ESRF	**Experiment title:** **Novel type of metal-supported catalysts characterised by anomalous small-angle X-ray scattering**	**Experiment number**: SC1489
Beamline: ID1	**Date of experiment**: from: 13/02/2005 to: 17/02/2005	**Date of report**: 25/02/06
Shifts: 9	**Local contact(s)**: Peter Bösecke	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Dingenouts, Nico*;

Polymer Institut, Universität Karlsruhe, Kaiserstr. 12, 76128 Karlsruhe, Germany

Ballauff, Matthias; Rosenfeldt, Sabine*; Haupt, Bjoern*;

Physikalische Chemie I, Universität Bayreuth, Universitätsstraße 30, D-95440 Bayreuth, Germany

Report:

In this experiment we have investigated new metal-based catalysts with anomalous small-angle X-ray scattering (ASAXS). In co-operation with the university of Stuttgart we have prepared metal-supported catalysts using supercritical fluid reactive deposition (SFRD) and platinum as metal component (Pt, L-III edge at 11563.7 eV). As shown by some preliminary tests [Watkins et al., 1999], this new method is able to achieve an extremely fine dispersion of the metal component and therefore results in a high reactivity for the catalyst-material.

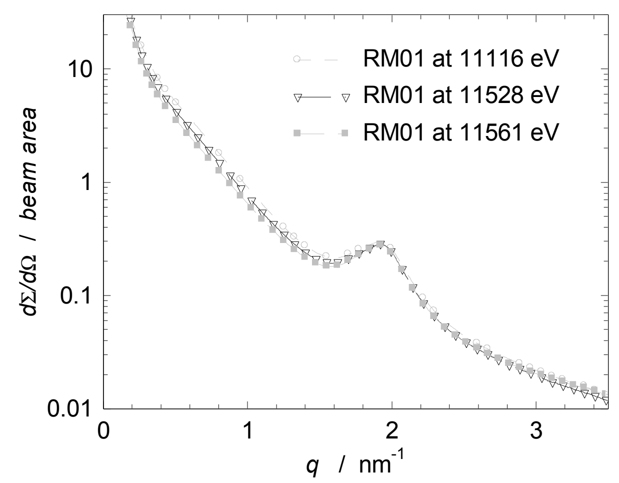

Fig. 1 Scattering intensity of sample RM01 at different energies of the primary beam

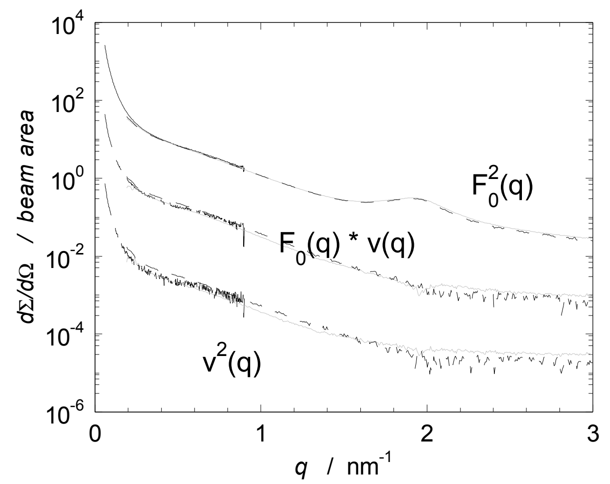

Fig. 2 Comparison of three separations according eq. 1 from three independent measuring runs shows the good reproducibility of the separation

We prepared sample using different substrates using different conditions and methods for the deposition of the metal component. For all samples, we measured 5 energies near the L-III edge of Pt. Fig1. shows the resulting scattering intensity for a sample with a high metal volume fraction.

After performing a measurement at at least three different energies, the measured ASAXS intensity can be modelled in term of eq.(1):

$$I(q) = F_0^2(q) + 2f'(E)F_0(q)v(q) + [f'(E)^2 + f''(E)^2]v^2(q) \qquad (1)$$

The first term is the scattering intensity of a single particle far from the edge and can be rendered as the square of the (real) scattering amplitude $F_0(q)$. The second term is the cross term between $F_0(q)$ and the Fourier-transform v(q) of the radial distribution of the metal ions.

