Design of Ultra-stretchable Physical Hydrogels cross-linked by Cubosomes: Structural Changes Revealed by SANS during In-Situ Polymerisation and Mechanical Deformation

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Estimation of D_{inter}

The mean radius values can be used to estimate the mean distance between the center of two different PT domains. This quantity is linked to the PT volume fraction and the mean radius by the equation:

$$D_{inter} = 2R_{mean}\sqrt[3]{\frac{6\Phi_{PT}}{\pi}} \quad (1)$$

Using the nominal value for Φ_{PT} of this sample and the fitting values founded for R_{mean}, we found $D_{inter} = 355$ Å, $D_{inter} = 593$ Å and $D_{inter} = 287$ Å for the pristine Cubosomes, the Cubosomes+AAm dispersion and the final hydrogel, respectively. It can be observed that at each step of the synthesis D_{inter} is smaller than the mean diameter (i.e. $2R_{mean}$) of the domains. This means that PT domains are extremely close to each other. This implies that the domains are interacting and cannot be considered as isolated.

Details on the fits of the inner morphology of Cubosomes at the different polymerisation steps and Evolution of the invariant as a function of the elongation .

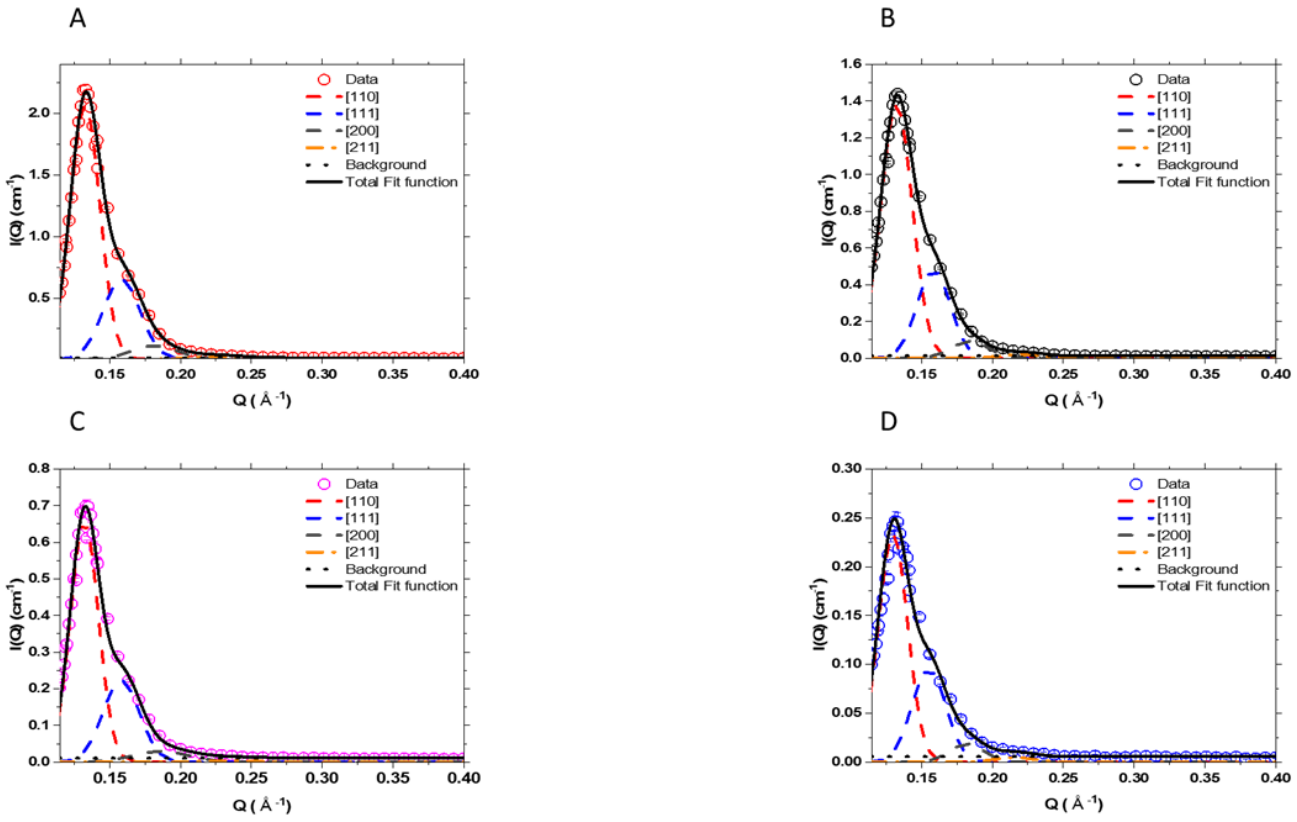

Figure S1 Diffraction pattern of the pristine Cubosomes: 10 wt% PT/1 wt% Lap (A), 10 wt% PT/0.5 wt% Lap (B), 5 wt% PT/1 wt% Lap (C), 5 wt% PT/0.5 wt% Lap (D). The points are the experimental data, the continuous lines are the fit function (see main text), the dashed lines are the Gaussian functions composing the fit function and the dotted lines are the flat background.

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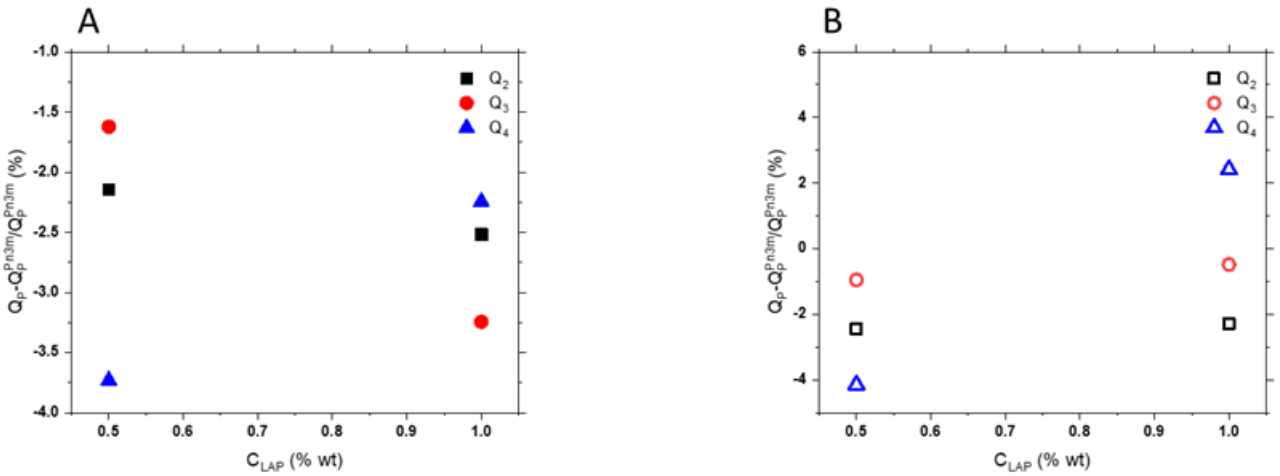

Figure S2 Percentage deviation between the peaks positions found by the data fitting and the equivalent Pn3m phase in function of the Laponite concentration for the pristine Cubosomes at 10 wt% PT (A) and at 5 wt% PT (B). The position of the Pn3m structures have been calculated using the Q_1 value as the position of the $\{h,l,k\} = \{110\}$ peak.

