

## **Experimental Section**

### **Materials**

Different viscosity grades of Hydroxy propyl methyl cellulose (AnyAddy, 15, 50, 100 and 4000cps, Samsung Fine Chemicals) were received as generous gift samples from Harke FoodTech, Germany. Sunflower oil was received as a gift sample from Vandemoortele Lipids N.V., Belgium. Nile red was purchased from Sigma Aldrich Inc., USA.

### **Preparation of organogels**

The organogel preparation included following steps:

- 1) Aqueous foam preparation and drying: Accurately weighed samples of HPMC were dissolved in water to achieve 2 %wt polymer solutions. These solutions of different viscosity grade HPMC were then aerated with the use of high dispersing unit (Ultraturrax<sup>®</sup>). The aqueous foam was then frozen at -23 °C overnight before subjecting them to freeze drying using VaCo5 lyophilizer (ZirBus Technology, Germany).
- 2) Organogel formation: Accurately weighed quantities of sunflower oil was sorbed onto the dried cryogels and left overnight. The oil-sorbed cryogels were sheared at 11,000 rpm using Ultraturrax<sup>®</sup> to get organogels. Organogels with varying amount of polymer weight (1-5 %wt) were prepared for comparison.

### **Microstructure studies**

The microstructure of aqueous foam, cryogel and organogel was studied using optical & polarized light, confocal and electron microscopy.

Optical and polarized light microscopy was used to image both the aqueous foam and the organogel on a Leitz diaphan microscope, Lietz Wetzlar Germany). For foam, a diluted sample was used for better viewing of the air bubbles.

For confocal microscopy, Nile red was first dissolved in sunflower oil and this oil was then used for preparing the organogel samples. Samples were imaged using a Nikon A1R confocal microscope (Nikon Instruments Inc., USA). Excitation was performed by means of a 488nm Ar laser and fluorescence was detected through a 525/50 bandpass filter. Images were acquired and processed with Nikon NIS Elements

software.

For cryo-scanning electron microscopy, cryogel sample was placed on a rivet-type holder, plunge-frozen in liquid nitrogen and transferred to the cryo-chamber of an Oxford Cryo-Trans 1500 cold stage attachment (Oxford Instruments, Tubney Woods, Oxon, UK). Sample was subsequently cryoplaned in a cryo-ultramicrotome (Leica) and shortly sublimated (freeze-etched) at  $-90^{\circ}\text{C}$ . The fractured sample was sputter-coated with Au/Pd, and examined in a JEOL JSM6340F SEM (JEOL Ltd, Tokyo, Japan) operated at 3kV.

### **Oil uptake study**

For studying the oil uptake, weighed sample of cryogel ( $X$ ) was placed on a sieve attached to a holder at a height of around 5 cm on top of a weighing balance. Weighed amount of oil sample was dropped on the cryogel using a pipette until the sample was saturated with oil and could no longer absorb any oil. The oil-sorbed sample was left on the sieve for an hour and any oil spilled on the balance was weighed and subtracted from the initial amount of oil added to get  $O_T$ . The oil swelling capacity as calculated by following formula:-

$$\text{Oil swelling capacity} = [O_T/X] * 100$$

For better visualization, Nile red was dissolved in oil and this oil was used to demonstrate rapid uptake of oil by cryogel. Precisely, 0.2 g sample of cryogel was placed in petridish containing 25 g oil (containing Nile red) and pictures were clicked every 60 seconds to follow the oil uptake.

### **Rheological measurements**

The rheological measurements were carried out on an advanced rheometer AR 2000ex (TA Instruments, USA) using a parallel plate geometry of diameter 40mm. The viscoelastic parameters such as storage ( $G'$ ) and loss ( $G''$ ) moduli were measured as a function of time, temperature and frequency. For LVR studies, the samples were subjected to a varying stress from 0 to 250 Pa at a constant frequency of 0.25 Hz at  $20^{\circ}\text{C}$ . Temperature and frequency sweeps were carried out at a constant stress of 10 Pa and varying temperatures (from 20 to  $100^{\circ}\text{C}$ ) and frequencies (from 0 to 100 Hz) respectively. Temperature-time related behaviour was studied by measuring the viscoelastic modulus over 20 minutes at increasing temperatures from 20 to  $80^{\circ}\text{C}$ .

