Electronic Supplementary Material (ESI) for Journal of Materials Chemistry B.

Supporting information

Monitoring the formation of a colloidal lipid gel at the nanoscale: Vesicle aggregation driven by a temperature-induced mechanism.

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Differential Scanning Calorimetry

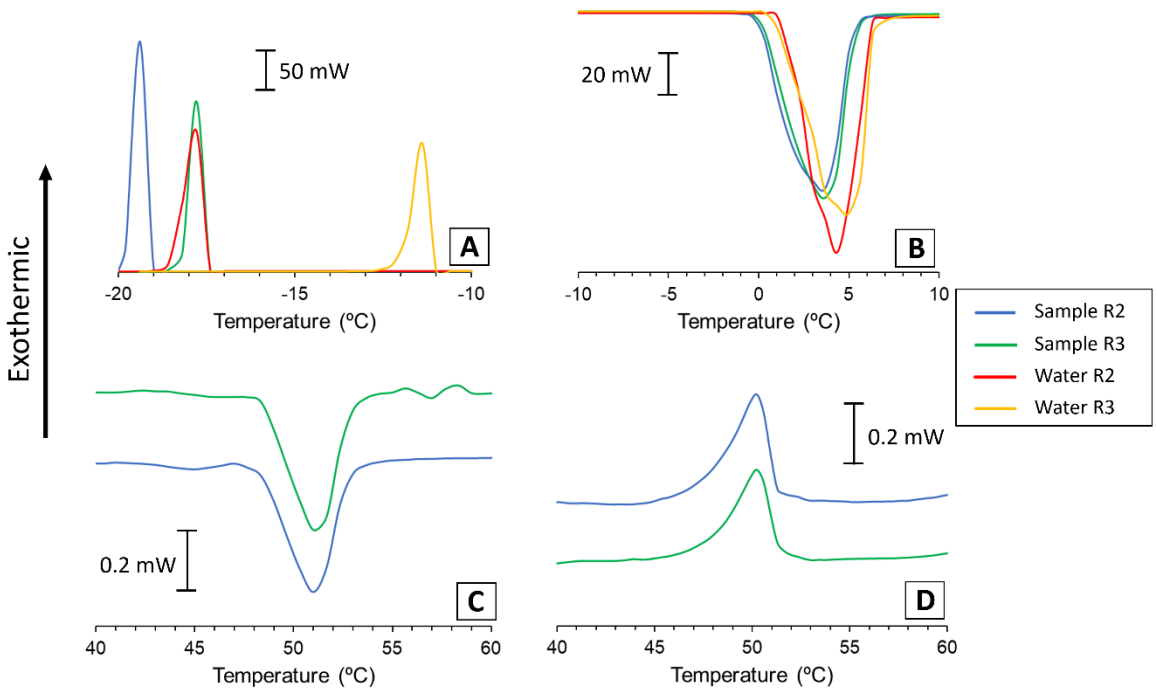

Fig. S1. DSC scan of the replicates corresponding to the lipid dispersion sample and the water control: Exothermic bands associated with freezing of step 2 (A), endothermic bands associated with melting of step 3 (B), and the endothermic band corresponding to the main phase transition of the lipid membranes from L_β to L_α in step 4 (C) and from L_α to L_β in step 5 (D).

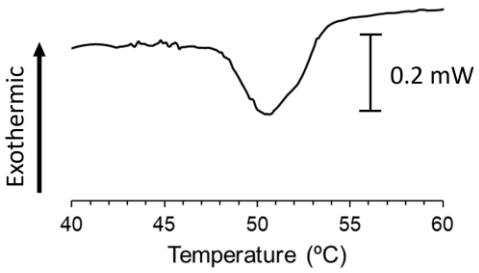

Fig. S2. DSC scan corresponding to the heating of the initial dispersion (a) from 25 to 70 ºC, showing the endothermic band of the main phase transition of the lipid membranes from L_β to L_α.

Development of the models corresponding to a bilayered cylinder and a spherical vesicle

To build this model, points were randomly distributed either within a spherical or cylindrical shell with the following electron density distribution in relation to the distance from the centre of the vesicle or the cylinder (Fig. S3).

