

## Supporting Information

### **Peptide-Stabilized Emulsions and Gels from an Arginine-Rich Surfactant-Like Peptide with Antimicrobial Activity**

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**SAXS models.** The SAXS intensity from a scattering object without a particular orientation can be approximated as the following equation:<sup>1-2</sup>

$$I(q) \propto \langle F^2(q)S(q) \rangle \quad (1)$$

where  $F^2(q)$  is the scattering particle form factor and  $S(q)$  is the interparticle structure factor.

The SAXS curves for samples containing only A<sub>9</sub>R were fitted using a form factor for random oriented cylindrical shell with circular cross section.<sup>3</sup> The fitting parameters were the core radius  $R$ , the shell thickness  $D$ , the scattering length density of the core, shell and solvent  $\eta_{\text{core}}$ ,  $\eta_{\text{shel}}$  and  $\eta_{\text{core}}$  and the background BG.

The SAXS curves for vesicle were fitted using a form factor for a Gaussian lipid bilayer together with a structure factor for a multilayer structure influenced by thermal fluctuations.<sup>4-5</sup> The details of the Gaussian lipid bilayer model are given elsewhere.<sup>6</sup> Briefly, the model assumes an electron density profile comprising Gaussian functions for the head groups on either side of the bilayer and another Gaussian for the hydrocarbon chain interior. The midpoint of the bilayer is defined as  $z = 0 = z_C$ . In our model we assumed a Gaussian distribution of inter-head group thicknesses  $2z_H$ , with an associated degree of polydispersity  $\Delta_{2z_H}$ . The fitting parameters of the model are the electron densities of the head group ( $\rho_H$ ), the layer thickness  $z_H$ , the electron density of the hydrocarbon chains ( $\rho_C$ ), the standard deviation of the position of the Gaussian peak at  $z_H$  ( $\sigma_H$ ), the standard deviation of the position of the Gaussian peak at  $z_C$  ( $\sigma_C$ ), and  $\Delta_{2z_H}$ .

The structure factor was modelled using the modified Caillé theory.<sup>4-5</sup>  $S(q)$  depends on the total number of layers ( $N$ ) within a scattering domain, the diffuse background of uncorrelated bilayers ( $N_{diff}$ ) the bilayer spacing ( $d$ ) and the Caillé parameter ( $\eta$ ):

$$\eta = \pi k_B T / 2d^2 (BK_c)^{1/2} \quad (2)$$

which is a measure for the bilayer fluctuations and depends on the bilayer rigidity  $K_c$  and the bulk modulus of compression  $B$ . All fitting was done using the software SASfit.<sup>7</sup>

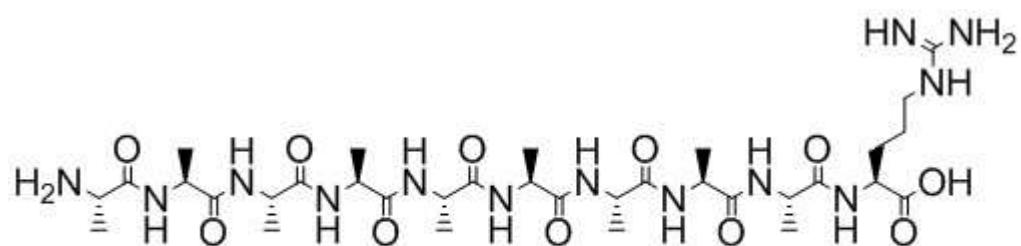
**Table S1.** Parameters extracted from the fitting of the SAXS curve in Figures 1e,2c,S3: core radius  $R$ , shell thickness  $D_r$  and scattering length density of the core, shell and solvent  $\eta_{\text{core}}$ ,  $\eta_{\text{shell}}$  and  $\eta_{\text{solv}}$  and background BG.

	0.08 wt% A <sub>9</sub> R	0.5 wt% A <sub>9</sub> R	1 wt% A <sub>9</sub> R	9.5 wt% A <sub>9</sub> R
$R \pm \Delta R$ (Å)	$18 \pm 10$	$18 \pm 10$	$20.3 \pm 10.8$	$15 \pm 10$
$D_r$ (Å)	4	4	3	2
$\eta_{\text{core}}$ (a.u.)	$7.17 \times 10^{-8}$	$1.39 \times 10^{-6}$	$1.7 \times 10^{-6}$	$4.3 \times 10^{-2}$
$\eta_{\text{shell}}$ (a.u.)	$8.25 \times 10^{-9}$	$1.75 \times 10^{-7}$	$-5.0 \times 10^{-7}$	$8.5 \times 10^{-2}$
$\eta_{\text{solv}}$ (a.u.)	0	0	0	0
BG	$2 \times 10^{-4}$	$1.1 \times 10^{-3}$	$1.4 \times 10^{-3}$	10