The flow behaviour of the samples was also studied by measuring the change in viscosity at a shear rate of  $0.1 \text{ s}^{-1}$  over time for organogels prepared at different polymer weights (1-5 %wt). The thixotropic and shear-thinning behaviour was studied by measuring viscosity over time at 2 alternative shear rates ( $0.1$  and  $10 \text{ s}^{-1}$ ).

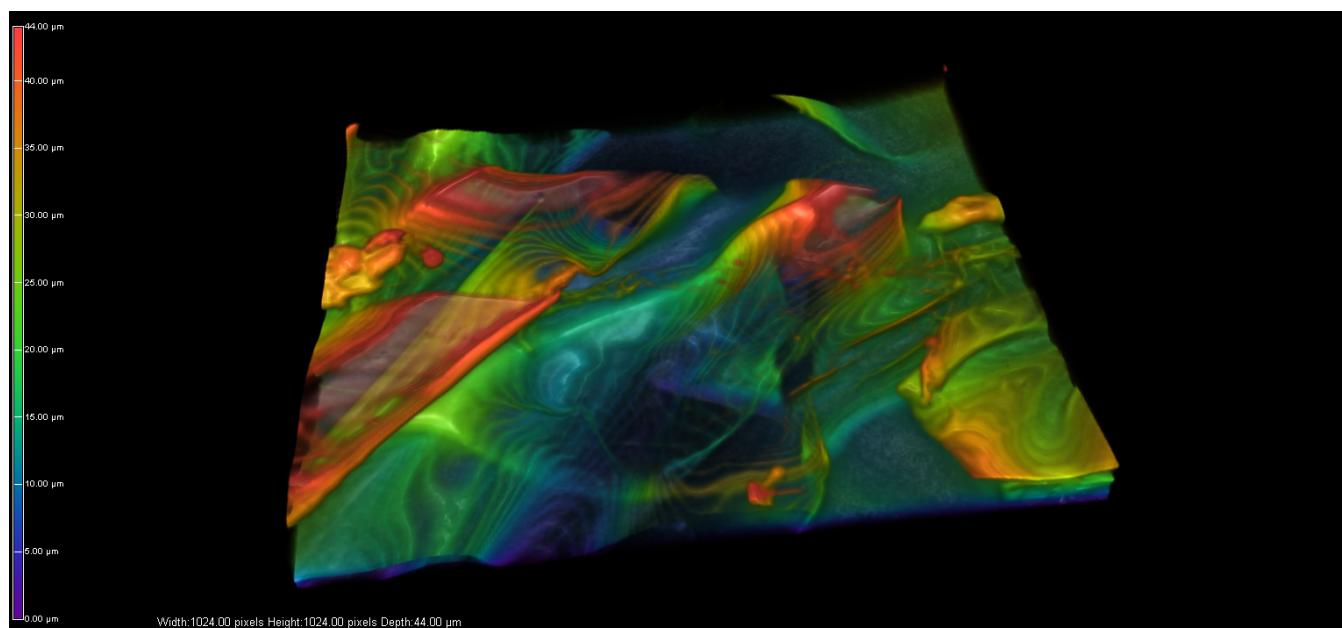
## Tables & Figures

**Table. S1.** Foaming properties of different viscosity grades of HPMC. Initial volume of HPMC solution, initial foam volume (at time  $t_0$ ) and final volume after 30 minutes (at time  $t_{30}$ ) were measured and used for calculation of % overrun and % foam stability.