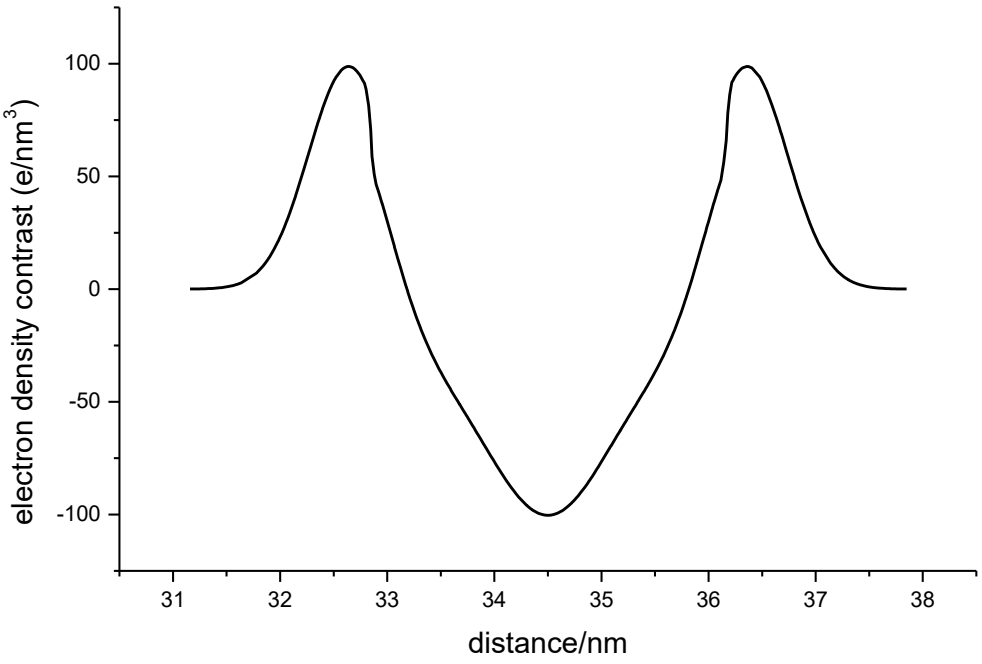

Fig. S3. Electron density profile of the bilayer forming the spherical and cylindrical shell.

The pair distribution function of the spherical vesicle, built from those randomly assigned points, is shown in Fig. S4 (black line). The corresponding function for the cylinders (red line) extends to a much larger distance (up to the length of the cylinders, 1000 nm) and only the first part appears in this figure. The complete plot is shown in Fig. S5; note that the y-axis has been enlarged to show the complete profile. Both functions are similar at very short distances (bilayer thickness range) and at longer distances (vesicle/cylinder diameter range).

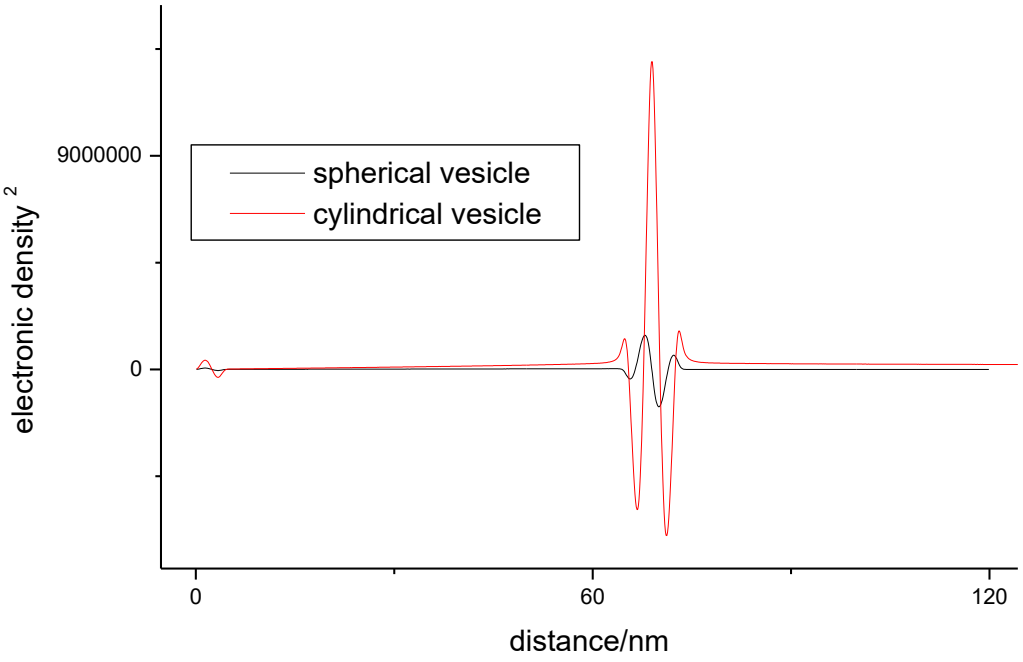

Fig. S4. Pair distribution function of the spherical and cylindrical vesicles.

