Experimental Details

Materials: Rapeseed oil (> 98% triacylglycerols) and native sunflower lecithin (SL, > 90% phosphatidylcholine) were supplied by Vandemoortele Lipids N.V., Belgium. Sucrose esters (SE) sample SP10 with monoester content of 10% (fatty acid composition = stearate/palmitate) and HLB 2 was generously donated by Sisterna, Netherlands. Chemical structures of SE and SL (a & b respectively) are provided below:



R = Palmitic acid/stearic acid



R = fatty acids

Preparation of two-phase gels systems and oil foams

SE was accurately weighed and heated to 80°C followed by slow addition of pre-heated oil under continuous stirring to avoid lump formation. After complete dispersion of SE, SL was added to this mixture under stirring and the oil dispersion was then cooled to room temperature under quiescent conditions to obtain two-phase gel systems (TPgel). Similarly, oil + SE and oil + SL were also prepared for comparison.

To prepare oil foams, oil dispersions were aerated using a kitchen coffee frother for 2 minutes at high temperature (80°C) before they were allowed to cool to room temperature. The amount of air incorporation was determined by calculating the overrun (OR) using following formula:

OR [%] = V_I x 100%, where V_I and V_{II} are volumes measured before and after aeration respectively.

Gels and foams were stored at 5°C until further use as this is the storage temperature for most lipidbased food products.

Note: For selection of foaming temperature, foams of 10% wt SE samples were prepared at different temperatures to understand the effect of increasing bulk phase viscosity on air incorporation. OR calculated at different temperatures are shown in the plot below along with the viscosity values. As seen from the plot, there was a progressive drop in the overrun with increase in viscosity values of continuous phase.



Characterization of two-phase gels systems and oil foams

Microstructure studies

 $V_{II} - V_I$

Microstructure of gel and foam was studied using an optical microscope (Lieca Microsystems, Germany) under normal and polarized light. In order to calculate the dimension of air bubbles in foams, images were recorded over 4 different field of views for each sample. In each field of view the dimensions of about 130 air bubbles were measured and histograms were plotted to obtain size distribution of air bubbles of different foam samples. Temperature responsive of foams was also studied by heating the samples from 5 to 80°C at a constant rate of 5°C/min.

Foams were also visualized under cryo-SEM, the samples were placed in the slots of a stub, plunge-

frozen in liquid nitrogen, and transferred into the cryo-preparation chamber (PP3010T Cryo-SEM Preparation System, Quorum Technologies, UK). There were freeze-fractured and then sputter-coated with Pt. The examination was performed using JEOL JSM 7100F SEM (JEOL Ltd., Tokyo, Japan).

Rheological measurements

Rheological measurements of samples were carried out on an advanced rheometer AR 2000ex (TA Instruments, USA) equipped with a 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. A range of experiments including oscillatory experiments (amplitude stress and frequency sweep) and flow tests (like thixotropy, shear ramp and flow viscosity) were carried out at 5°C (except for flow viscosity test). For flow viscosity measurement, the samples were first heated from 5 to 90°C and then cooled down to 5°C at constant rate of 5°C/min and shear rate of 0.1 s⁻¹. For thixotropy test, standard protocol for a 3 interval thixotropy test was applied. Samples were subjected to alternating steps of low and high shear rate (0.1, 10 and 0.1s⁻¹) in 3 steps for 15, 5 and 10 minutes respectively. Resultant viscosity values were then plotted as a function of time to understand the structure recovery properties of samples.

Thermal analysis

The thermal parameters were studied using a Q1000 Differential scanning calorimeter (TA Instruments, USA) on samples weighing 10 mg in flat-bottomed aluminium pans. The samples were subjected to heating and cooling cycles from 5 to 80°C and back at cooling rates of 5°C/min. The thermal parameters were obtained from the heat flow curves with the help of TA Universal Analysis software.



Figure S1. PLM images of SE:SL, 10:0 and 8:2 samples diluted with rapeseed oil.



Figure S2. DSC melting curve for SE:SL, 0:10 sample heated from 5° to 80°C at 5°C/min.



Figure S3. DSC curves of sucrose esters displaying the crystallization and melting behavior.



Figure S4. On the left an image of coated air bubbles is shown, the sharpness of the image is enhanced to show the contours formed by overlapping layers. On the right, an image of the curvature of the air-oil boundary is shown that confirms the layered arrangement.