

## Experiment Report Form

**The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.**

Once completed, the report should be submitted electronically to the User Office via the User Portal:

<https://www.esrf.fr/misapps/SMISWebClient/protected/welcome.do>

### ***Reports supporting requests for additional beam time***

Reports can be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

### ***Reports on experiments relating to long term projects***

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

### ***Published papers***

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

### **Deadlines for submission of Experimental Reports**

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

### **Instructions for preparing your Report**

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.



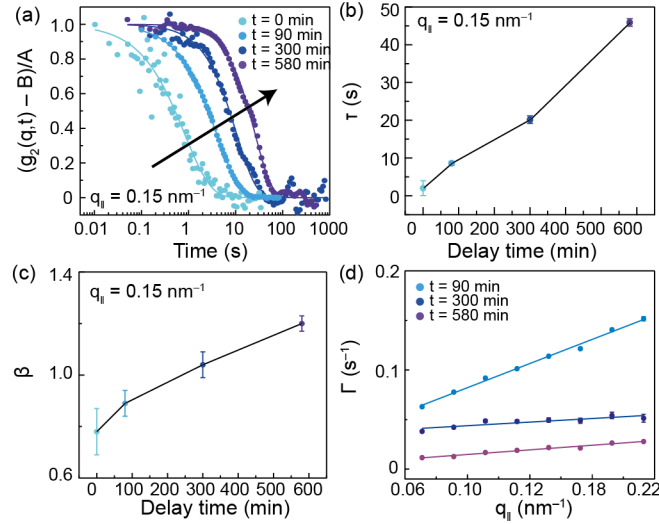
	<b>Experiment title:</b> Unraveling Gelation of Biopolymer at Oil/Water Interface by Using Grazing-Incidence X-Ray Photocorrelation Spectroscopy (GI-XPCS)	<b>Experiment number:</b> SC 4949
<b>Beamline:</b> ID 10	<b>Date of experiment:</b> from: 14.11.2018 to: 20.11.2018	<b>Date of report:</b> 13.02.2019
<b>Shifts:</b> 18	<b>Local contact(s):</b> Yuriy Chushkin	<i>Received at ESRF:</i>
<b>Names and affiliations of applicants</b> (* indicates experimentalists): Federico Amadei <sup>1,*</sup> and Motomu Tanaka <sup>1,2</sup>  <sup>1</sup> Physical Chemistry of Biosystems, Institute of Physical Chemistry, Heidelberg University, D69120 Heidelberg, Germany  <sup>2</sup> Institute for Integrated Cell-Material Science (WPI iCeMS), Kyoto University, 606-8501 Kyoto, Japan		

## Report:

The primary goal of the proposed experiment is to quantitatively investigate the sol-gel transition kinetics of sodium alginate caused by the diffusion to the interface of calcium chloride nanoparticles suspended in oil and surfactant by means of GI-XPCS.

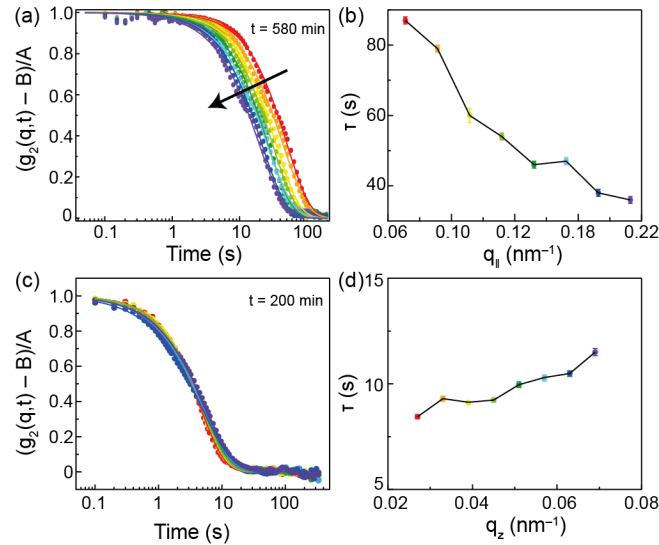
The change in the interface dynamics was followed by monitoring the time evolution of the autocorrelation function (ACF):  $g^2(q, t) \sim \exp[-2(t/\tau_q)^\beta]$  of silanized SiO<sub>2</sub> nanoparticles ( $\emptyset \sim 100$  nm) confined at the oil water interface.

Thanks to the knowledge and experience acquired during the previous beam time (SC 4654, February 2018), in this instance we were able to overcome the challenges dictated by the complexity of our system, which led us to obtain promising data to better understand the 2D gelation dynamics of polyalginate at the oil/water interface. We focused on the gelation of 1.5 wt% polyalginate by the addition of cross-linkers (CaCl<sub>2</sub> nanoparticles) suspended in vegetable oil. We monitored how the dynamics at a microscale changed and eventually slowed down by acquiring data at different time points. Figure 1a shows a selection of normalized autocorrelation function at different time points and at constant  $\mathbf{q}_{\parallel}$ . Interestingly, as the gelation proceeds, the characteristic decay time of the autocorrelation functions increases (Fig. 2b), which is accompanied by the increase of the form factor  $\beta$  (Fig 3c) which is reflected in a more prominent shoulder-like profile in the autocorrelation function. Last but not least, the relaxation time  $\Gamma$  plotted vs  $\mathbf{q}_{\parallel}$  suggests the suppression of interface dynamics by gelation.



**Figure 1.** (a) Normalized intensity autocorrelation functions (ACFs) measured at  $t = 0, 90, 300,$  and  $580$  min after the injection of  $\text{CaCl}_2$  NP. at  $q_{\parallel} = 0.15 \text{ nm}^{-1}$ . Solid lines represent the fits obtained with equation (XX). Temporal evolution of the (b) structural relaxation time  $\tau$  and (c) form factor  $\beta$ . (d) Relaxation rate  $\Gamma$  plotted vs.  $q_{\parallel}$  at  $t = 90, 300$  and  $580$  min, implying the suppression of interface dynamics by gelation. Solid lines represent linear regressions.

Furthermore, the dependency of the relaxation time on  $q_{\parallel}$  and  $q_{\perp}$  was investigated. Fig. 2 shows how the ACF decay time depends strongly on  $q_{\parallel}$  whereas it is almost negligible on the account of  $q_{\perp}$ .



**Figure 2. Dependence of the relaxation time  $\tau$  on  $q_{\parallel}$  and  $q_{\perp}$ .** (a) Normalized intensity ACFs at  $t = 580$  min. ACFs collected at various  $q_{\parallel}$  are presented color-coded. (b) Relaxation time  $\tau$  plotted as a function of  $q_{\parallel}$ . (c) Normalized intensity ACFs at  $t = 200$  min. ACFs collected at various  $q_{\perp}$  are presented color-coded. (d) Relaxation time  $\tau$  plotted as a function of  $q_{\perp}$ .

These findings suggest that the dynamics of nanometric domains at the oil/water interface slows down as the sol-gel transition proceeds.

Further experiment and data analysis are needed to shed light on the impact of polymer and cross-linker concentration on the reaction dynamics at the oil/water interface.