Experimental

Materials

Fumed silica (Aerosil® 150 and 972) samples were gifted by Evonik Inudstries AG, Germany. Locust bean gum (LBG), kappa-carrageenan (Car) were generous gift samples from Cargill R&D (Vilvoorde, Belgium). Sunflower oil (>98 %wt triacylglycerols) were received as gift samples from Vandemoortele R&D Izegem, Belgium. Nile Red was purchased from Sigma Aldrich Inc. (USA). Distilled water was used for all the experiments.

Preparation of samples

The oil dispersions of silica particles were prepared by mixing accurately weighed amount of fumed silica powder to sunflower oil followed by shearing the dispersion at high speed (11,000 rpm) using a high energy dispersing unit (Ultraturrax® (IKA®-Werke GmbH & Co. KG, Germany). At 10 and 15 %wt of fumed silica, viscoelastic gels (organogels) were formed. The water gel was prepared at a total polymer concentration of 1 %wt and LBG:CAR ratio of 1:1 by dispersing weighed amount of polymer powders to distilled water under continuous stirring. To prepare bigel samples, the organogel and melted water gel in varying proportion using mild stirring. All the samples were stored at 5 °C until used for further experiments.

Characterization studies

Microstructure studies

The microstructure of fumed silica powder, organogel and bigel was studied using scanning electron microscope. For organogel sample, de-oiling was carried out using butanol to remove the surface liquid oil in order to visualize the underlying network of silica particles. The samples were than plunge-frozen in liquid nitrogen and transferred into the cryo-preparation chamber (PP3010T Cryo-SEM Preparation System, Quorum Technologies, UK) where they were freeze-fractured and subsequently sputter-coated with Pt and examined in JEOL JSM 7100F SEM (JEOL Ltd, Tokyo, Japan). For bigel samples, an additional step of sublimation was carried out after the freeze fracture to get rid of water phase. Energy dispersive x-ray spectroscopy system (Oxford Instruments, UK) was used for creating elemental map of Si in the bigel sample. For confocal microscopy, Nile Red was dissolved in oil phase which was then used to prepare organogel and consequently the bigel. 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.

Rheological measurements

The rheological measurements were carried out on an advanced rheometer AR 2000ex (TA Instruments, USA) equipped with Peltier system for temperature control. A parallel plate cross hatched geometry of diameter 40 mm was used and the geometry gap was set at 1000 μm. Amplitude sweeps (stress = 10^{-2} to 10^{4} Pa) were carried out to determine the linear viscoelastic region of organogels, water gel and bigels. The frequency sweeps (0.1 to 300 $rads^{-1}$) were done to study the viscoelastic parameters as a function of applied rate of deformation. Stress ramps (shear stress = 1 to 400 Pa) and 3-interval-time-test (3ITT) in rotation were carried out to study the flow and thixotropic behaviour of samples. For 3ITT, the samples were subjected to 3 interval of alternate low and high shear rates (0.1, 10 and 0.1 s^{-1} respectively). All these rheological measurements were carried out at 5 °C. The samples were also subjected to temperature ramps (5 to 80 °C and back at a constant rate of 1 °C/min) to study the temperature behaviour.

The flow curves of stress ramp were fitted to Hershel Bulkley model (Eq. 1) to obtain the values of flow parameters

$$\sigma = \sigma_o + K\dot{\gamma}^n \quad \text{(Eq. 1)}$$

Where σ is shear stress, K and η are consistency coefficient and flow index respectively and $\dot{\gamma}$ is the shear rate.

Figures and Tables

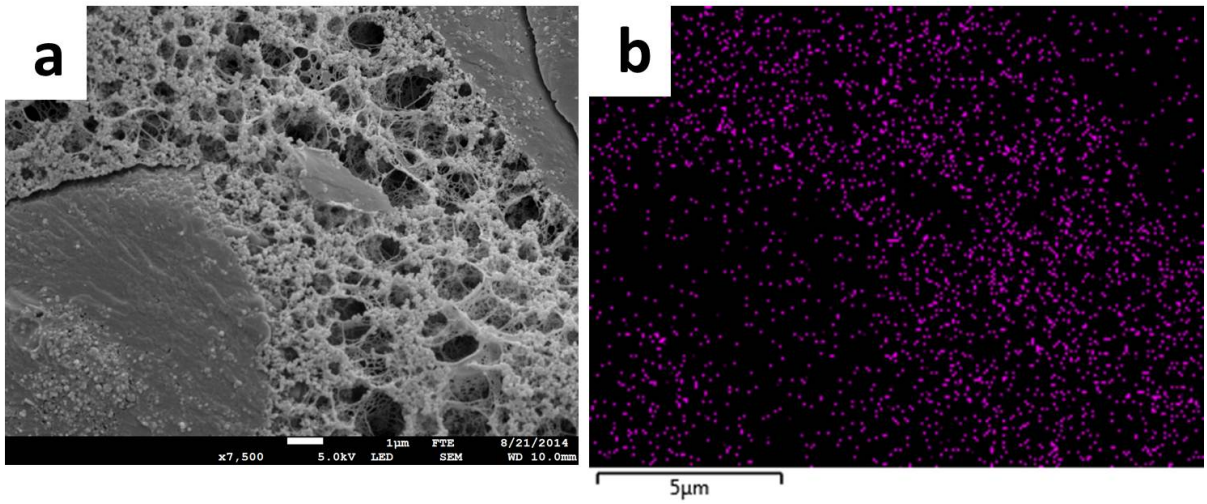

Fig S1: a & b) Cryo SEM and elemental mapping of Si respectively of bigel sample prepared using O:W 6:4.