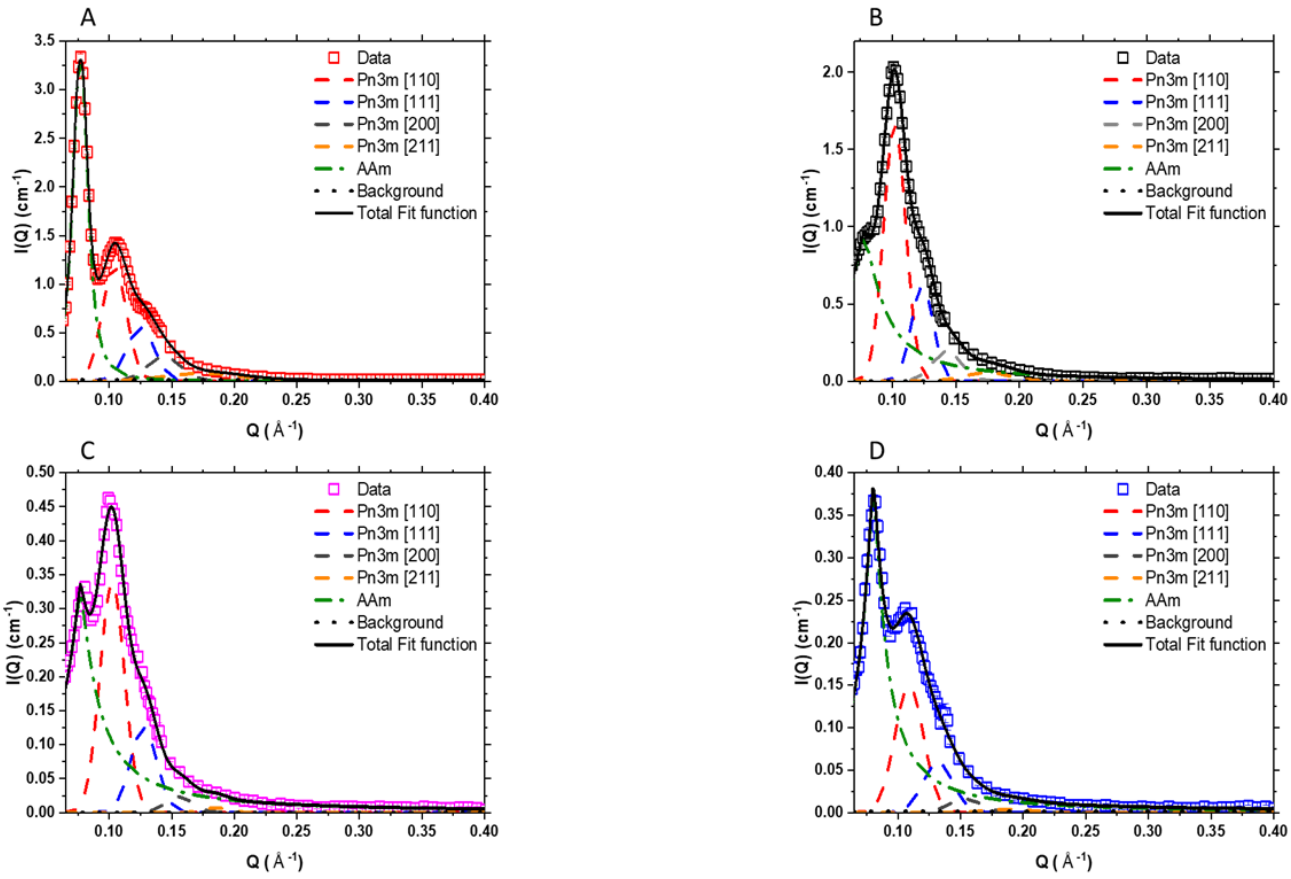

Figure S3 Diffraction pattern of the Cubosomes in presence of AAm monomers: (A) 10 wt% PT/1 wt% Lap, (B) 10 wt% PT/0.5 wt% Lap, (C) 5 wt% PT/1 wt% Lap, (D) 5 wt% PT/0.5 wt% Lap. The points are the experimental data, the continuous lines are the fit function (see main text), the dashed lines are the Gaussian functions composing the fit function, the dashed dotted lines are the peak function due to the monomers and the dotted lines are the flat background.

