



	Experiment title: Novel strain-adaptive molecular bottlebrush-based bioadhesives and injectable elastomers	Experiment number: SC-4982
Beamline: ID02	Date of experiment: from: 29.10.20 to: 01.11.20	Date of report: 13.01.21
Shifts: 9	Local contact(s): Michael Sztucki , Peter Boesecke	<i>Received at ESRF:</i>
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Report:

Currently, bottlebrush polymers are gaining importance as reprocessable and extremely soft materials for various applications[1,2]. There is a lot of discussion about how such systems self-organize into semi-ordered structure and what factors drive this organization. In present work we have studied a series of bottlebrush-based polymers by means of SAXS/WAXS combined with mechanical stretching and swelling of chemically cross-linked elastomers. We have studied two types of systems: chemically heterogeneous bottlebrushes where both side-chain and backbone are based on acrylic polymers and chemically homogeneous where backbone is acrylic and side-chain is PDMS-based.

Main structural feature is bottlebrush backbone-to-backbone correlation peak[3,4] which was previously studied by us. Here, we continue our investigation into the nature of this correlation. During this experiment we have found that loosely grafted heterogeneous brushes does not exhibit such correlation peak. This can be explained by the absence of electron density contrast, because side-chain conformation does not allow to form dense bottlebrushes, because there are not enough side-chains to form an envelope. This effect is shown in Figure 1, where one can observe scattering curves for PBA brushes with constant side-chain lengths and decreasing grafting density (increasing n_g). It is noteworthy that correlation peak almost vanishes for loosely grafted samples, which is not the case for heterogeneous bottlebrushes, where phase separation between side chain and backbone does not allow them to mix and form uniform density.

In addition we have studied structural variations upon mechanical stretching. It was found, that upon elongation bottlebrush correlation peak exhibits strong orientation with minimas located in the stretching direction. That confirms our suggestion that this peak arises from backbone-to-backbone correlation. This result is in line with our previous experiments.