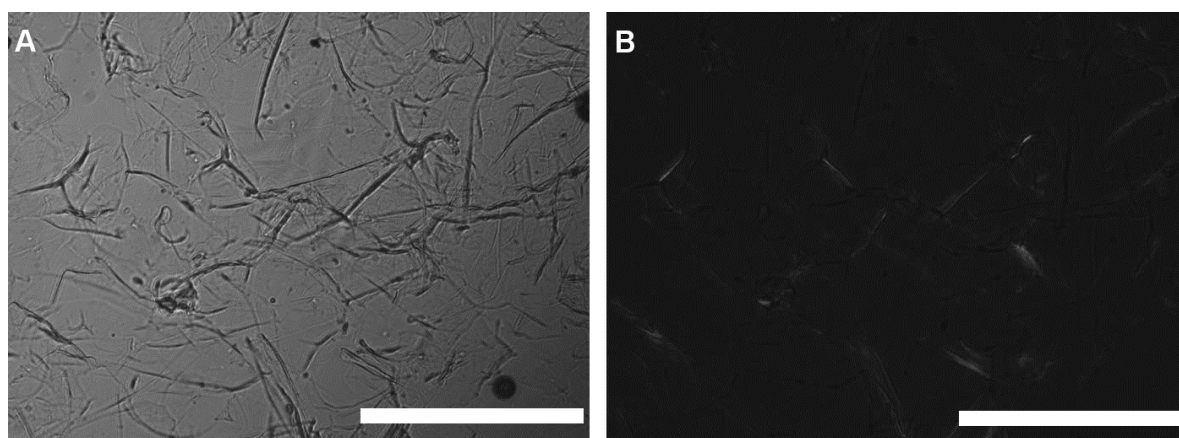
Viscosity grade (cPs)	15	50	100	4000
Initial volume (mL)	10	10	10	10
Volume foam $t_0$ (mL)	42,5	37,5	35	30,5
% overrun	325	275	250	205
Volume foam $t_{30}$ (mL)	10	11	13	27
% foam stability	0	9.6	12.5	82.9



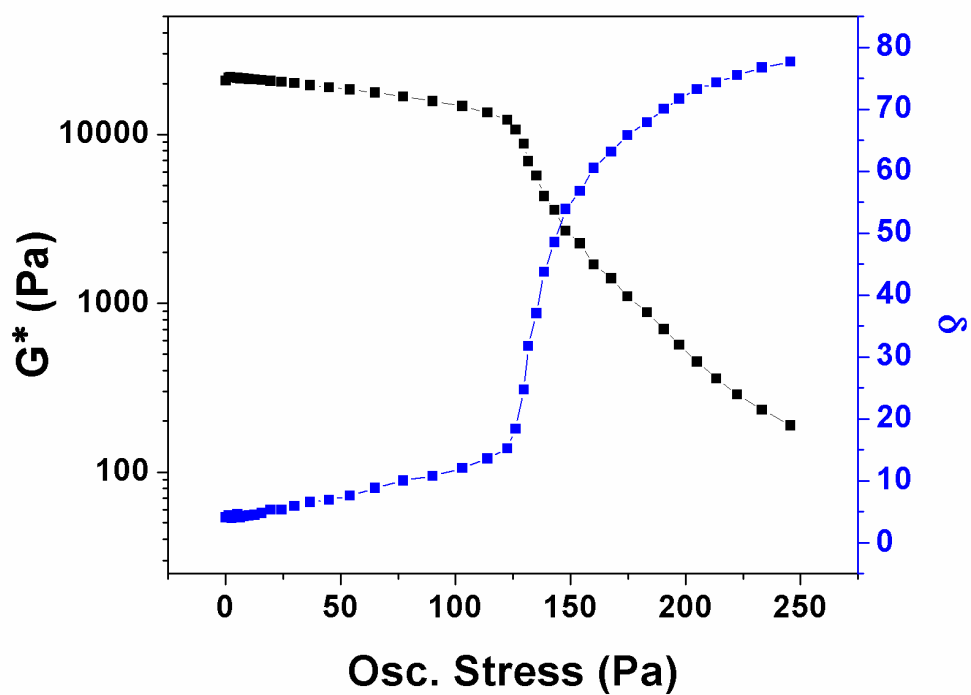
**Fig. S1.** Photograph of an organogel with 98 %wt liquid oil prepared by shearing oil-sorbed cryogels.



**Fig. S2.** Volume view generated from the stacks of confocal images. Polymer sheets are seen uniformly dispersed through the sample as indicated by their presence at several depths of the sample.



**Fig. S3.** Microscopy images of sunflower oil organogel (prepared at 2 %wt polymer weight) under optical light (a) and polarized light (b).



**Fig. S4.** Complex modulus and phase angle as a function of oscillatory stress.



**Fig. S5.** Photograph of an organogel with 98 %wt liquid oil stored at 5°C for over 3 months. No oiling out or polymer sedimentation was observed on storage.