As already shown in previous ASAXS investigations of colloidal and polyelectrolyte systems carried out at the ESRF (Dingenouts et al., 2004), we could shows also for the catalyst materials the possibility to get not only the cross term but all three scattering terms of eq. 1. Fig. 2 shows the resulting separation for one sample. The comparison of independent measurements included in Fig. 2 shows the excellent reproducibility of the experiment and the separation.

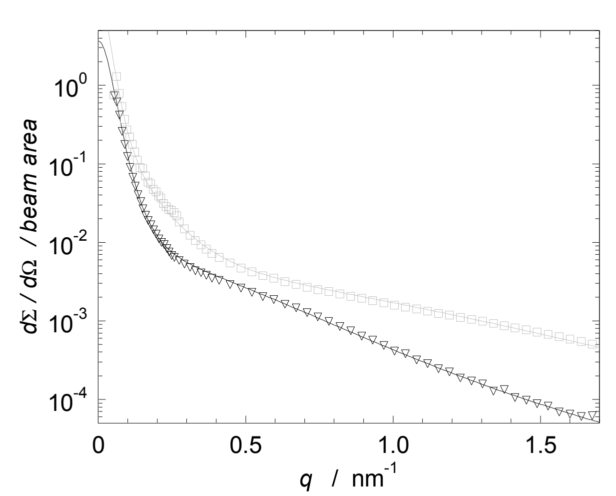

Fig. 3 Selfterm $v^2(q)$ for a sample with conventional preparation (black) and with SFRD (grey). Solid lines shows theoretical scattering based on the particle distribution shown in fig.4 .

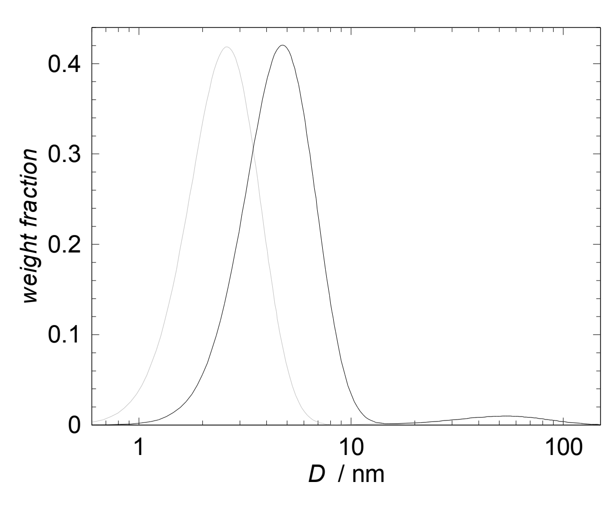

Fig. 4 Particle Size Distribution for or a sample with conventional preparation (black) and with SFRD (grey). Scattering intensities see fig. 3.

The last step in the evaluation process is the determination of the particle size and the distribution of the metal component. Therefore the only necessary scattering term is the self term v(q) of the metal particles. Fig. 3 shows this scattering term for two samples prepared by different deposition methods. A direct calculation of the particle size distribution was not possible due to the available scattering vector regime but it was possible to adjust a bimodal particle size distribution (fig. 4) so that the corresponding scattering intensity (fig. 3) reproduce the experimental scattering intensity.

By this evaluation method we could show that all samples have a bimodal particle size distribution consisting off a small amount of big particles and a high amount of small particles (see. fig. 4). In addition, we were able to demonstrate that preparation by supercritical fluid reactive deposition (SFRD) results in smaller metal particles, e.g. an improved dispersion of the metal component. A more detailed analysis and a first publication are in preparation.

N. Dingenouts, M. Patel, S. Rosenfeldt, D. Pontoni, T. Narayanan , M. Ballauff; *Macromolecules,* **37**, (2004) 8152-8159.

Watkins, J.J.; Blackburn, J.M.; McCarthy, T.J, (1999) *Chem. Mater.* **11**, 213-215.