**Table S2.** Parameters extracted from the fitting of the SAXS in Figure S2.

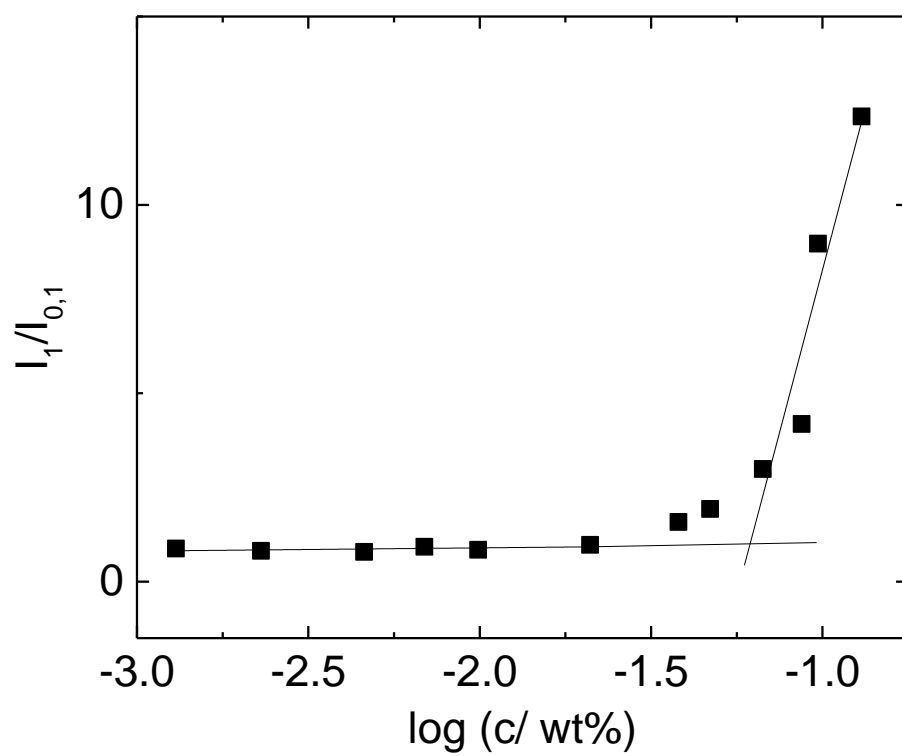
Sample \ Parameter	* $\phi_{\text{POPG}} = 0.2$	$\phi_{\text{POPG}} = 0.2 + 0.08 \text{ wt\% A}_9\text{R}$	* $\phi_{\text{DOPC}} = 0.2$	$\phi_{\text{DOPC}} = 0.2 + 0.08 \text{ wt\% A}_9\text{R}$	
N1 [arb. units]	1	--	0.25	--	--
$\Delta_{2z_H}$ [Å]	5	--	5	--	--
$2z_H$ [Å]	44	49	44	38.5	38.5
$\sigma_H$ [Å]	3	2.3	3.1	2.2	2.8
$l_H = 2z_H + \sigma_H$	47	51.3	47.1	40.7	41.3
$\rho_H$ [rel. units]	$2.2 \times 10^{-3}$	$3.25 \times 10^{-6}$	$1 \times 10^{-3}$	$3.6 \times 10^{-6}$	$1 \times 10^{-6}$
$\sigma_C$ [Å]	1	1	1	1.37	1
$\rho_C$ [rel. units]	$-8.5 \times 10^{-3}$	$-3.93 \times 10^{-5}$	$-5.5 \times 10^{-3}$	$-1.58 \times 10^{-5}$	$-1 \times 10^{-5}$
$N$	--	2	8	2	6
$d_o$	--	81	64	65	65
$l_w = d_o - 2z_H$	--	29.7	17	24.3	23.7
$N_{diff}$	--	0.3	0.3	0.25	0.2
$\eta$	--	0.5	0.5	3.1	0.5
$BG$	0.1	--	--	0.2	0.2

**Key: Gaussian bilayer form factor:** scale factor N1, Gaussian half-width at half-maximum for polydispersity  $\Delta_{2z_H}$ , inter-head group thicknesses  $2z_H$ , Gaussian half-width for outer layer surface  $\sigma_H$ , electron density for headgroup  $\rho_H$ , Gaussian half-width for inner layer  $\sigma_C$ , relative electron density for inner layer  $\rho_C$ . **Caillé structure factor:** total number of layers  $N$ ; layer thickness  $d_o$ ; Caillé parameter  $\eta$ ; diffuse background  $N_{diff}$ . \*data extracted from ref.<sup>8</sup> are displayed here only as a reference.  $l_w$  and  $l_H$  are the thickness of the water and the lipid bilayer respectively.

