

# Spin Crossover-Macromolecule Composite Nano Film Material

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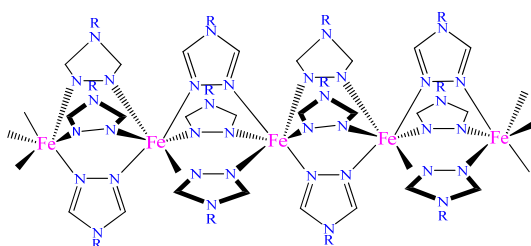
<sup>1</sup> *These authors contributed equally to this work.*

## Supporting Information

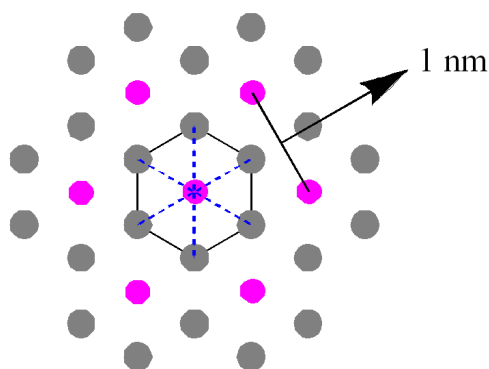
### Experimental Section

**General Remarks.** All synthesis were carried out under nitrogen using standard Schlenk techniques. Ethanol was purified and distilled under nitrogen before use. NH<sub>2</sub>trz and PVP were purchased from Alfa Aesar and used as received. The **IR spectra** were recorded on Bruker TENSOR-27 infrared spectroscopy at ambient temperature. The **XPS spectra** measurements were recorded using a Kratos Axis Ultra DLD spectrometer employing a monochromated Al-K $\alpha$  X-ray source, hybrid optics and a multi-channel plate and delay line detector at ambient temperature. The **X-ray diffraction** were performed on a D/Max-2500 X-ray diffractometer using Cu-K $\alpha$  radiation. **Small angle X-ray scattering** experiments were performed on a Bruker AXS NANOSTAR using a monochromatic X-ray beam and a two-dimensional detector for recording the scattering intensity. The distance between the samples and the detector was 270 mm and  $\lambda = 1.54 \text{ \AA}$  for the radiation beam. **Transmission electron micrographs (TEM) image** were taken at TecnaiG<sup>2</sup>20S-TWIN microscope. **Mössbauer spectra** were measured on MS-500 Mössbauer instrument at 295, 240 and 85 K. **Magnetic susceptibility measurements.** These were performed under a helium atmosphere using Quantum Design MPMS-XL SQUID susceptometer in 2000 Oe applied field at a temperature range of 10-300 K.

**Synthesis of the [Fe(NH<sub>2</sub>trz)<sub>3</sub>(ClO<sub>4</sub>)<sub>2</sub>]-PVP composite film.** An EtOH (10 ml) solution containing NH<sub>2</sub>trz (0.026 g, 0.3 mmol) and PVP (0.0355 g, 0.3 mmol) was added into an EtOH (10 ml) solution of Fe(ClO<sub>4</sub>)<sub>2</sub> (0.0255 g, 0.1 mmol), the mixture was stirred for 2 hours. Slight yellow transparent films were obtained after keeping the mixture in a vacuum desiccator for two days. The film can also be obtained by spin-coating on quartz substrate.

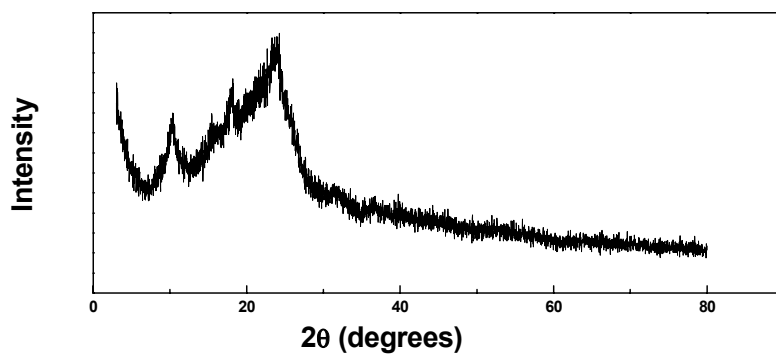


**Scheme S1.** Schematic structure of the [Fe<sup>II</sup>(4-R-trz)<sub>3</sub>](anion)<sub>2</sub> family.

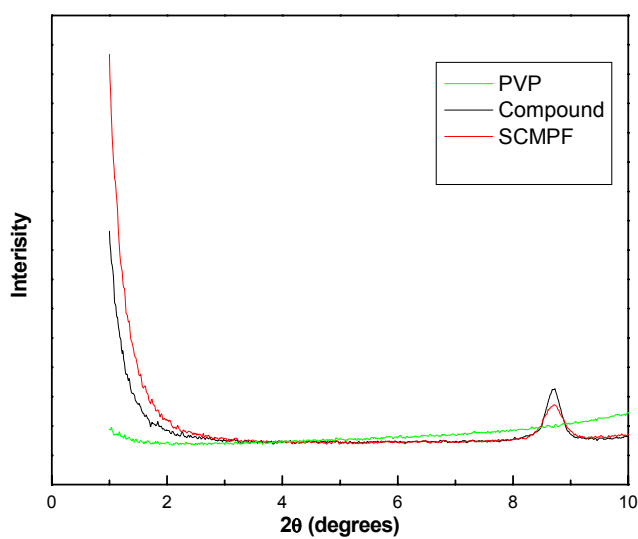


**Scheme S2.** View along the chains of PVP and the [Fe(NH<sub>2</sub>trz)<sub>3</sub>]<sup>2+</sup>. The [Fe(NH<sub>2</sub>trz)<sub>3</sub>]<sup>2+</sup> chains are in purple and the PVP chains are gray. The blue dashed lines are the hydrogen bonds between them. Parts of the structure are omitted for clarity. A hexagonal columnar structure is formed by the six PVP chains with one [Fe(NH<sub>2</sub>trz)<sub>3</sub>]<sup>2+</sup> in the middle. The distance between iron chains is *ca.* 1 nm ( $2\theta = 8.7^\circ$ ).

**Synthesis of the [Fe(Htrz)<sub>3</sub>(ClO<sub>4</sub>)<sub>2</sub>]-PVP film as comparison.** The procedure is similar as the synthesis of the [Fe(NH<sub>2</sub>trz)<sub>3</sub>(ClO<sub>4</sub>)<sub>2</sub>]-PVP composite film.



**Figure S1.** The XRD patterns of the film of  $[\text{Fe}(\text{Htrz}_3)(\text{ClO}_4)_2]\text{-PVP}$  which indicate the amorphous structure because of lack of hydrogen-bonds between the compound and PVP molecules.



**Figure S2.** The small angle X-ray scattering of PVP, compound and the composite film.