

Supplementary

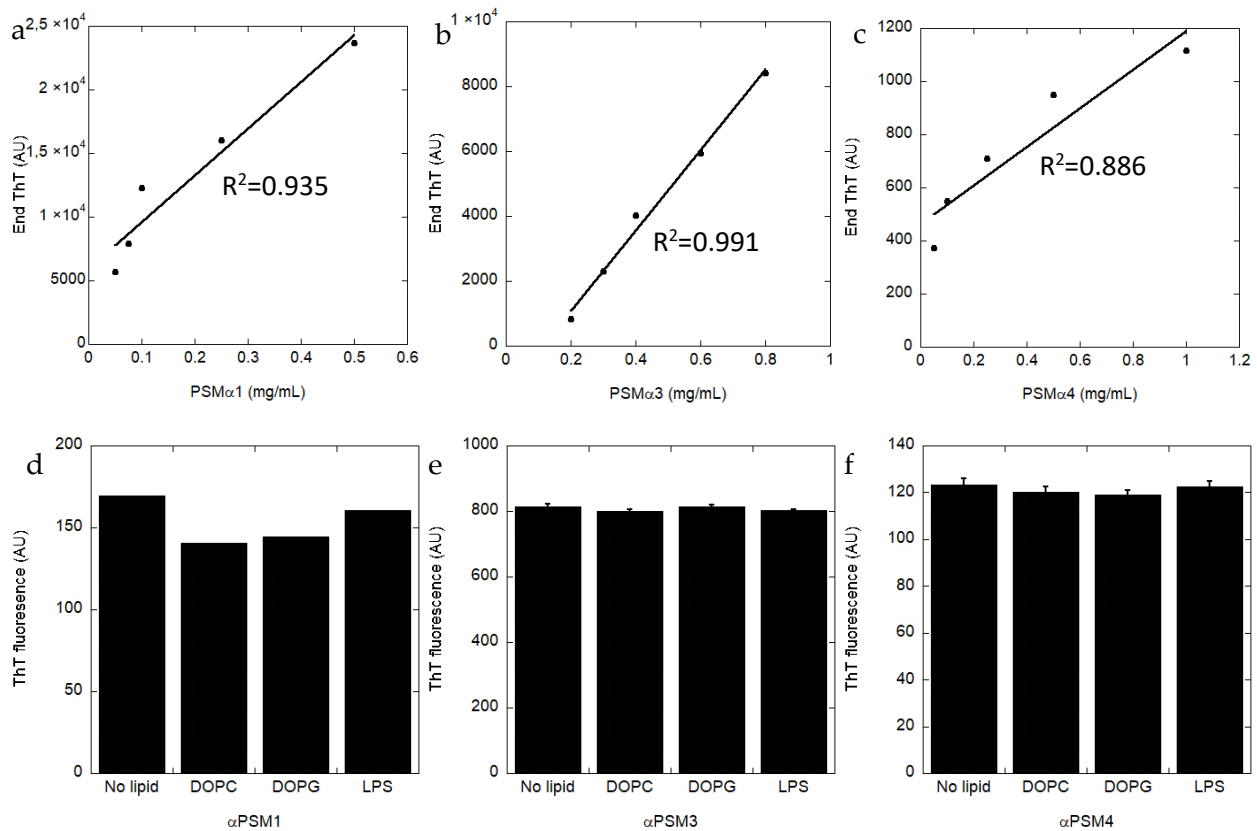


Figure S1. Experimental controls. (a) End ThT fluorescence signal for α PSM1. (b) End ThT fluorescence signal for α PSM3. (c) End ThT fluorescence signal for α PSM4. (d) ThT fluorescence signal of preformed α PSM1 aggregates (0.25 mg/mL) in the presence and absence of 100 μ g/mL DOPC, DOPG, and LPS lipid vesicles. (e) ThT fluorescence signal of preformed α PSM3 aggregates (0.25 mg/mL) in the presence and absence of 100 μ g/mL DOPC, DOPG, and LPS lipid vesicles. (f) ThT fluorescence signal of preformed α PSM4 aggregates (0.25 mg/mL) in the presence and absence of 100 μ g/mL DOPC, DOPG, and LPS lipid vesicles.