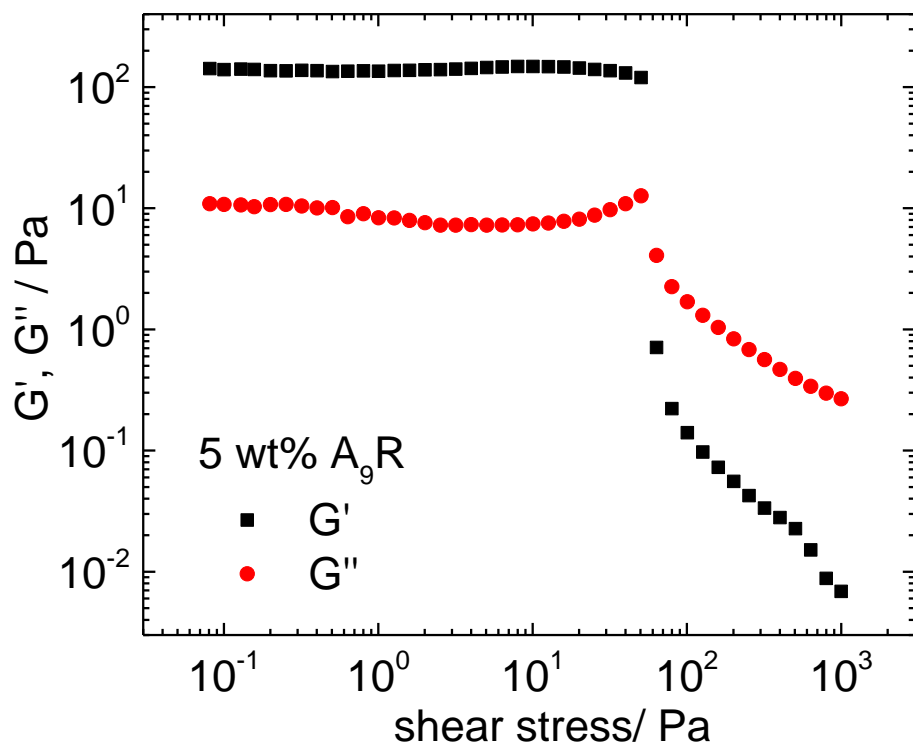


A<sub>9</sub>R

**Scheme S1.** Molecular structure of A<sub>9</sub>R

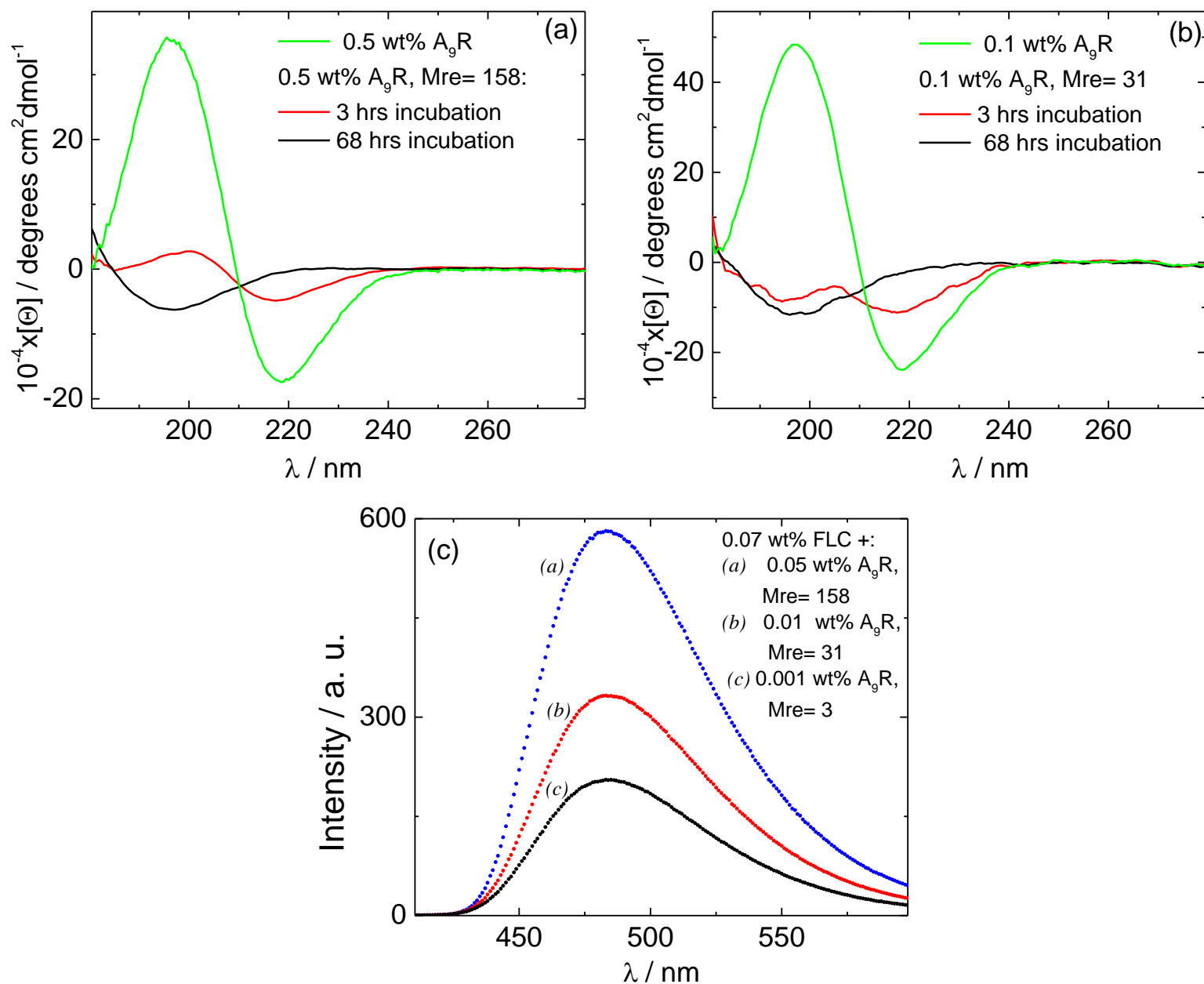


**Figure S1.** Pyrene assay for A<sub>9</sub>R .