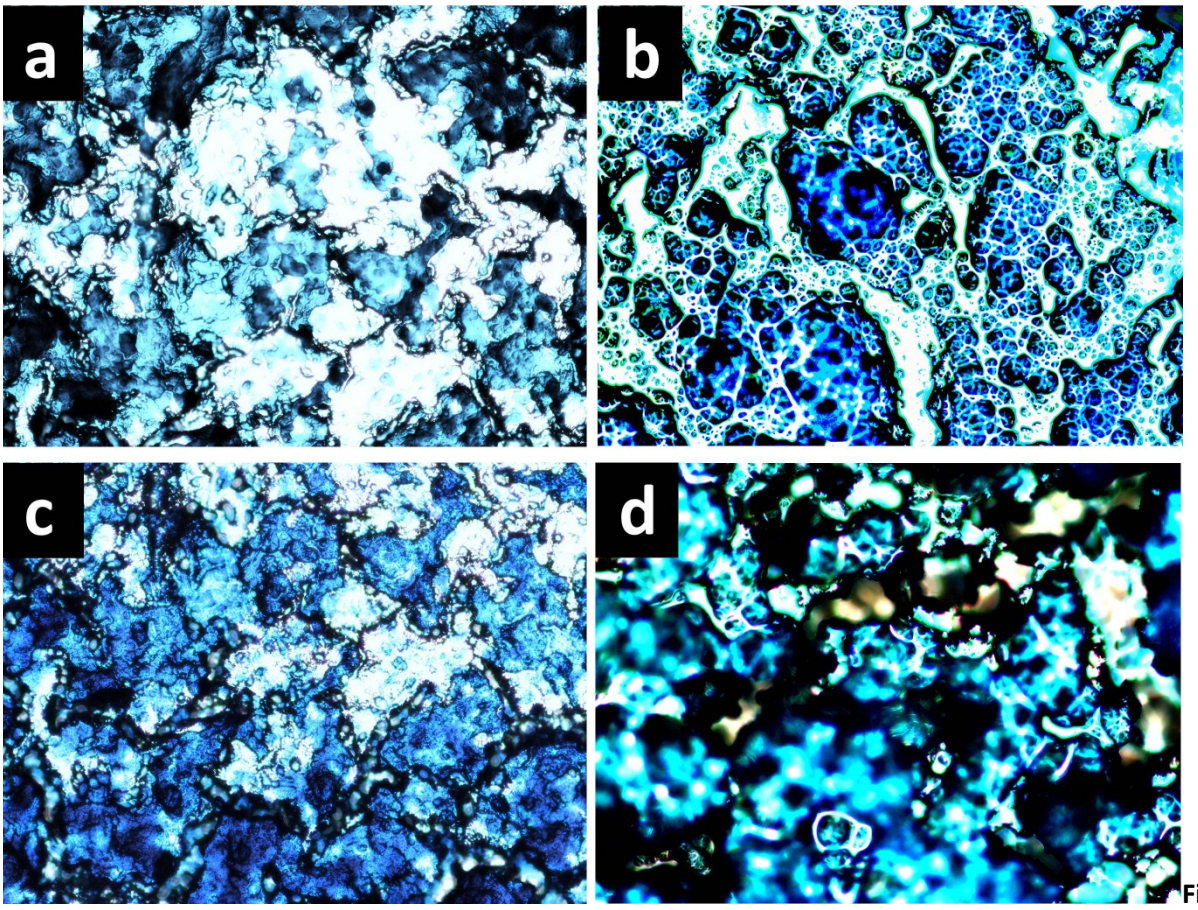

Fig S2: a) to d) Optical microscopy images of bigel samples prepared at O:W ratios of 9:1, 8:2, 7:3 and 6:4 respectively. The water phase doped with a water soluble dye (fast green FCF) was used for bigel preparation. (*Image width: 500 µm*).

Table S1: Data from 3 interval thixotropy test done on water gel, organogel and bigels, the unit for vioscosity is Pa.s.

Samples	**Viscosity at the start of interval 1** (η_1)	**Viscosity at the end of interval 1**	**Peak viscosity in interval 3** (η_3)	**structure recovery (%)** (η_3 / η_1) x **100**
Water gel	332 ± 21	359 ± 39	97 ± 1.9	29,36 ± 1.3
Organogel	3599 ± 338	12753 ± 1275	5163 ± 188	>100
Bigel (O:W, 9:1)	42035 ± 926	12375 ± 1201	5705 ± 161	13,56 ± 0.9
Bigel (O:W, 8:2)	31945 ± 816	14130 ± 1371	5298 ± 43	19.98 ± 1.9
Bigel (O:W, 7:3)	25250 ± 1640	13345 ± 1096	1766 ± 81	6,96 ± 1.2
Bigel (O:W, 6:4)	25065 ± 120	15785 ± 247	3960 ± 29	15,10 ± 2.1