

Abstract No. giba0263

Time-resolved *In Situ* Grazing Incidence Small Angle X-ray Scattering Experiment of Evaporation Controlled Self-assembly

A. Gibaud⁺, D. Grosso⁺⁺, B. Smarsly^{*}, A. Baptiste⁺, J. F. Bardeau⁺, J. Brinker^{*}, D. Doshi^{*}, Z. Chen^{*}, F. Babonneau⁺⁺, and C. Sanchez⁺⁺,

⁺ LPEC Le Mans France, ⁺⁺ LCEC Jussieu France, ^{*} Univ. of NM USA, [‡] Sandia National Lab
Beamline(s) : X22A

The synthesis of silica-based mesostructured materials by using supramolecular self-assembly of surfactant molecules to template the condensation of inorganic species has attracted considerable interest in the past decade¹. Geometrical considerations show that above the CMC (Critical Micelle Concentration), surfactant molecules can self-assemble into spherical, cylindrical or lamellar shapes micelles². As shown in 1992, by researchers of the Mobil Corporation micelles can further self assemble into well-organized 2D or 3D mesostructural phases³⁻⁴ and template the condensation of inorganic materials. Mesoporous phases with tailored porosity^{5,6} can be obtained by thermally removing the surfactant. Recently, there have been some attempts of in-situ characterization, using resolved fluorescence-depolarization experiments or in-situ luminescence of probe molecules but those techniques do not provide any information on the organization of the film⁷. We report here a GISAXS experiment of the influence of the rate of evaporation of the solvent together with the composition and aging time of the sol. Despite the complexity of this process, we present a first insight of the evaporation role in the meso-structuration. This is made possible by studying the dynamics of organization using in-situ time-resolved GISAXS experiments performed during thin and thick films processing from strictly identical initial sols but in different conditions of evaporation. Starting from a selected solution, we have observed as shown in figure 1 that one can obtain either the 2D-hexagonal (p6m), the 3D-hexagonal (P6₃/mmc), the cubic (Pm3n) or mixed structures depending on the evaporation conditions. This suggests that a one fixed sol composition (same aging time) is not attached to a single final hybrid mesophase but more to the pathway taken during the ECSA.

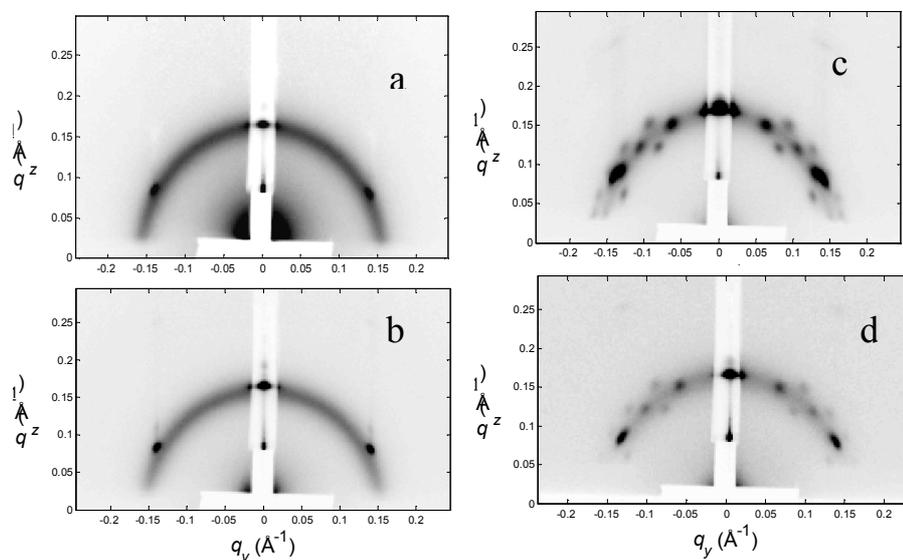


Figure 1: Observation of a 2D hex pattern for fast assisted evaporation rate 2a, 2b and of a cubic phase for slower evaporation rates 2c, 2d.

Acknowledgements : This work was supported by the UNM/NSF Center for Micro-Engineered Materials, the French ACI "Nanostructure" under project N° 03-01, the AFOSR, the DOE Basic Energy Sciences Program and SNL's Laboratory Directed R&D program. Research carried out in part at the National Synchrotron Light Source, Brookhaven National Laboratory, which is supported by the U.S. Department of Energy, Division of Materials Sciences and Division of Chemical Sciences.

REFERENCES

- 1- For a recent review see Ying, J.Y., Mehnert, C.P. and Wong, M.S. *Angew. Chem. Int. Ed.*, 38, 56-77, 1999
- 2- Israelachvili, J., "Intermolecular and Surface forces" 2nd Ed., Academic Press, London 1992.
- 3- Kresge, C.T., Leonowicz, M.E., Roth, W.J., Vartuli, J.C., and Beck, J.S., *Nature*, 359, 710-712, (1992)
- 4- Beck, J.S., Vartuli, J.C., Roth, Leonowicz, M.E., W.J., Kresge, C.T., Schmitt, K.D., Chu C.T.W., Olson, D.H., Sheppard, E.W., McCullen, S.B., Higgins, J.B. and Schlenker J.L. *J. Am. Chem. Soc.*, 114, 10834-10843, (1992)
- 5- Lu, Y., Cangull, R., Drewlen, C.A., Anderson, M.T., Brinker, C.J., Gong, W., Guo, Y., Soyez, H., Dunn, B., Huang and Zing, J.I. *Nature*, 389, 364, (1997).
- 6 - Zhao, D., Yang, P., Melosh, N., Feng, J., Bradley, B.F., Cmelka, F. and Stucky, G.D., *Adv. Mater.*, 10, 1380, (1998).
- 7- Chen, C.Y., Burkett, S.L., Li, H.X. and Davis, M.E., *Microporous Mater.*, 2, 27-34, (1993)