# Experimental report

05/02/2019

**Proposal:** 9-11-1862 **Council:** 4/2018

**Title:** Entanglement Dynamics in Ultra-High Molecular Weight Polyethylene andits Composites with Graphene Oxide

**Research area:** Soft condensed matter

**This proposal is a new proposal**


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**Local contacts:** Ingo HOFFMANN


**Samples:** reduced Graphene Oxide
Ultra-High Molecular weight Polyethylene
Deuterated Ultra-High Molecular weight Polyethylene

| Instrument | Requested days | Allocated days | From | To |
|---|---|---|---|---|
| D33 | 1 | 0 | | |
| D22 | 1 | 0 | | |
| D11 | 1 | 0 | | |
| IN15 Std+Small echo | 7 | 3 | 05/10/2018 | 08/10/2018 |


## Abstract:

Ultra-High Molecular Weight Polyethylene (UHMWPE) is an engineering polymer which is used in a wide range of applications such as medical implants or bullet-proof vests. However, when melted, its extremely long chains lead to a high density of entanglements that complicates processing. Our group has developed a controlled synthesis method that reduces remarkably the number of entanglements in the amorphous phase. In this experiment, we will characterize for the first time disentangled UHMWPE using neutron spin echo (NSE) spectroscopy and its composites with reduced graphene oxide (rGON). Backed by rheology measurements, we expect to see a slow down of the entanglement dynamics in the presence of rGON, which affects strongly the chain confinement. This experiment will be the link between the rheological response and the microscopic chain dynamics picture provided by NSE, and a significant advance in the understanding of the behaviour of entangled polymer melts in the presence of nanofillers.

**EXPERIMENTAL REPORT**

**Proposal 9-11-1862**

# Entanglement Dynamics in Ultra-High Molecular Weight Polyethylene and its Composites with Graphene Oxide

## Experiment objectives

The objective of our experiment is to measure the single chain dynamic structure factor in melts of disentangled UHMWPE and its composites with rGON using NSE spectroscopy. We aim to identify the relaxation mechanisms that govern chain dynamics in the melt and obtain valuable parameters such as the reptation tube diameter, as well as assessing the influence of the presence of rGON in the entanglement formation.

## Experiment report

The Panel allocated 3 days (out of the 7 requested) of beamtime for carrying out preliminary work “to find the appropriate time window on pure components”. Therefore, during our experiment we used pristine UHMWPE samples of 2 molecular weights (2.1 Mg/mol and 5.4 Mg/mol) with 10 wt% of hydrogenated chains. We explored 3 different temperatures (435 K, 473 K, and 509 K) to be able to choose the right time and q ranges that allow us to visualize the reptation plateau. As shown in Figure 1, we found out that for a q range between 0.05 $Å^{-1}$ and 0.13 $Å^{-1}$ the reptation plateau is reached within times much smaller than the detection range (which goes all the way to 400 ns, a zoomed in spectra is shown here for clarity). As temperature increases, the initial Rouse decay is faster, facilitating the analysis of the reptation plateau. The dynamic structure factor is not normalized to one because a measurement of the 100% sample was not performed to subtract the background. We are currently working on the fitting of these curves using the DeGennes expression to obtain a value of the tube diameter to compare with other references in the literature.

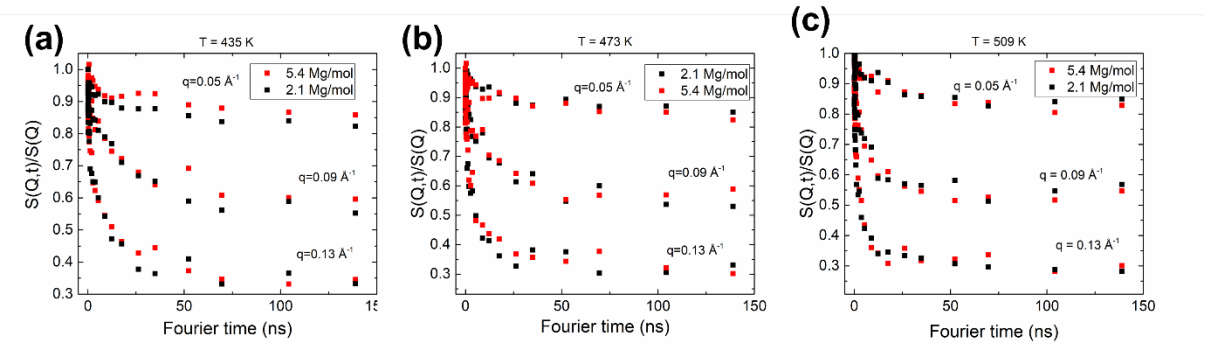


Figure 1. Single chain dynamic structure factor of UHMWPE for two different molecular weights (2.1 Mg/mol and 2.1 Mg/mol) at (a) T = 435 K, (b) T = 473 K, and (c) T = 509 K.

Therefore, and as requested by the panel, in these preliminary experiments we found out the appropriate temperature (T = 509 K), q range (0.05 $Å^{-1}$ – 0.13 $Å^{-1}$), and time range (0 ns – 200 ns).

**Likely outcome (s)**

This experiment will lead to the following outcomes:

1) An EASY proposal to measure a fully deuterated sample and normalize our dynamic structure factors to one.
2) A follow-up proposal at ILL for beamtime in February 2019 to perform spin echo on the rGON/UHMWPE systems initially proposed as well as SANS to determine the radius of gyration of our samples
3) The experiments are likely to be included in the PhD thesis of one of the experimenters (S. Drakopoulos)
4) If more beamtime is awarded and the composite systems are measured, a journal publication will be prepared.