Characterization of Europium-Doped Silica Thin Films as Luminescent Materials by Small Angle X-Ray Scattering

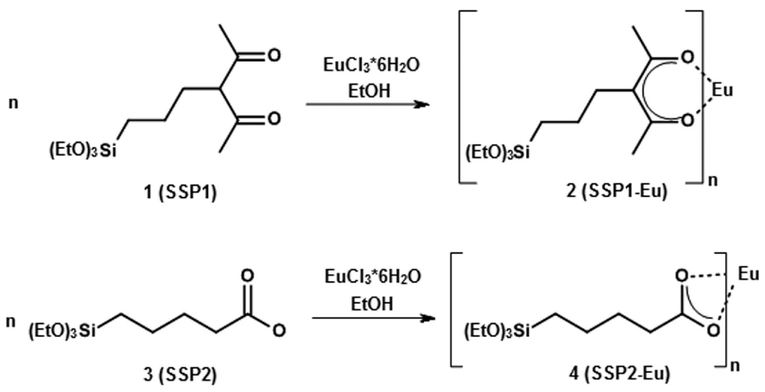
J. Akbarzadeh*; A. Feinle†; F. Lavoie-Cardinal§; C. Wickleder§; N. Hüsing†; H. Peterlik*

*University of Vienna, Faculty of Physics, Dynamics of Condensed Systems, Austria
†Paris-Lodron University Salzburg, Materials Chemistry, Austria
§ University of Siegen, Inorganic Chemistry II, Siegen, Germany

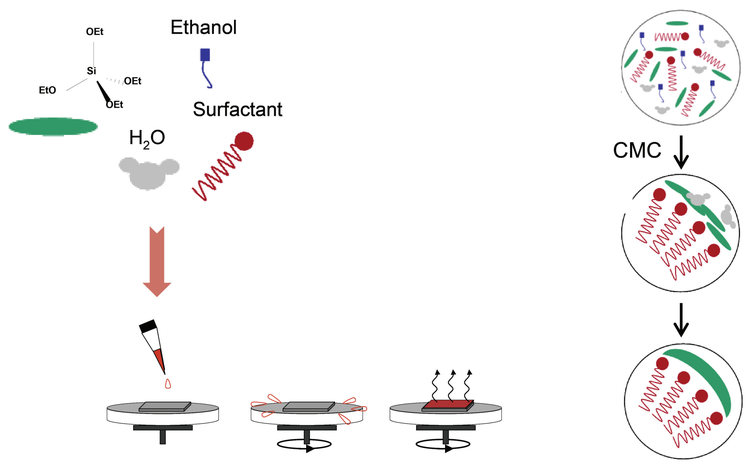
Abstract

The unique luminescence properties of the rare earth elements allow the development of various technological devices. Especially Eu^{3+} ions are widely used as a red phosphor in television sets, energy saving and discharge lamps, sensors, and also in the anti-counterfeiting materials in Euro banknotes due to their sharp emission peak at approximately 611 nm. The wide field of application of the rare earth elements, in our daily life is the reason for the world wide increasing interest during the last years.
A still remaining challenge in the production of luminescent materials is to achieve long-time high thermal and photochemical stability of the final materials. It can be assumed that the incorporation of the lanthanide ions into an inorganic mesoporous host, e.g. silica matrix, will improve the properties as well as the stability to a high extent.
The investigated samples in this work were prepared via a solvent evaporation-induced self-assembly (EISA) approach of different single-source precursors (SSP) in the presence of Pluronic™ P123 acting as a structure-directing agent.
The influence of the different SSPs and the different Si/Eu^{3+} ratios on the final mesostructure of the films has been investigated by small-angle X-ray scattering (SAXS) and grazing incidence small angle X-ray scattering (GISAXS).

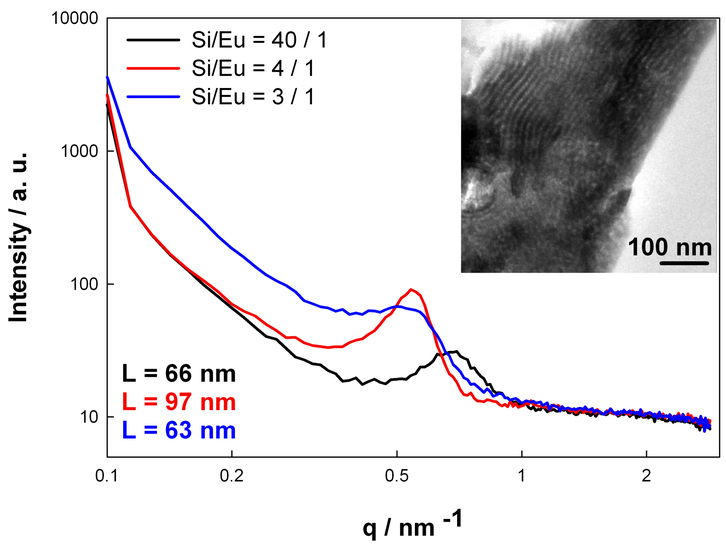
Synthesis of Metal-Coordinated Eu^{3+} Single-Source Precursor (SSP) Molecules



