

Spin-Crossover Confinement in Mesoporous Matrices Enables Tunable Porosity for Molecular Sensing

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Área temática: A. Síntesis de nanomateriales

The integration of molecular functionality within hierarchically organized micro- and nanostructures represents a transformative strategy for the development of next-generation stimuli-responsive materials. This work addresses this challenge by coupling bistable spin-crossover (SCO) nanodomains with tunable mesoporous networks supported on a sustainable cellulose-paper scaffold. By employing a surfactant-templated sol-gel route with tetraethyl orthosilicate (TEOS) and Pluronic F127, we successfully functionalized cotton linter paper microfibers with an ordered mesoporous silica-based framework. This architecture provides the precise confinement necessary for the subsequent loading of the $[\text{Fe}(\text{II})(\text{Trz})_3]\text{BF}_4$ (Trz = 1,2,4-triazole) SCO complex. We demonstrate that sol-gel chemistry serves as the definitive enabling technology to establish a continuous structural connection from the microscale paper matrix down to the molecular level, creating a unique confinement-driven coupling between structural hierarchy and electronic bistability. Our results show that the mesoporous framework significantly modulates the thermodynamic behavior of the Fe(II) spin transition and induces strong cooperative interactions between the SCO molecular domains and the porous host. Notably, this hybrid self-assembly enhances accessible surface areas (a remarkable finding considering that the bulk SCO complex is inherently non-porous). Furthermore, the hybrid material exhibits guest modulated-responsive transitions, where molecular adsorption produces measurable shifts in the spin transition temperature by modifying the local coordination environments within the composite. These multiscale cooperative effects reveal a profound synergy between the storage capacity of the mesoporous host and the molecular responsiveness of the SCO complex. This research offers a robust and versatile platform for the design of adaptive, paper-based materials with high potential for application in advanced sensing, smart packaging, and responsive technologies.

REFERENCIAS

1. L. B. Pizarro, et. al. *Advanced Materials Interfaces* 13 (2026) e01103
2. J. J. Mikolei, et. al., *Nanoscale* 15 (2023) 9094–9105
3. A. Tissot, et al., *Chemical Communications* 55 (2019)194–197