KATHOLIEKE UNIVERSITEIT LEUVEN

# Graphene Sheets: Characterization in suspension and at interface

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## Abstract

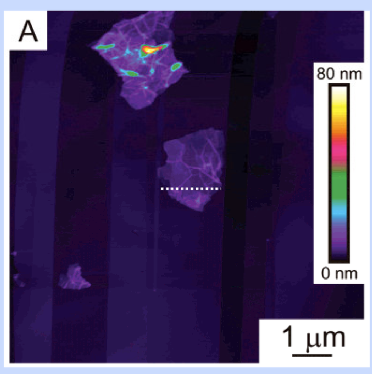
Graphene has been called 'a rising star of material science and condensed matter physics'[1]. The great attention that it has gained in the field of physics is due to its 2-dimensional nature, which allows to design systems with highly dense structures and yet high surface area (almost close to theoretical limit), good electronic properties and high mechanical strength (greater than carbon nanotubes), etc. Most of the past research has been focused on the mechanical properties of individual sheets and of composite materials. Yet, the 2-dimensional nature of graphene sheets and its effects on the behavior in complex systems has not been addressed explicitly.

In this work we investigate the behavior of functionalized graphene sheets (FGS) at the interface as a mean of exploiting the bi-dimensional nature of the sheets in a bi-dimensional assembly: we show how graphene is a strong surface-active material and how it can be used as a low cost and high yield emulsion stabilizer. Moreover we present the rheology of water-in-oil emulsions in both oscillatory and steady shear measurements. The reason of the strong compressibilities and high elasticities observed for FGS-laden interfaces is searched in the compressibility and elasticity of the single graphene sheet itself, through dichroism measurements of suspended FGS under oscillatory shear flow.

[1] A. K. Geim and K. S. Novoselov, Nature Materials 6, 183 - 191 (2007)

