Article

Tuning the Spin-Crossover Behaviour in Fe(II) Polymeric Composites for Food Packaging Applications

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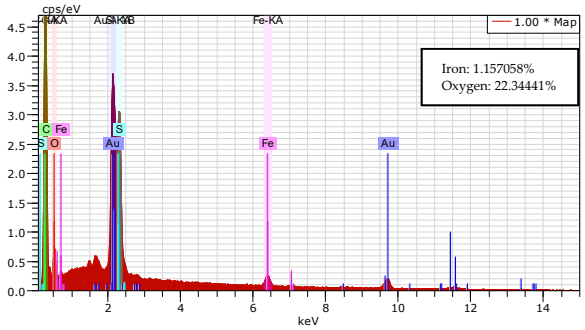
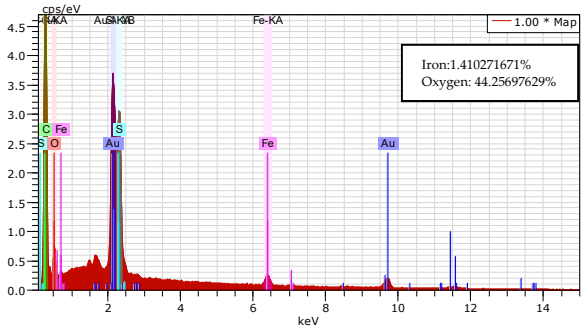
1. Composites' Preparation Procedure, Morphological Characterisation, and Migration Release Study

For the film preparation, a film casting method was used. For each composite, the polymer matrix (i.e., PLA and PSF) and the SCO compound (SCO1 and SCO2) were separately dissolved in CH_2Cl_2 (dichloromethane) under stirring. The percentage of SCO compounds in the composites was 10% w/w. The SCO solution was added to the polymer solution, and after 10 min of stirring, the solution was poured into a Petri dish which was left at room temperature for 24 h until solvent evaporation. The films were subsequently put in a vacuum oven equipped with an oil pump (T= 25 °C), in order to make sure that all the amount of solvent had been evaporated. The films' thickness was in the range of 100-120 μm.

The SCO:polymer percentage ratio of the composites was determined through EDX analysis. For quantification, the % percentage of Fe:O was calculated since Fe exists only in the SCO complex and O only in the two polymers. Note that each SCO complex possesses one Fe centre, while PLA and PSF, two and four O atoms, respectively. Therefore, the percentage Fe:O for PLA and PSF is anticipated to be approximately 5% and 2.5%, respectively. In Figure S1, the EDX analysis results for the PLA–SCO2 (**a**) and PSF–SCO2 (**b**) composites, which were further investigated by Raman spectroscopy, are provided.



(a) PLA–SCO2



(b) PSF–SCO2

Figure S1. Representative results from the EDX analysis for the PLA–SCO2 (**a**) and PSF–SCO2 (**b**) composites, where SCO2 is the complex $[Fe(SCN)_2(abpt)_2]$; the percentage of iron existing in the SCO2 and oxygen existing in the two polymers (PLA and PSF) is provided.

For the migration release studies (MRS), appropriate stainless steel migration cells systems, purchased from LABC-Labortechnik (http://www.labc.de/, accessed on 17 Sep. 2021), were used (Figure S2). The cells were placed inside a shaking incubator at stable speed (speed 50 rpm) and temperature (40 °C) conditions for 15 days. For the quantification of the potential release, 2 mL aliquots were withdrawn each time, and UV–Vis absorption spectra were obtained; the aliquots were subsequently returned back in the cell after the measurements. The corresponding concentration calculations of the migrated quantity were calculated based on the calibration curve of the SCO materials (Figure S2).

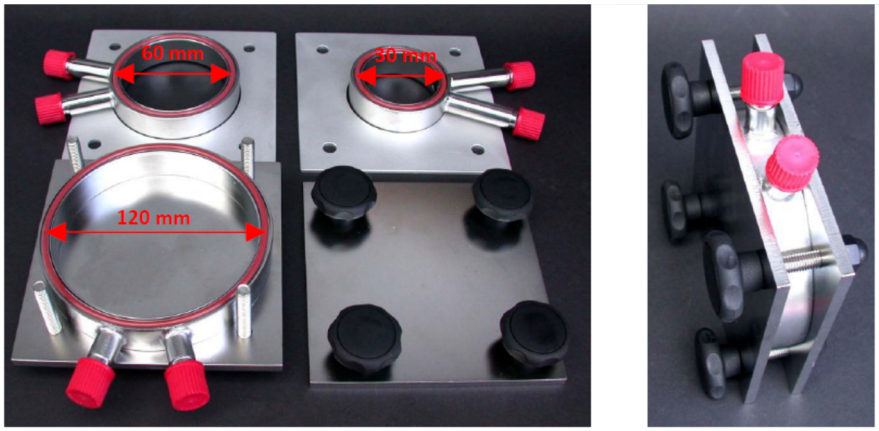


Figure S2. (Left) The components of the migration cells. Cells of three different geometrical characteristics; the cell possessing 30 mm diameter was used for the migration experiments. (Right) A typical assembly of the cell that is ready to be put into the incubating chamber.

In Figure S3, the UV–Vis spectra of solutions in 50% EtOH (left) of the SCO compounds SCO1 (**a**) and SCO2 (**c**) and the subsequent calibration curves ((**b**) and (**d**), respectively) are shown.