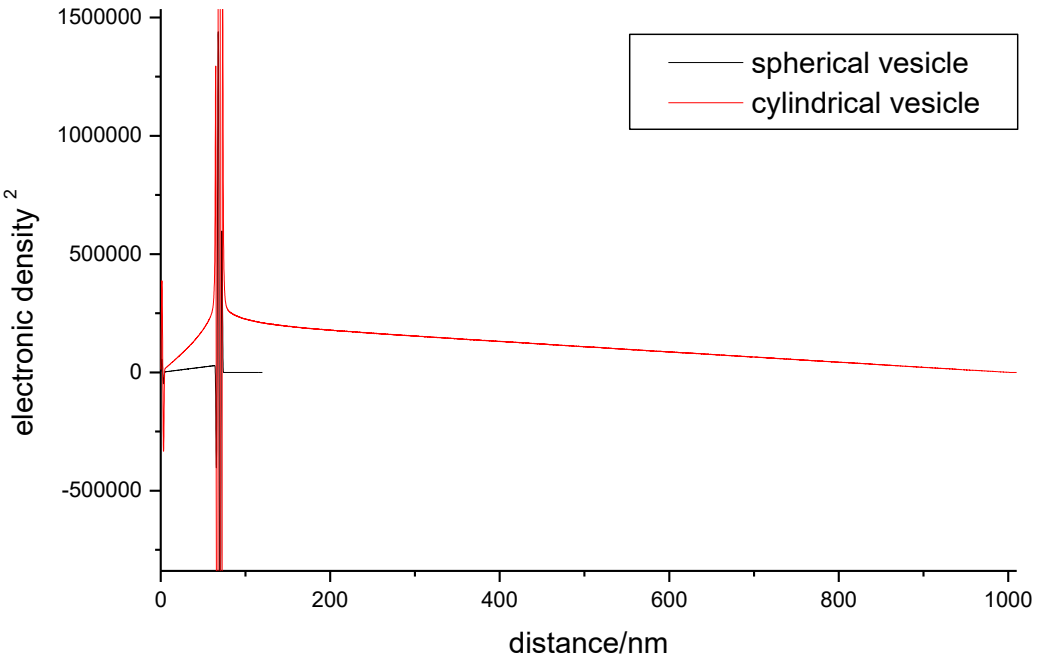

Fig. S5. Complete plot of the pair distribution function of the spherical and cylindrical vesicles.

The sinus Fourier transformation of the pair distribution function of the vesicles or cylinders generates an oscillating function whose period depends on vesicle or cylinder diameter (Fig. S6). A detailed view is presented in Fig. S7, illustrating that the different harmonics have maxima and minima that when we perform the summation of the different radii reduce the oscillations, mainly for $q>1nm^{-1}$. Note that the contribution of each radii has been weighed according to the polydispersity index determined from the fit.

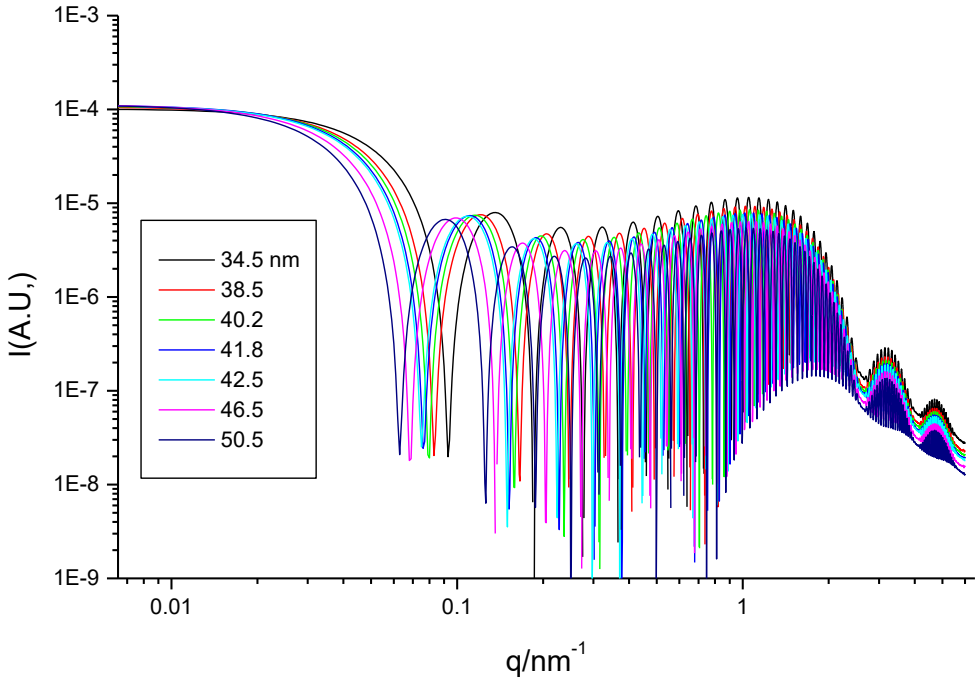

Fig. S6. Sinus Fourier transformation of the pair distribution function of the modelled vesicles to illustrate the effect of vesicle radius.