## *THE SAMPLE – dimensions and behavior at the interface*

***FUNCTIONALIZED GRAPHENE SHEETS (FGS)***



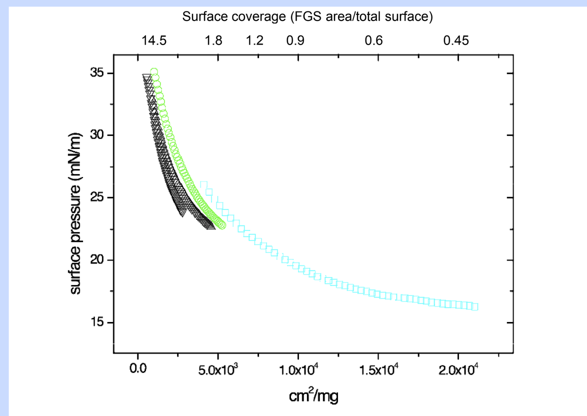

Mean thickness *1.7 nm*

Surface area

*600-900 m²/g (BETNitrogen)*

***1850 m²/g*** *(MB dye Ethanol Suspension)*

MJ McAllister, *Chem. Mater.* ***2007, 19, 4396-4404***



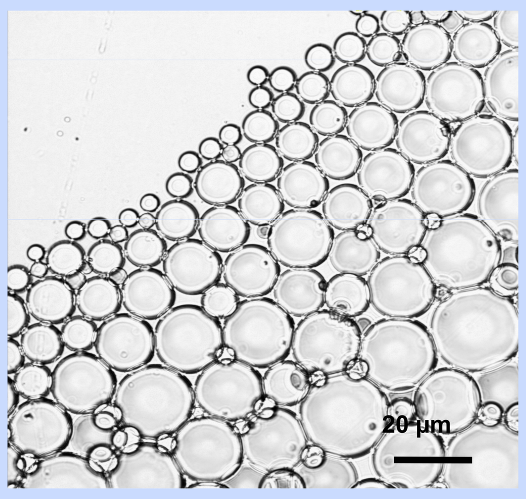

**FGS at water-air interface**

Pressure-Area isotherms at different surface coverage

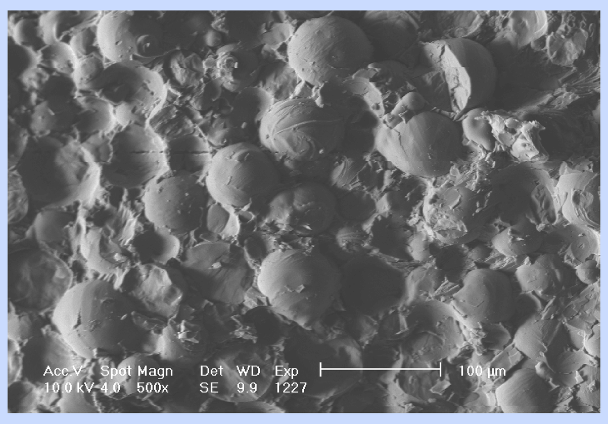
Surface pressure $\pi = \gamma_{clean} - \gamma$

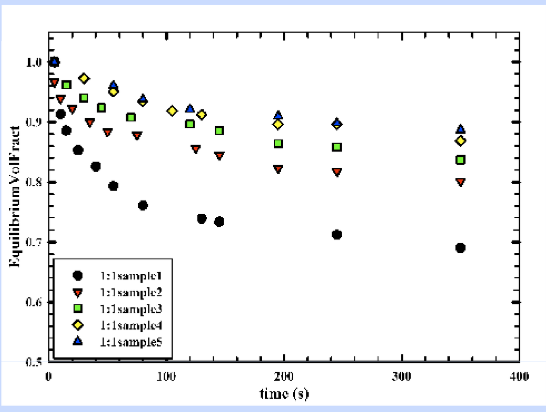
## *THE EMULSION*

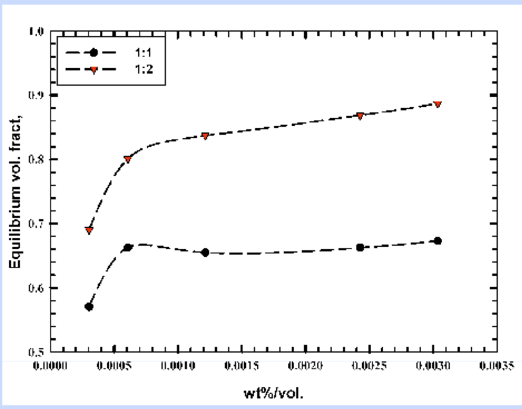
### *Optimization of FGS concentration and phase ratio*