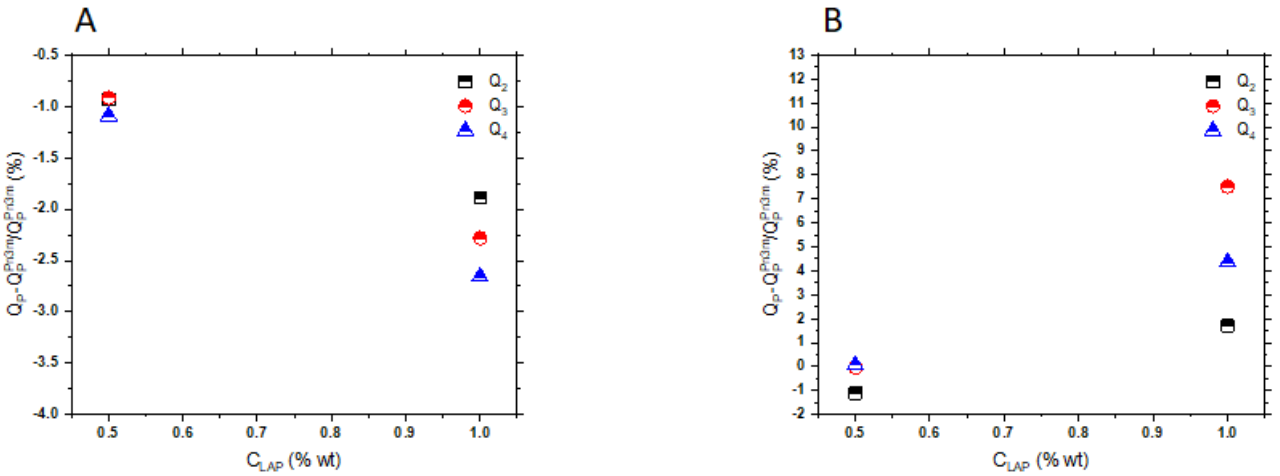

Figure S4 Percentage deviation between the peaks positions found by the data fitting and the equivalent Pn3m phase in function of the Laponite concentration for Cubosomes in presence of AAm monomers at 10 wt% PT (A) and at 5 wt% PT (B). The position of the Pn3m structures have been calculated using the Q_1 value as the position of the $\{h,l,k\} = \{110\}$ peak.

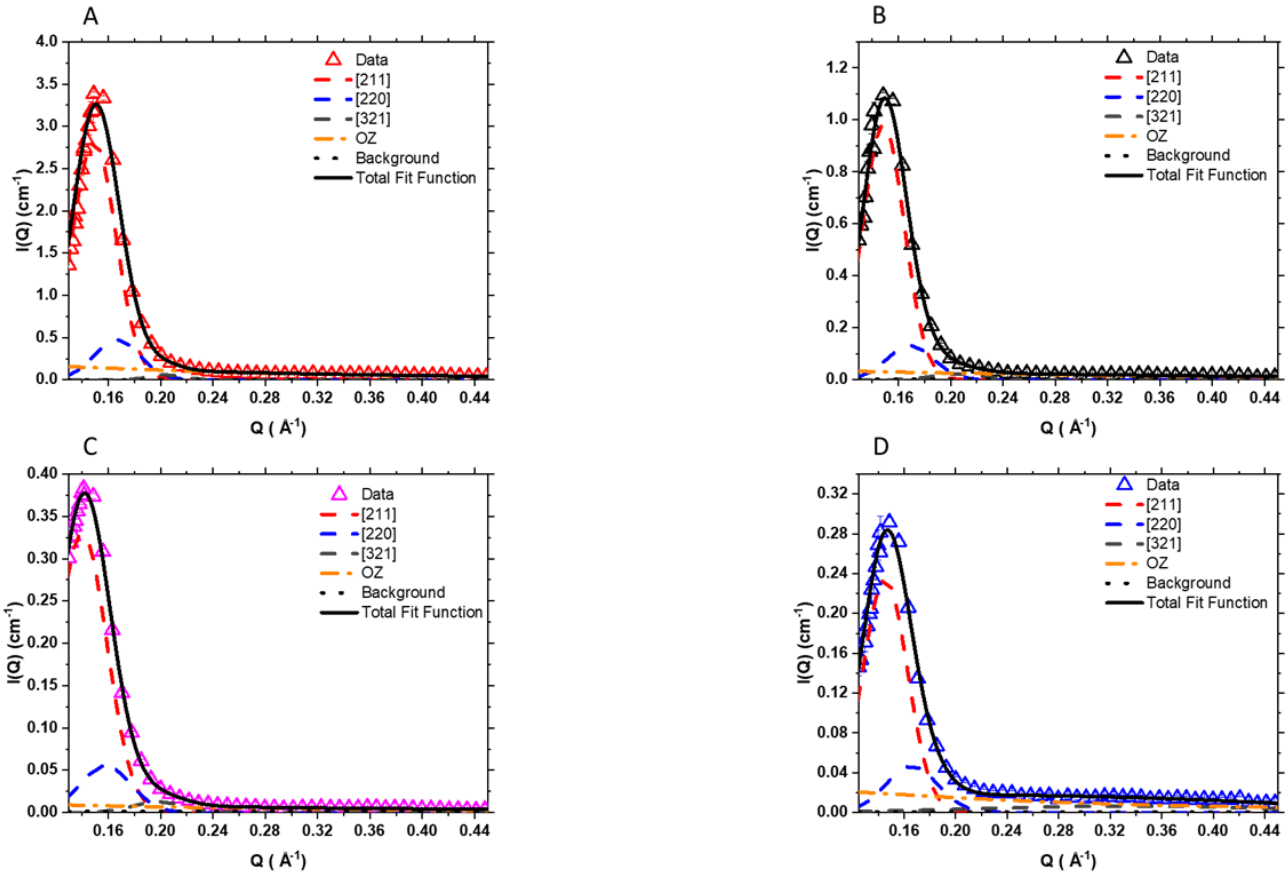

Figure S5 Diffraction pattern of the formed hydrogels: (A) 10 wt% PT/1 wt% Lap, (B) 10 wt% PT/0.5 wt% Lap, (C) 5 wt% PT/1 wt% Lap, (D) 5 wt% PT/0.5 wt% Lap. The points are the experimental data, the continuous lines are the fit function (see main text), the dashed lines are the Gaussian functions composing the fit function, the dashed dotted lines the OZ functions due to the polymer network and the dotted lines are the flat backgrounds.