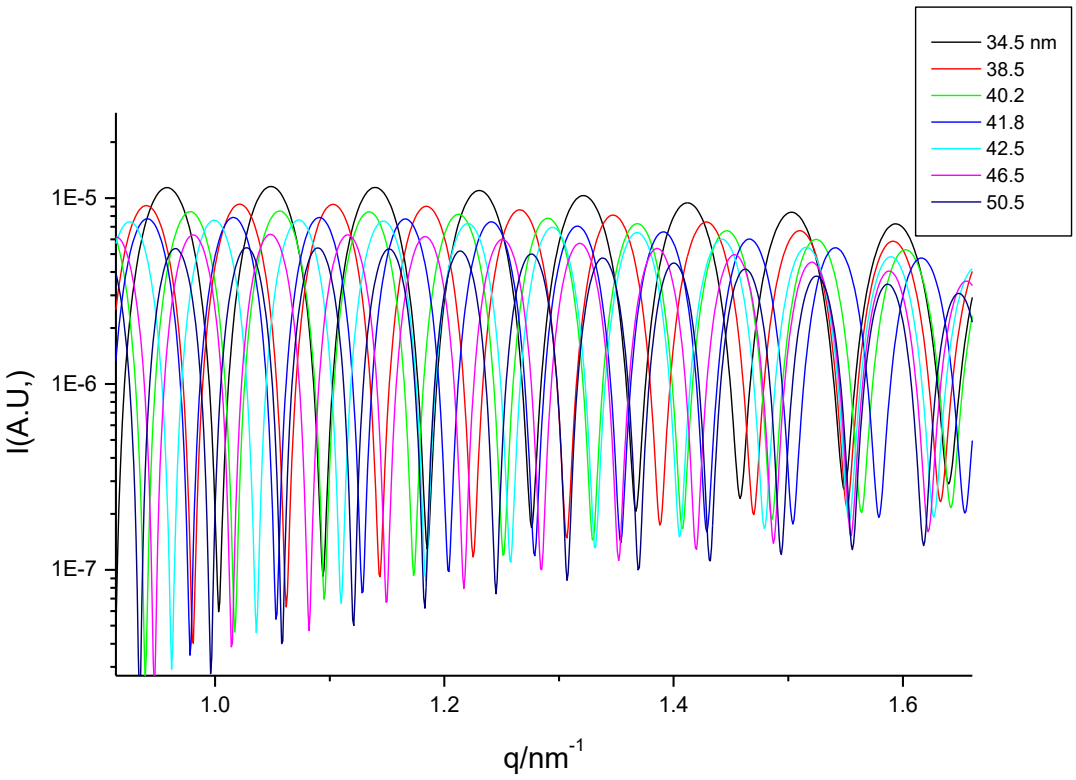

Fig. S7. Detailed view of the sinus Fourier transformation of the vesicles pair distribution function.

Fig. S8 compares a monodisperse vesicular system (1 size) with polydisperse populations that include the summation of 3 and 7 different sizes. From this plot is clear that polydispersity causes a reduction of the oscillations. Besides, this reduction is much more significant below q = 0.4 nm^{-1} than above. Using a continuum of sizes the maxima and minima would cancel except for the leftmost oscillations.