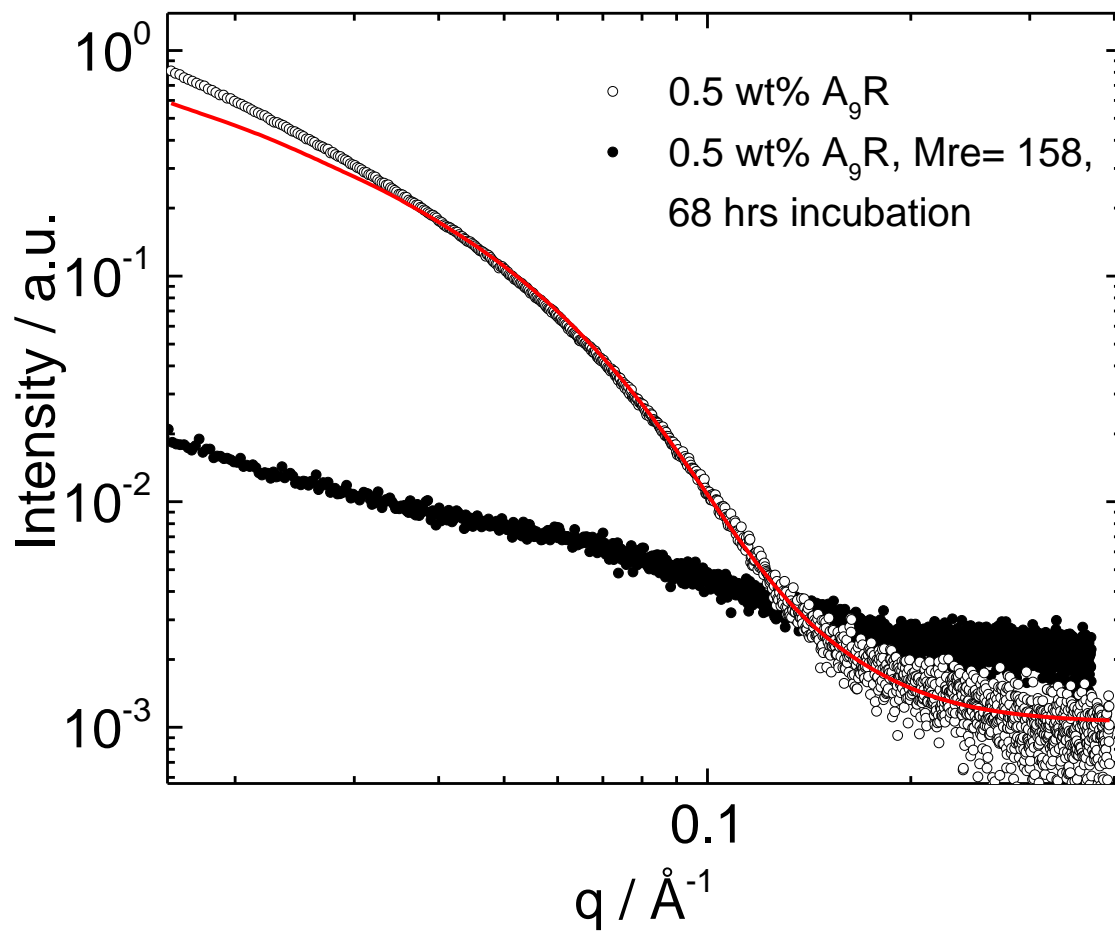


**Figure S2.** Plot of dynamic shear moduli versus shear stress to determine the linear viscoelastic regime for 5 wt%  $A_9R$  hydrogel.