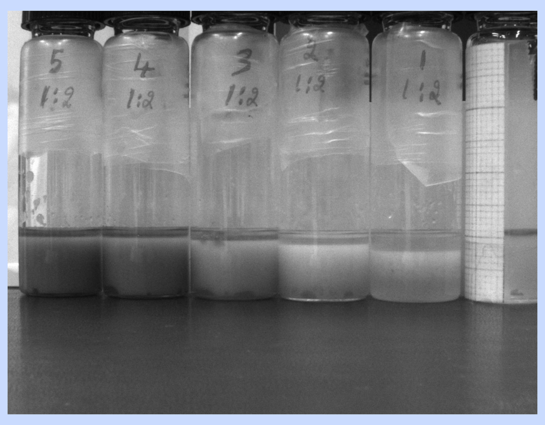


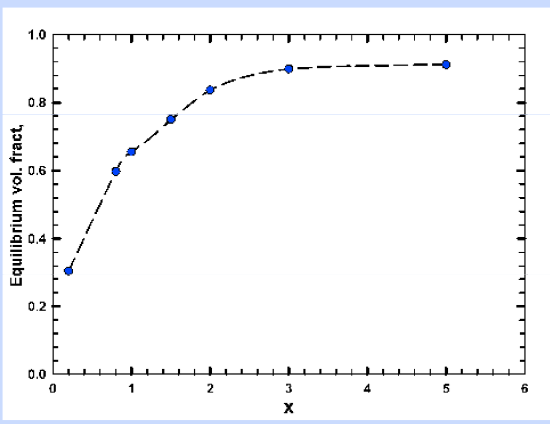

**WATER IN DECANE EMULSION**









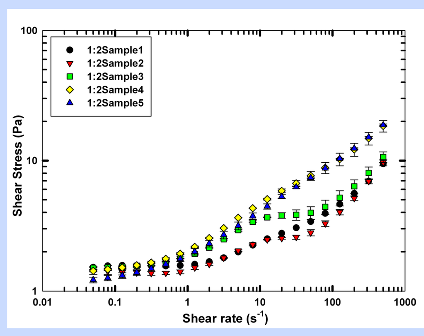


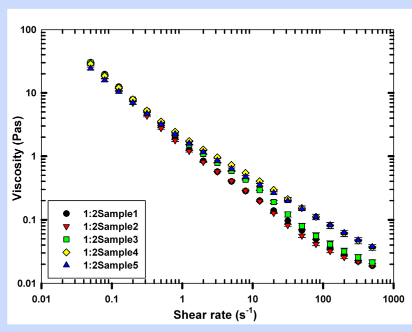

All emulsions show an excellent stability over time (> 6 months). The efficiency in emulsion formation is high even at very low FGS concentrations **(E-3 wt%/vol)** . The emulsification power increases with FGS content and water fraction up to a constant volume fraction of **0.85** (emulsion:total volume).

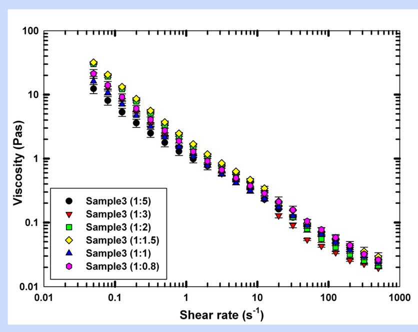
Optimal conditions are decane:water 1:2 vol and FGS 1.2 E-3 wt% /vol