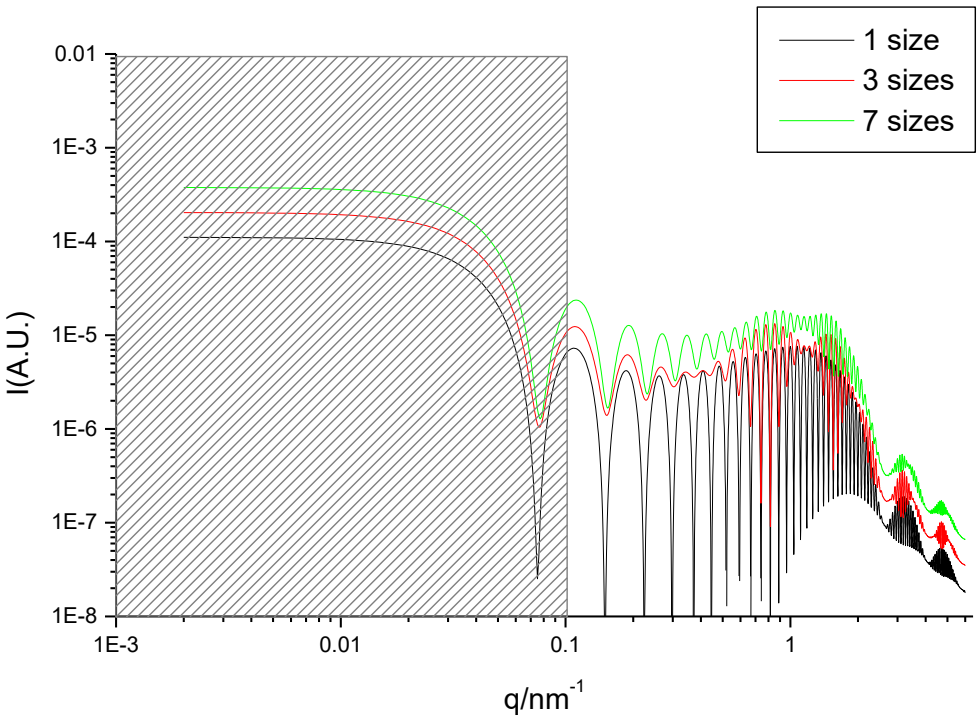

Fig. S8. Sinus Fourier transformation of the pair distribution function of the modelled vesicles and their progressive summation to illustrate the effect of system polydispersity. The shadowed area corresponds to the non-accessible very small angle scattering.

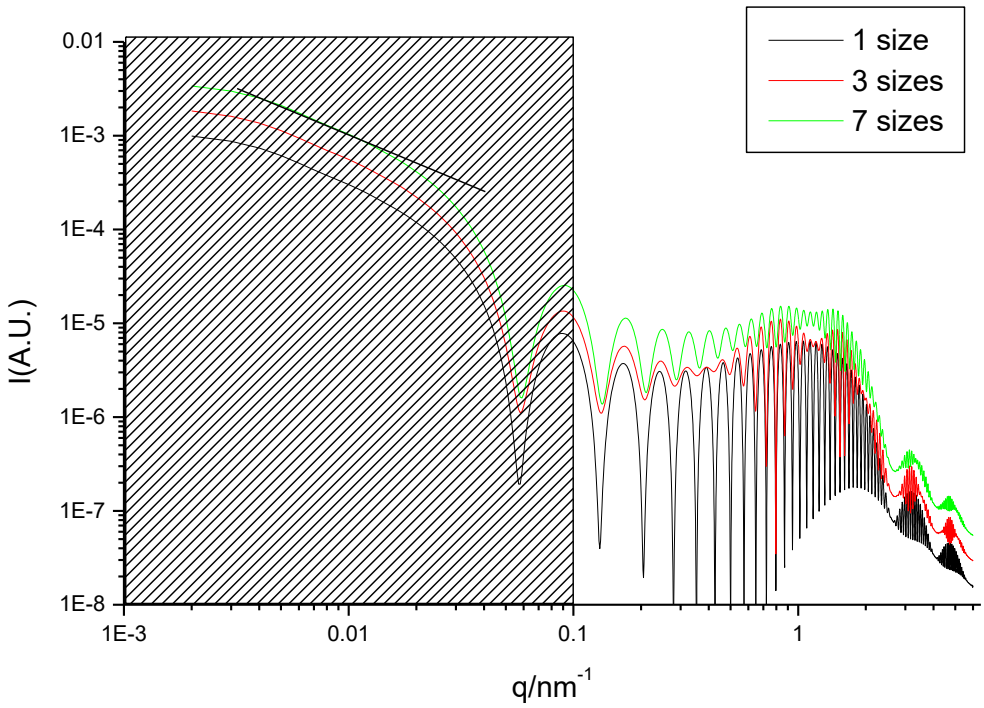

Fig. S9. Sinus Fourier transformation of the pair distribution function of the modelled cylinders and their progressive summation to illustrate the effect of system polydispersity. The shadowed area corresponds to the non-accessible very small angle scattering.

Fig. S9 shows the results for cylinders in the same fashion that Fig. S8 does for vesicles. Note that the strongest difference is in the slope at very small q, region not accessible with the present experimental setup. Maxima and minima in the curves in Fig. S8 and S9 are similar, however their positions are different. While for vesicles the position of the maxima is fitted to $q_n=A+Bn$ where A/B=0.5, for cylindrical vesicles A/B=1/4.