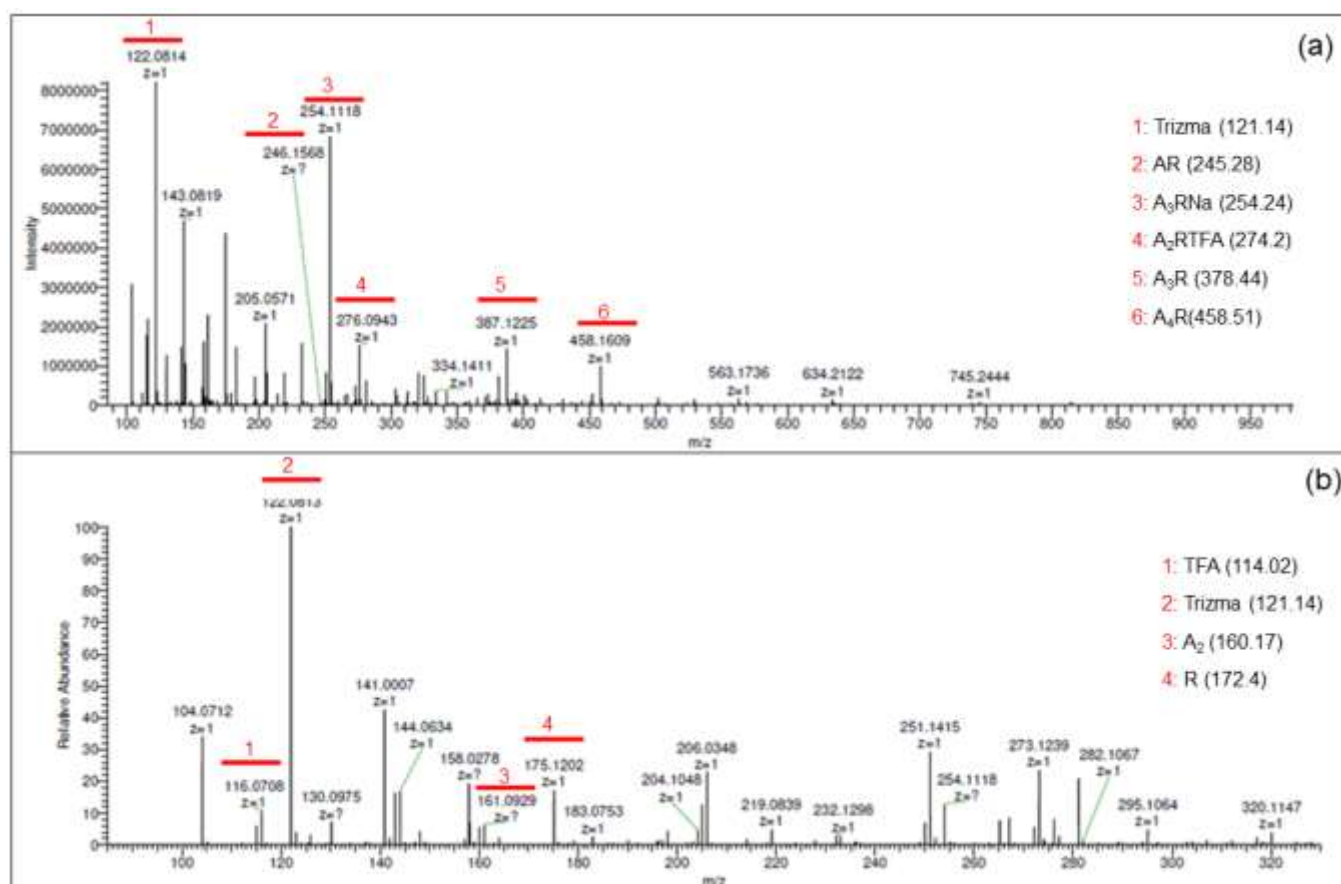




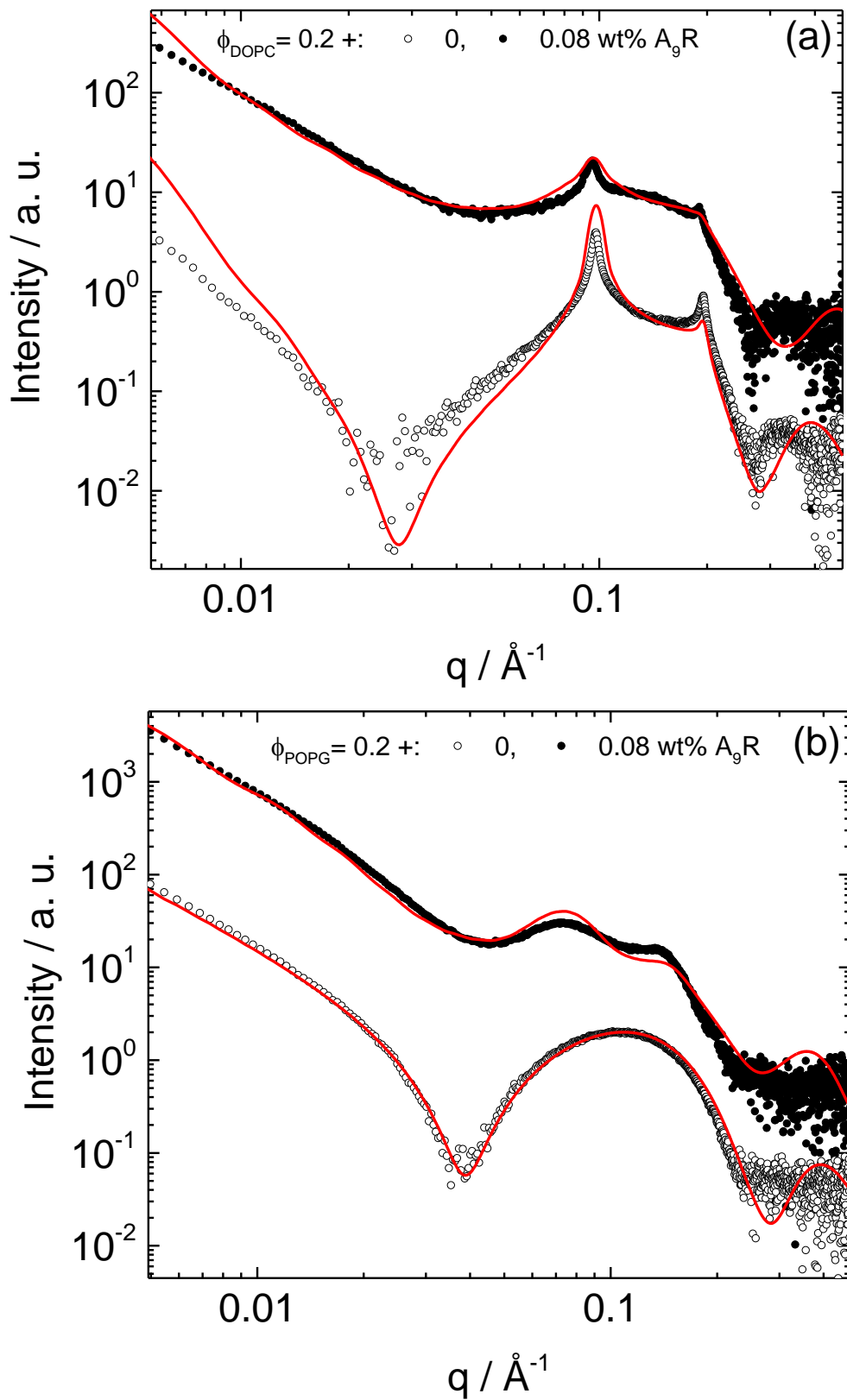
**Figure S3.** Elastase enzymatic assay for A<sub>9</sub>R: CD data for (a) 0.5 wt% A<sub>9</sub>R, M<sub>re</sub> = 158 and (b) 0.1 wt% A<sub>9</sub>R, M<sub>re</sub> = 31. CD controls for pure A<sub>9</sub>R solutions are displayed in (a-b). (c) FLC assay results at 0.07 wt% FLC and A<sub>9</sub>R/elastase ratios used in Figures (a-b). Data for 0.001 wt% A<sub>9</sub>R (M<sub>re</sub> = 3) is included to confirm the trend of the fluorescence intensity.