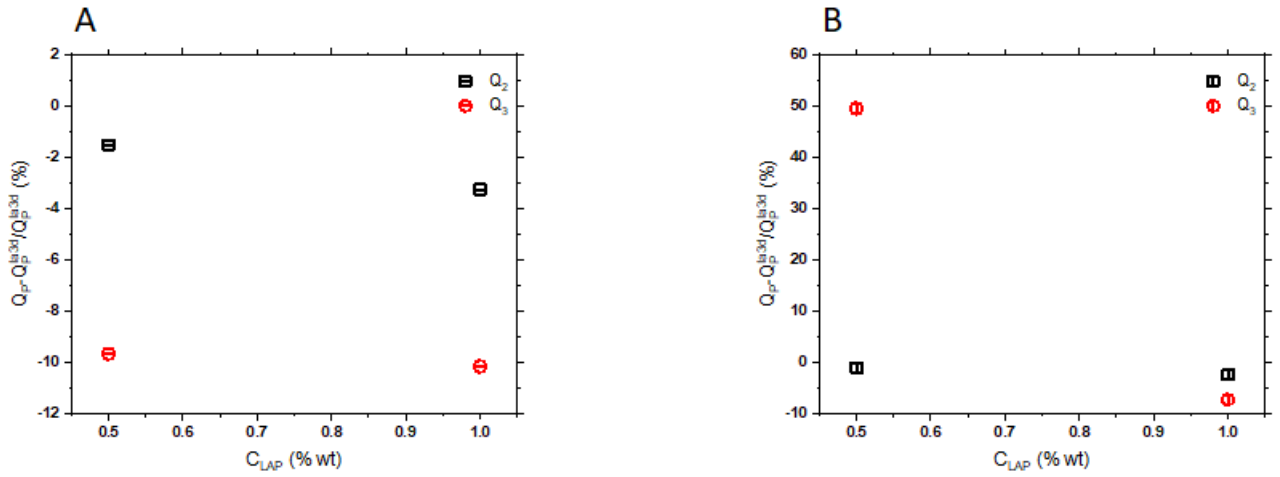

Figure S6 Percentage deviation between the peaks positions found by the data fitting and the equivalent Ia3d phase in function of the Laponite concentration for the formed hydrogels at 10 wt% PT (A) and at 5 wt% PT (B). The position of the Ia3d structures have been calculated using the Q_1 value as the position of the $\{h,l,k\} = \{211\}$ peak.

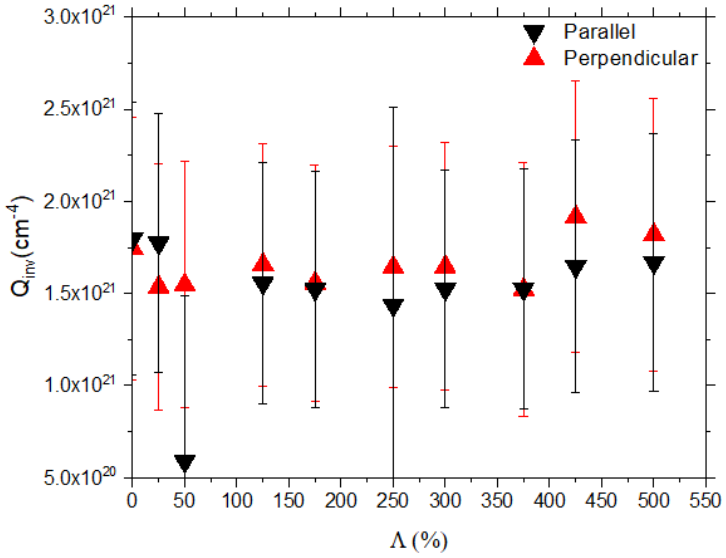

Figure S7 Evolution of the scattering invariant Q_{inv} as a function of the stretching ratio Λ for the parallel direction (black) and the perpendicular direction (red).