Experimental report

15/09/2018

Proposal: **9-11-1813**

Council: 10/2016

Title: Shear-induced disentanglement transition in polymer solutions and melts

Research area: Physics

This proposal is a resubmission of 9-11-1779

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Samples: PDMS

Instrument	Requested days	Allocated days	From	To
D11	2	2	26/06/2018	28/06/2018
IN15	7	6	28/06/2018	04/07/2018

Abstract:

Polymer solutions containing sufficiently long chains are known to show a strain plateau once a certain ammount of stress is reached. This plateau is discussed to have its origin in the formation of shear bands or in a disentanglement transition. With small angle neutron scattering it has been shown that long polymer chains under shear may stretch along the flow direction. However, rheological experiments probe dynamics properties in a sample and it is expected that transitions found in rheological measurements manifest themselves in the local dynamics of the polymer chains as well. Having shown in a proof-of-principal measurement that it is possible to monitor the dynamics of molecules and molecular segments in flowing media, we propose a full neutron spin echo experiment combined with a small angle neutron scattering study to shed light upon the origins of shear thinning in polymeric fluids.

Experimental report 9-11-1813

Proposers: Tomoko Akiyama[1], Maciej Kawecki[1], Airidas Korolkovas[2], Peter Falus[2], Philipp Gutfreund[2], Max Wolff[1]

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The aim of the current experiments was to look for evidence of polymer chain disentanglement under applied shear. The entanglements in polymers are known to result in the reptative motion of the chains. These topological constrains are a purely dynamical effect and can only be resolved by high resolution neutron spectroscopy, neutron spin echo spectroscopy. Recently, we had shown that such experiments are in principle possible. We constructed a non-magnetic shear device and showed that shear experiments on IN15 are possible without significant compromises on the instrument resolution. Moreover, we have shown that Doppler scattering can be avoided or at least accounted for [1]. The goal of this experiment was to built on this past experience and combine structural information obtained on D11 with measurements on IN15.

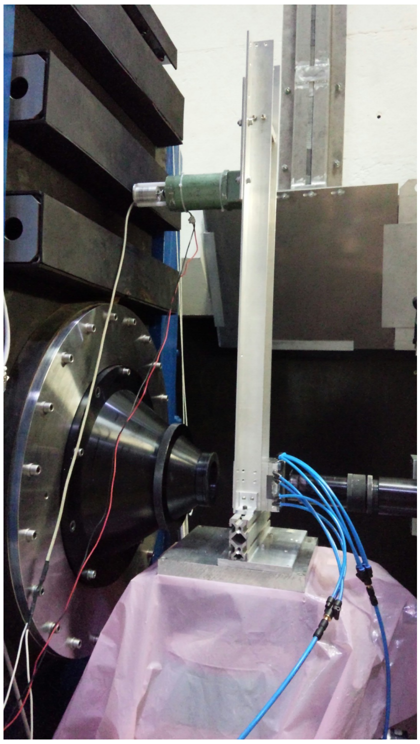

Figure 1 Cone-plate shear device mounted on D11. The beam enters from the right hand side. The detector tank is visible on the left.

To reach high enough shear rates to overcome the longest relaxation time of our polymer, we further improved the cone-plate shear device used previously by reducing the cone angle and replacing the dynamical sealing. As sample we chose a 92 kg/mol PDMS melt (37.4 % fully protonated chains, the rest deuterated, dissolved by toluene). The PDMS solution has the advantage that the viscosity can be tuned to some extend. To fill up the shear cell, 5 ml of PDMS were prepared and put into the shear cell which was closed with a crystal glass window. It turned out that the filling directly on the cone was challenging and we will further improve the procedure for upcoming experiments. A motor is connected to the shear cell by a non-magnetic belt at upper part (Fig. 1), and allows an adjustable rotation speed. During this beamtime data was collected at rotation speeds of the cone of 0, 45, 75 and 105 rpm. The temperature was controlled by a chiller connected via tubes to the shear cell window cover. All parts of the shear cell except the front window are made of aluminum since non-magnetic materials are required for NSE measurements.

During the NSE measurements, a graphoil spiral was used for the resolution measurements. The resulting data is shown in Fig. 2 (left panel). Clearly, for this shear rate and at larger Q values Doppler scattering affects the resolution of the instrument. This can be partially mitigated by analyzing only small parts of the detector, which will in turn result in a worse statistics. As mentioned above data was taken for four shear rates, 0, 45, 75 and 105 rpm of the cone, on both instrument D11 and IN15.

Figure 2 (right panel) shows exemplary data of the intermediate scattering function measured for 75 rpm on IN15. The data is corrected for the instrumental resolution. The beam wavelength was 10 Å. At larger Fourier times a clear difference between the data taken at rest and those under shear is visible. This might be related to the fact that the resolution effects are not properly taken into account. However, in that case the largest effect would be expected for the largest Q values, which is not in line with Fig. 2 (right panel). It might be that there is an indication of disentanglement but the final proof will only be possible after carefully reducing the data and further analysis, which is on the way.

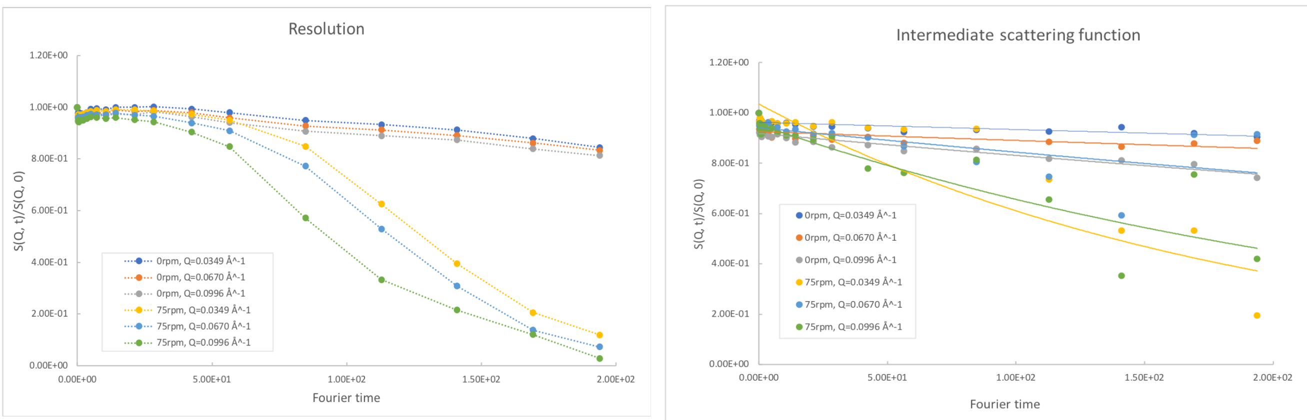

Figure 2: Resolution measured with a graphite foil mounted in the shear device on IN15 (left panel). The right panel depicts the intermediate scattering function at rest and for a shear rate of 75 rpm.

References:

[1] M. Kawecki, et al., J. Phys: Conf. series (JPCS) **746**, 012014 (2016).

[2] A. Korolkovas. et al., J. Chem. Phys. **149**, 074901 (2018).