Electronic Supplementary Material (ESI) for Biomaterials Science.

Impact of the biomolecular corona on the structure of PEGylated liposomes

Luca Digiacomo,[a,b] Daniela Pozzi,[a] Heinz Amenitsch,[c] and Giulio Caracciolo[a,*]

Electronic Supplementary Information

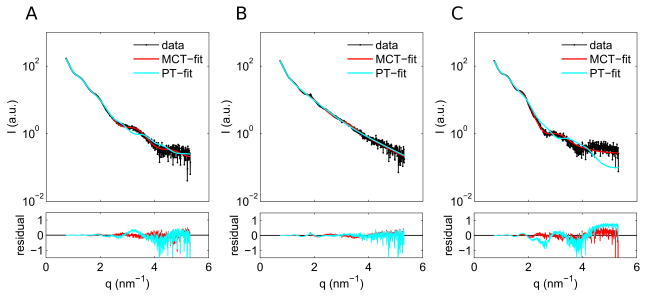

Fig. S1 SAXS data (black) and fitting curves (red for Caillé model and cyan for paracrystalline model) of CL-HP complexes. (A) DOTAP-DOPC, (B) DC-Chol-DOPE and (C) DOTAP-DOPC-DC-Chol-DOPE

Internal structure of PEGylated CL-HP complexes

SAXS curves of liposomes exposed to human plasma can not be described through the form factor only, due to the presence of Bragg peaks. The introduction of a structure factor implies a degree of spatial periodicity of the systems. In other words, SAXS curves of liposomes after incubation with human plasma are ascribable to the scattered intensity from stacks of bilayers. Therefore, these systems are described as multilamellar complexes. For these systems, different kinds of bilayer fluctuations contribute in different ways to shape the resulting SAXS pattern. As an instance, the paracrystalline theory describes the stacking disorder as due to displacements from the mean bilayer position and the modified Caillé theory focuses on the bending fluctuations, which are caused by bilayer undulations (Pabst et al. 2003). The expressions of the corresponding structure factors are

$$S_1(q) = N + 2\sum_{k=1}^{N-1}(N-k)\cos(kqd)\exp\left\{-\frac{k^2q^2\Delta^2}{2}\right\} \quad \text{(S1)}$$

for the paracrystalline theory and

$$S_2(q) = N + 2\sum_{k=1}^{N-1}(N-k)\cos(kqd)\exp\left\{-\frac{q^2d^2\eta[\gamma+\log(k\pi)]}{4\pi^2}\right\} \quad \text{(S2)}$$

for the modified Caillé theory. Here, N is the total number of layers within the scattering domain, Δ denotes the mean square fluctuations of the bilayers, d quantifies the extent of the scattering domain, η is the Caillé parameter (which is related to the bilayer bending rigidity) and γ is the Euler's constant. We employed both the structure factors in the fitting procedure of the experimental SAXS curves. The resulting profiles are shown in Fig. S1. Slight differences have been detected, according to which the modified Caillé theory describes better the investigated systems.

UnPEGylayed systems

In this work, bare liposomes (i.e. unPEGylated) have been used as a control. Fig. S2 shows synchrotron SAXS curve and corresponding fitting curves of unPEGylated CLs and unPEGylated CL-HP complexes.

[a]*Department of Molecular Medicine, "Sapienza" University of Rome, viale Regina Elena 291, 00161 Rome, Italy*
[b]*Department of Bioscience and Biotechnology, University of Camerino, via Gentile III da Varano, 62032 Camerino (MC), Italy*
[c]*Institute of inorganic Chemistry, Graz University of Technology, Stremayerg. 6/IV, 8010 Graz, Austria*
[*]*Corresponding author.* ✉ *giulio.caracciolo@uniroma1.it*

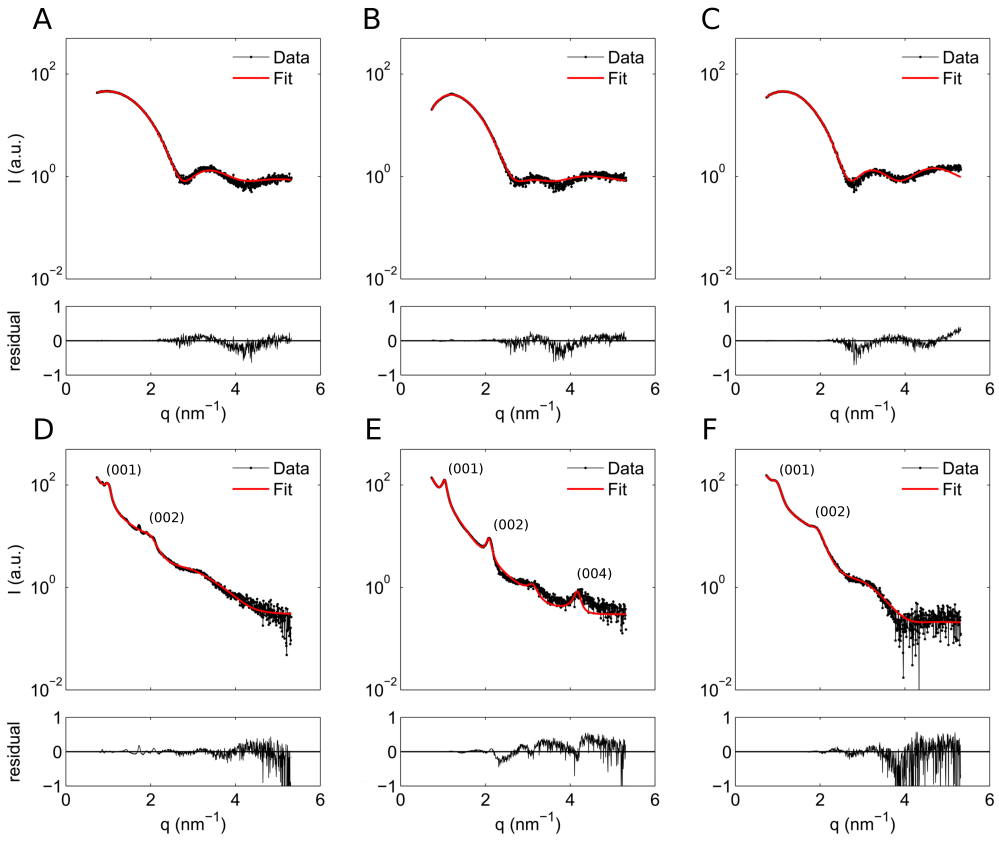

Fig. S2 High resolution synchrotron SAXS data (black) and fitting profiles (red) of unPEGylated CLs: (A) DOTAP-DOPC, (B) DC-Chol-DOPE, (C) DOTAP-DOPC-DC-Chol-DOPE. (D, E, F) Corresponding SAXS curves and fitting curves of unPEGylated CL-HP complexes. Residuals are expressed as relative differences between experimental data and fitting curves.