The Evaporation-Induced Self-Assembly (EISA) Method



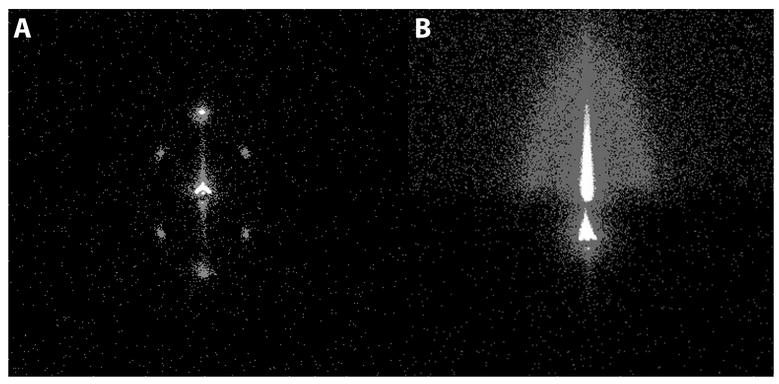
SAXS curves



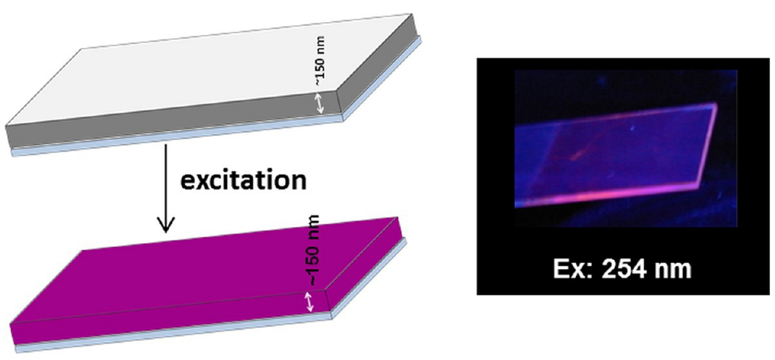
SAXS and GISAXS measurements

Right figure: GISAXS pattern of (A) a pure TEOS-coated silicon wafer (B) a wafer coated with the pure single-source precursor SSP2-Eu (Si/Eu = 3/1) The peak broadening is due to the small domain size.
Left figure: SAXS curves of Eu^{3+}-doped silica coatings using tetraethoxysilane (TEOS) and different amounts of the Eu^{3+}-coordinated organosilane (SSP2). The full width at half maximum (FWHM) of the short range order peaks allow the calculation of the domain sizes L.

GISAXS pattern



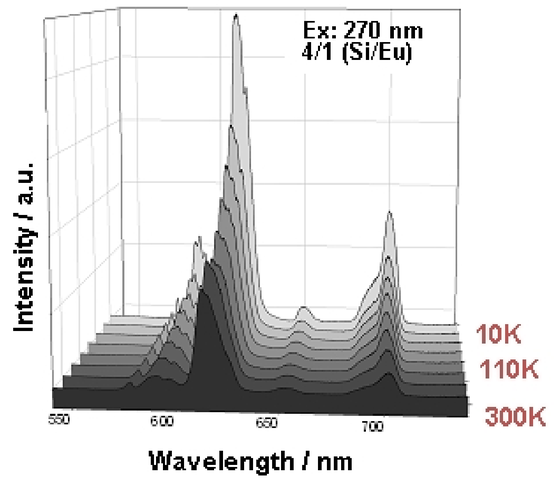
UV-Excitation



Luminescence Properties

Right figure: The photoluminescence spectra show characteristic emission peaks corresponding to the $^5D_0 \rightarrow {}^7F_J$ $(J = 0 - 5)$ transitions although the films have a very low thickness of 150 to 200 nm resulting in a red luminescence visible by eyes.
Left figure: Red emission of a coating with a Si/Eu ratio of 7/1 (12.5 mol % SSP1-Eu)

Luminescence Spectrum



Conclusions

- Porous Si/Eu^{3+} mixed metal oxide coatings on glass were successfully prepared via an EISA process using novel SSPs in the presence of structure-directing agents.
- The final Eu^{3+}-containing silica materials show a reduced ordering of the pores in the mesoscopic range and reduced specific surface areas compared to pure silica coatings.
- The thin coatings show luminescence properties with characteristic emission peaks according to the $^5D_0 \rightarrow {}^7F_J$ $(J = 0 - 5)$ transitions.
- The fact that the emission also is clearly visible for very thin films (150-200 nm) demonstrates the efficiency of this new type of Eu-containing material.

Literature

Feinle A.; Lavoie-Cardinal F.; Akbarzadeh J.; Peterlik H.; Adlung M.; Wickleder C.; Hüsing N.; *Chemistry of Materials* **2012**; DOI: 10.1021/cm300996j

Acknowledgements

The Austrian science foundation FWF (Proj.Nr. I449) is acknowledged for financial support. A. F. acknowledges the financial support of the Deutsche Forschungsgemeinschaft (No. HU 1427/4-1).