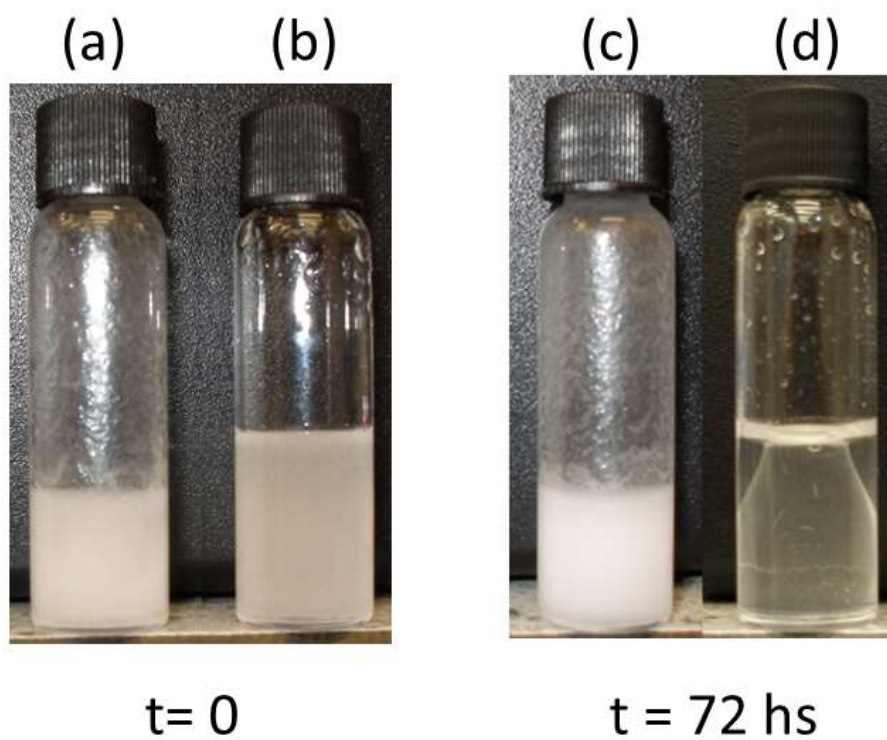
**Figure S4.** SAXS data for 0.5 wt%  $A_9R$  in water, and 0.5 wt%  $A_9R$  incubated with elastase at  $M_{re} = 158$  for 68 hrs (same sample as in Figure S3a). The full red line is a model form factor fit to the experimental data.



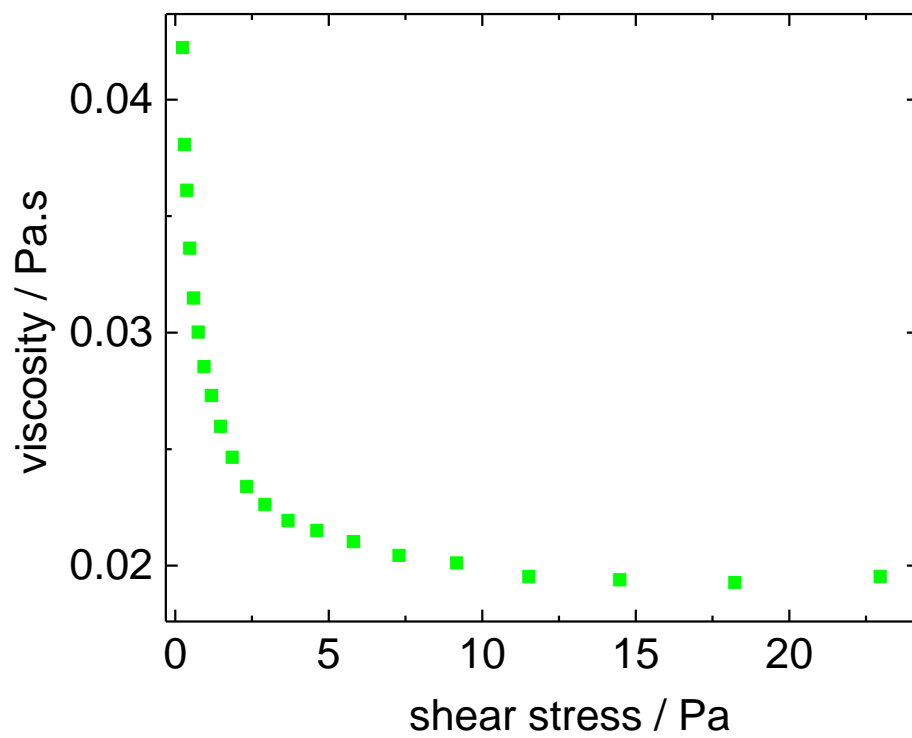
**Figure S5.** (a-b) ESI-MS for a sample containing 0.1 wt% A<sub>9</sub>R and M<sub>re</sub>= 30.8. Experiment performed for the same sample for which data is presented in Figure S2b (68 hrs of incubation). The band for the A<sub>9</sub>R molecular weight (814.9 g mol<sup>-1</sup>) is missing in the ESI spectra, due to elastase enzymatic degradation.