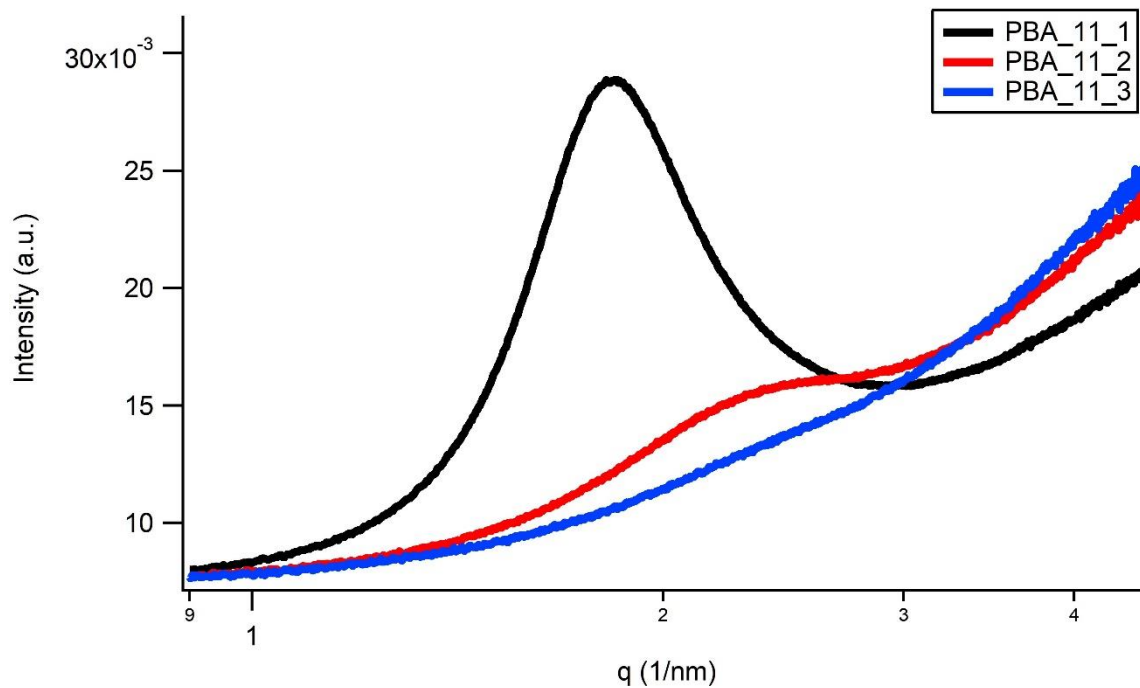


Figure 1. Scattering curves for samples with constant side chain length and varied grafting density for samples PBA_{n_{sc}}_{n_g}

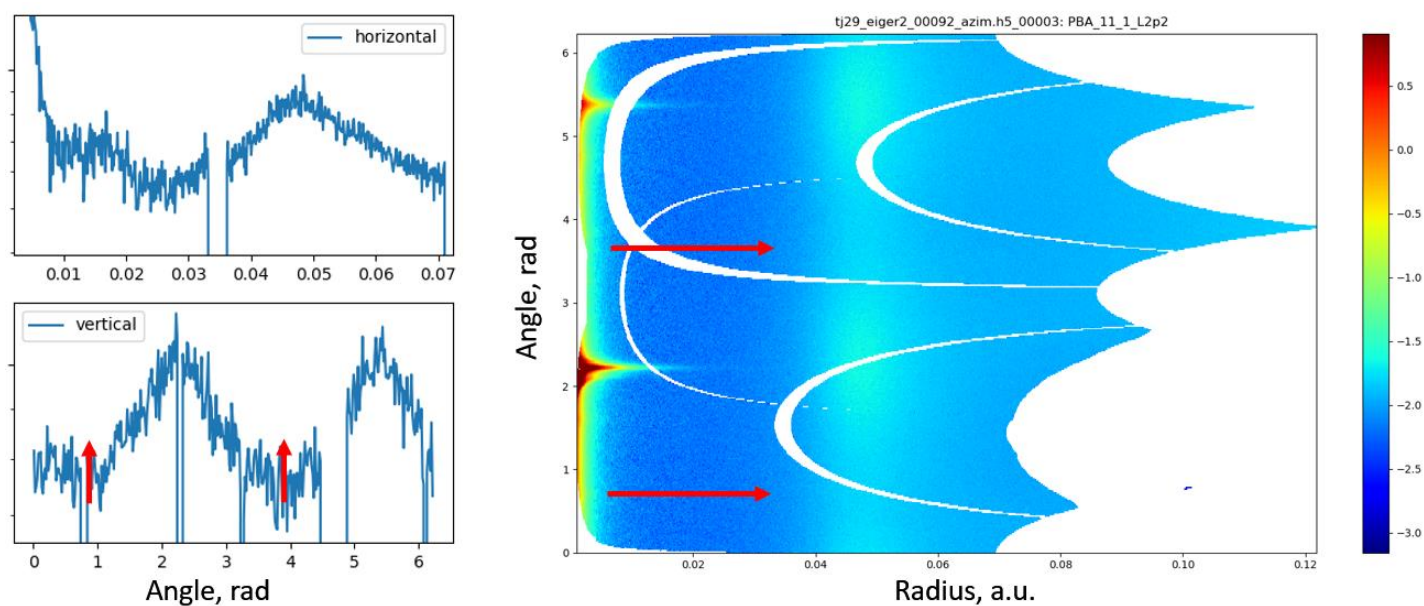


Figure 2. AZIM plot of stretched PBA_{11_1} bottlebrush sample. Red arrows indicate direction of stretching.

References

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- [3] Keith et al, *Macromolecules* 2020, 53, 21, 9306–931
- [4] Clair et al, *ACS Macro Lett.* 2019, 8, 5, 530–534
- [5] Sarapas et al, *PNAS* March 10, 2020 117 (10) 5168-5175