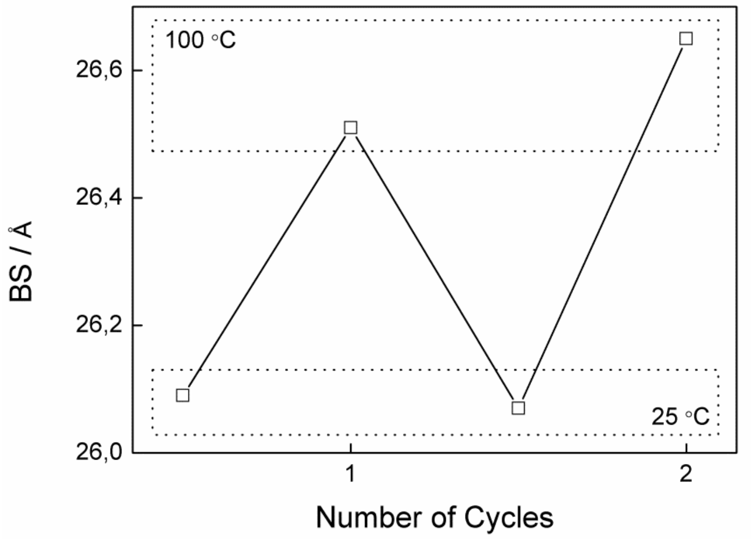
SI 7. Representation of the tautomerization of AO5 upon heating and cooling in the interlayer space of CoAl–AO5 following a Grotthuss-type proton migration mechanism. Adapted from Wang et al.[36]



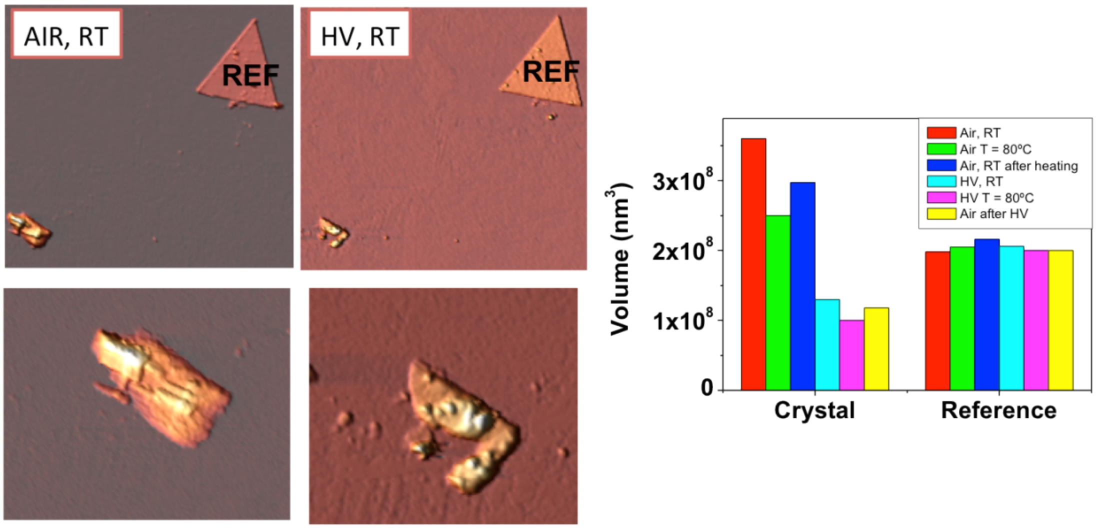
SI 8. Control experiment showing the reversible change in the XRD pattern of CoAl–SDS upon heating to 100 °C and cooling down to room temperature. The zoom-in highlights the movement and broadness increase of the (003) peak. In this case, the sample exhibits an increase in the basal space after heating.



SI 9. Control experiment showing the variation of the basal space of CoAl–SDS during two cycles of heating and cooling.



SI 10. AFM images of a crystal and a reference marker acquired at ambient conditions and in High Vacuum (HV) both at the same temperature (RT). As we can observe there is a clear change in the morphology as a consequence of the drastic water loss. In the graph we can observe the evolution of the volume of the same crystal and the reference marker for each measurement.



SI 11. Thermal variation of χ_m for the initial CoAl–AO5 (black), the corresponding heated (red) and final (blue) samples.