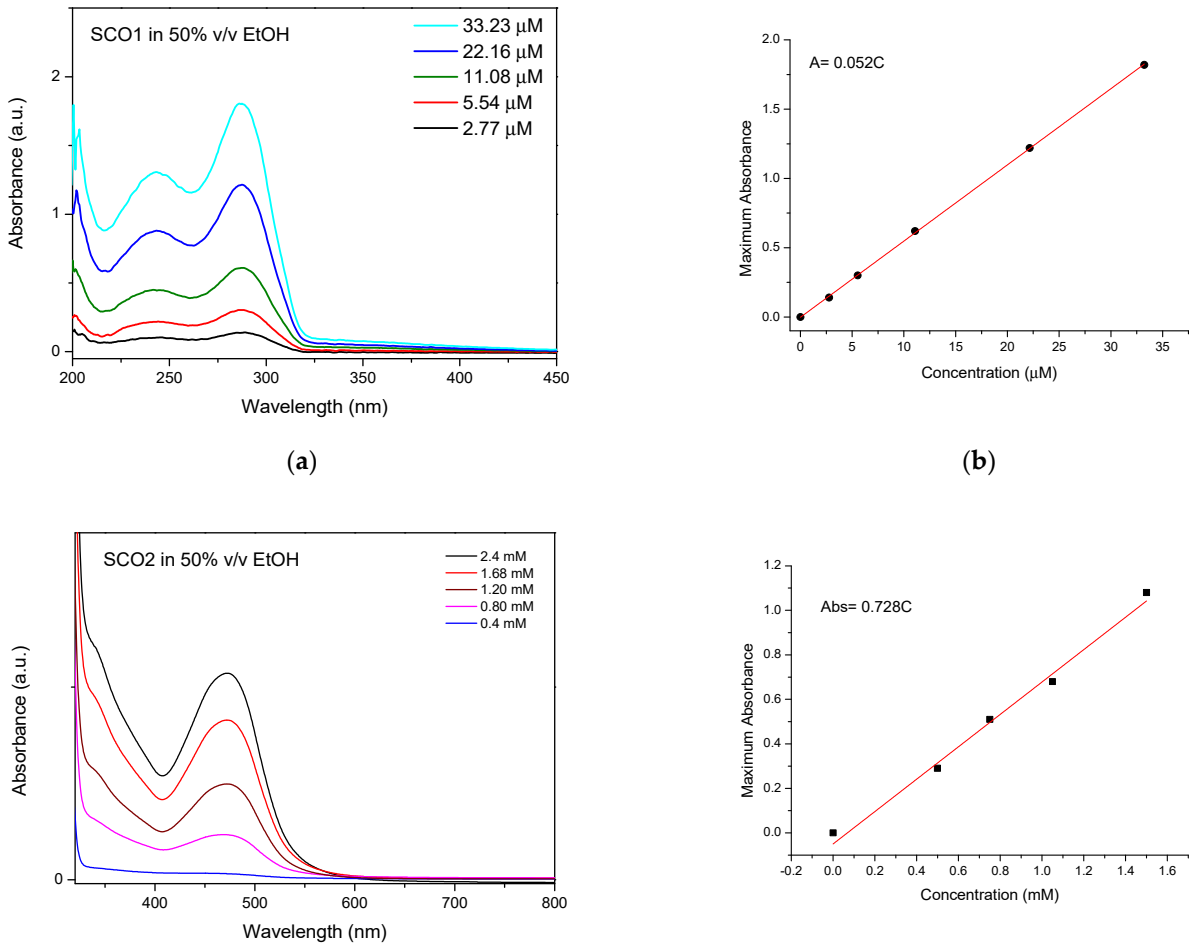
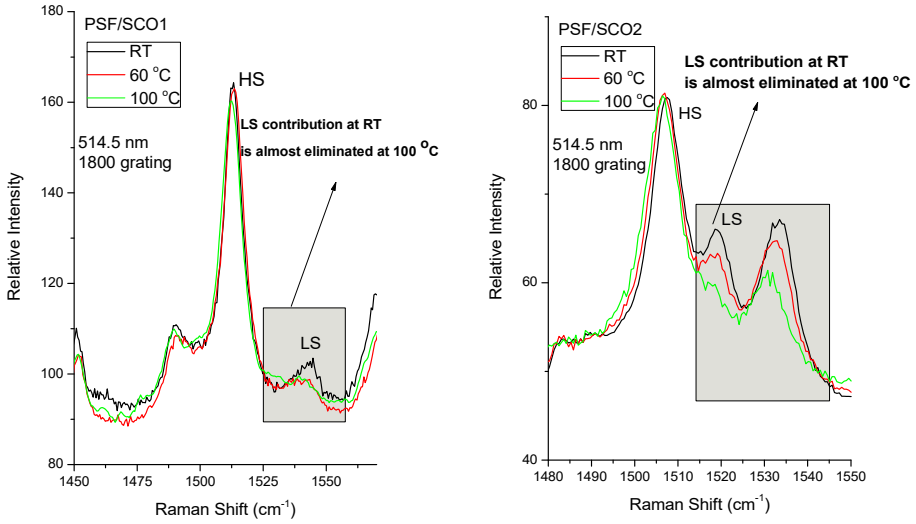


Figure S3. The UV–Vis spectra of the SCO coordination complexes SCO1 and SCO2 ((**a**) and (**c**), respectively) in 50% EtOH for various concentrations and the subsequent calibration curves based on the maximum absorbance value ((**b**) and (**d**), respectively).

2. Low-Spin (LS) Contribution at Room Temperature

In Figure S4, the Raman spectra of the composites PSF–SCO1 (**a**) and PSF–SCO2 (**b**) are presented. In these spectra, a contribution of the LS state was observed. By increasing the temperature, this contribution was eliminated.



(**a**) (**b**)

Figure S4. The disappearance of the LS contribution in the Raman spectra of the PSF–SCO1 (**a**) and PSF–SCO2 (**b**) composites with increasing temperature.