## *RHEOLOGY*

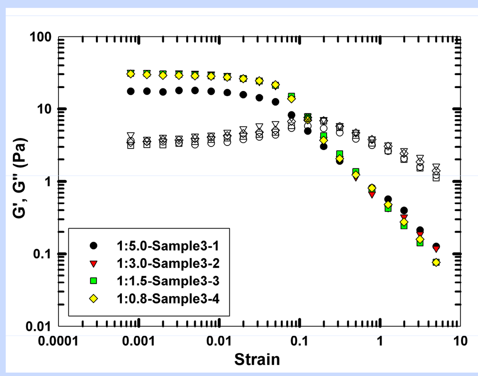
**Steady shear**

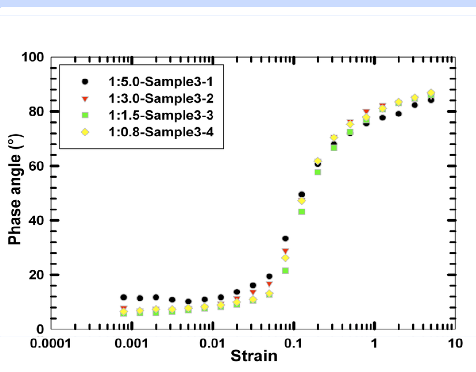


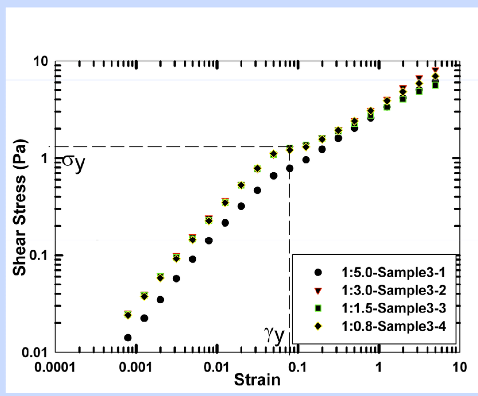


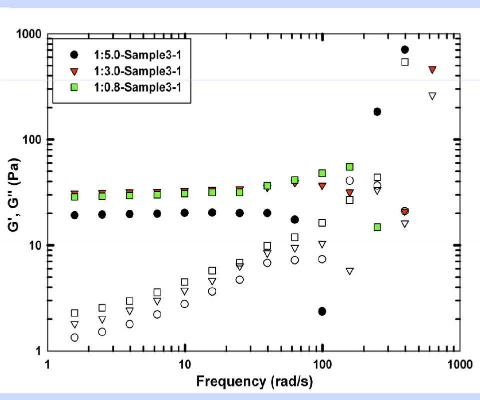


**Oscillatory measurements**







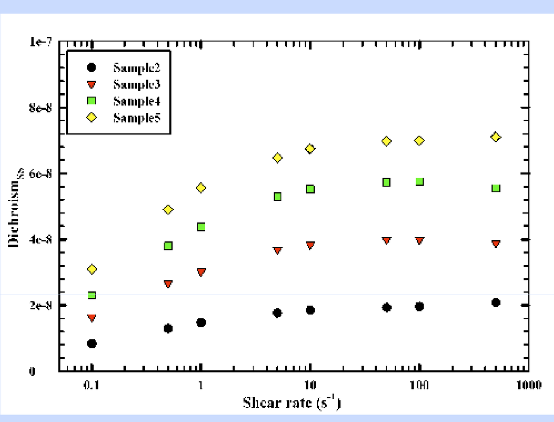


All emulsions show a strong shear thinning behavior. For emulsions with low FGS concentrations a stress plateau appears at a shear rate of approximately 10 $s^{-1}$. The position is not affected by the oil-water volume ratio.

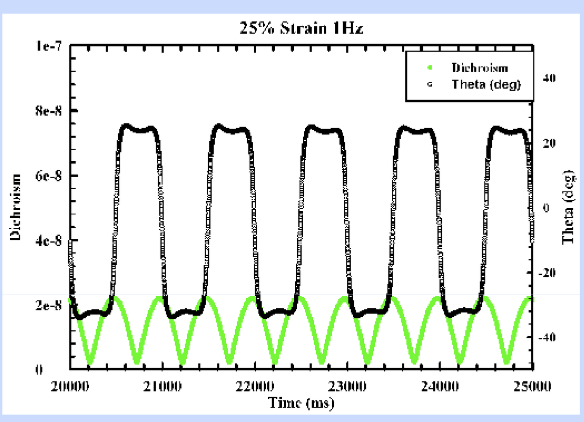
Oscillatory measurements show a strong elastic behavior in a wide range of strains and frequencies, up to a strain of 0.1 at which the emulsion begins to yield. The yield stress value of ~1 Pa is in excellent agreement with the plateau value of the steady shear stress.

## *RHEO-OPTICS – FGS in oil*

***Steady shear***

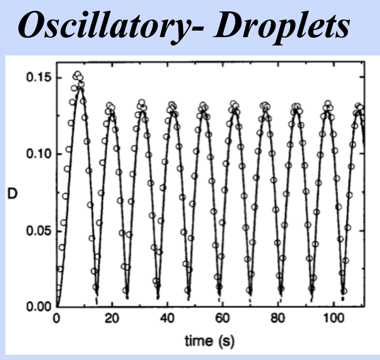


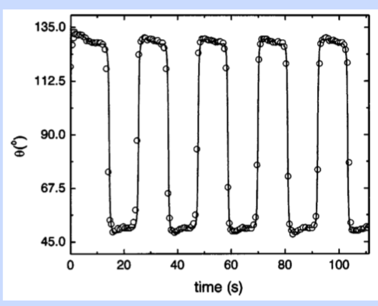
***Oscillatory***




***Stretching, crumpling and tumbling***

***Oscillatory- Droplets***





Cavallo *et al. -RheolActa.* **2003**, 42, 1

## *Conclusions*

**FGS are a strong surface-active material:**

- reduces surface tension
- adsorption at the interface is stable at different surface coverage and compression values.

**FGS stabilized water-in-oil emulsions:**

- High emulsification efficiency, up to 0.85 volume fraction even at low FGS concentrations (E-3 wt%/vol)
- Excellent stability over time, up to months
- Strong elastic behavior: rheology of a Bingham fluid with yield strain of 0.1 and yield stress of 1 Pa
- Dichroism of FGS in suspension: results suggest that graphene sheets stretch and crumble under oscillatory shear flow. The intrinsic elasticity of the single sheets could explain the high elastic behavior found for emulsions.

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