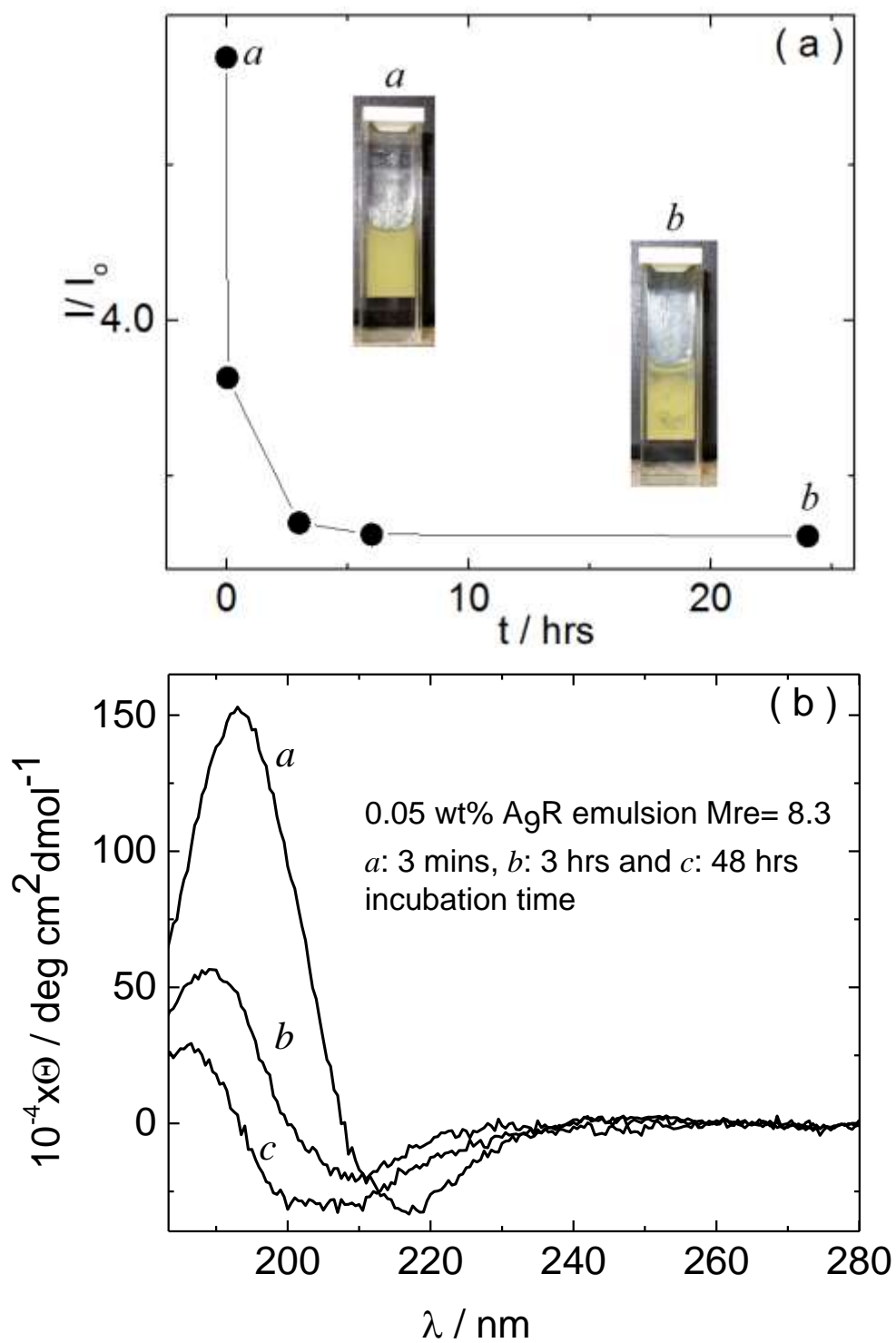
**Figure S6.** SAXS data for vesicles containing (a)  $\phi_{\text{DOPC}}=0.2 + 0$  or 0.08 wt%  $A_9R$  and (b)  $\phi_{\text{POPG}}=0.2 + 0$  or 0.08 wt%  $A_9R$ . The full red lines are fitted form factor profiles. Parameters from the fittings are displayed in Table S2. Results for 0 wt%  $A_9R$  are displayed as a reference.<sup>9</sup>



**Figure S7.** Stability of (a,c) 0.05 wt% A<sub>9</sub>R emulsion compared to (b,d) water/1-bromohexadecane 30/70 v/v mixture: samples immediately after mixing (a,b) and 72 hrs after mixing (c,d).



**Figure S8.** Viscosity versus shear stress for 0.05 wt% A<sub>9</sub>R emulsion.



**Figure S9.** 0.05 wt% A<sub>9</sub>R emulsion treated with elastase at  $M_{re}=8.3$ . (a) Time evolution of ThT fluorescence intensity ratio ( $I/I_0$ ) together with Images of the emulsion stained with ThT (i) immediately and (ii) after 24 hrs incubation time. (b) CD profiles as a function of the incubation time.

## References

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