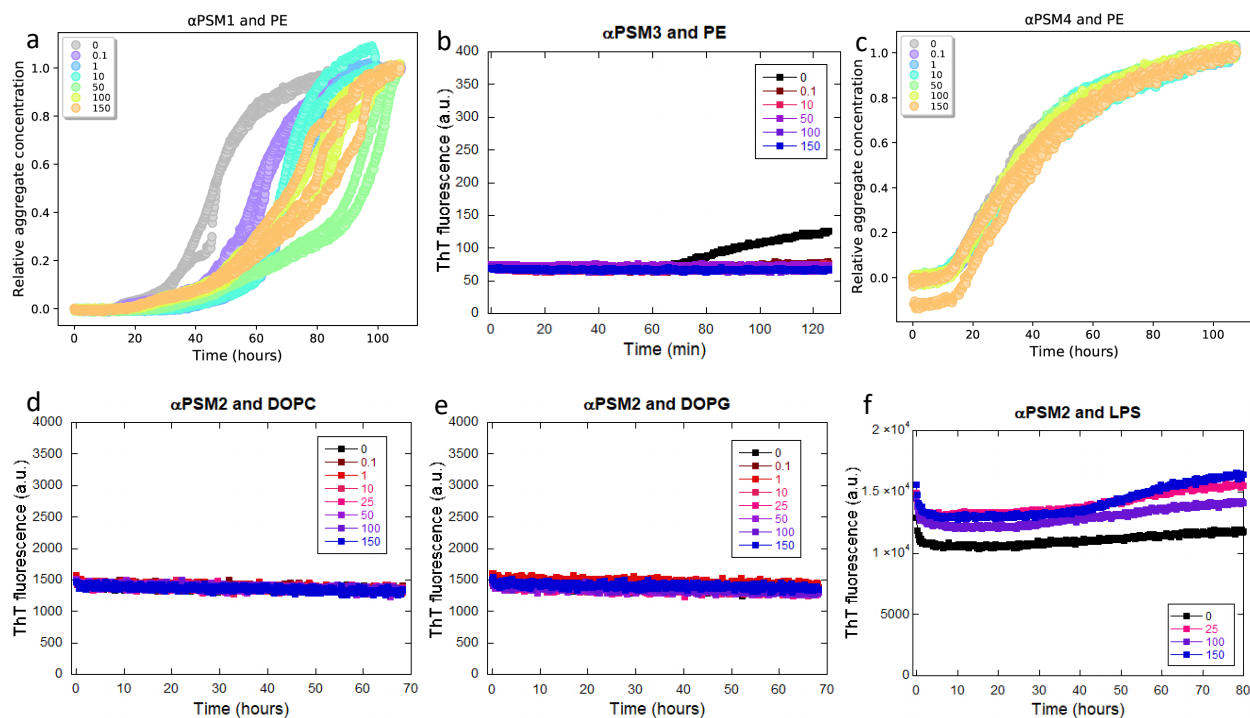


Figure S2. Additional experimental kinetic results. (a) Normalized kinetic data with α PSM1 (0.25 mg/mL) added PE (concentration stated in μ g/mL). (b) Raw kinetic data with α PSM3 (0.5 mg/mL) added PE (concentration stated in μ g/mL). (c) Normalized kinetic data with α PSM4 (0.25 mg/mL) added PE (concentration stated in μ g/mL). All kinetic experiments were carried out in triplicates. Please note the differences in the y- and x-axis between the figures. (d) Raw kinetic data with α PSM2 (0.25 mg/mL) added DOPC (concentration stated in μ g/mL). (e) Raw kinetic data with α PSM2 (0.25 mg/mL) added DOPG (concentration stated in μ g/mL). (f) Raw kinetic data with α PSM2 (0.25 mg/mL) added LPS (concentration stated in μ g/mL).

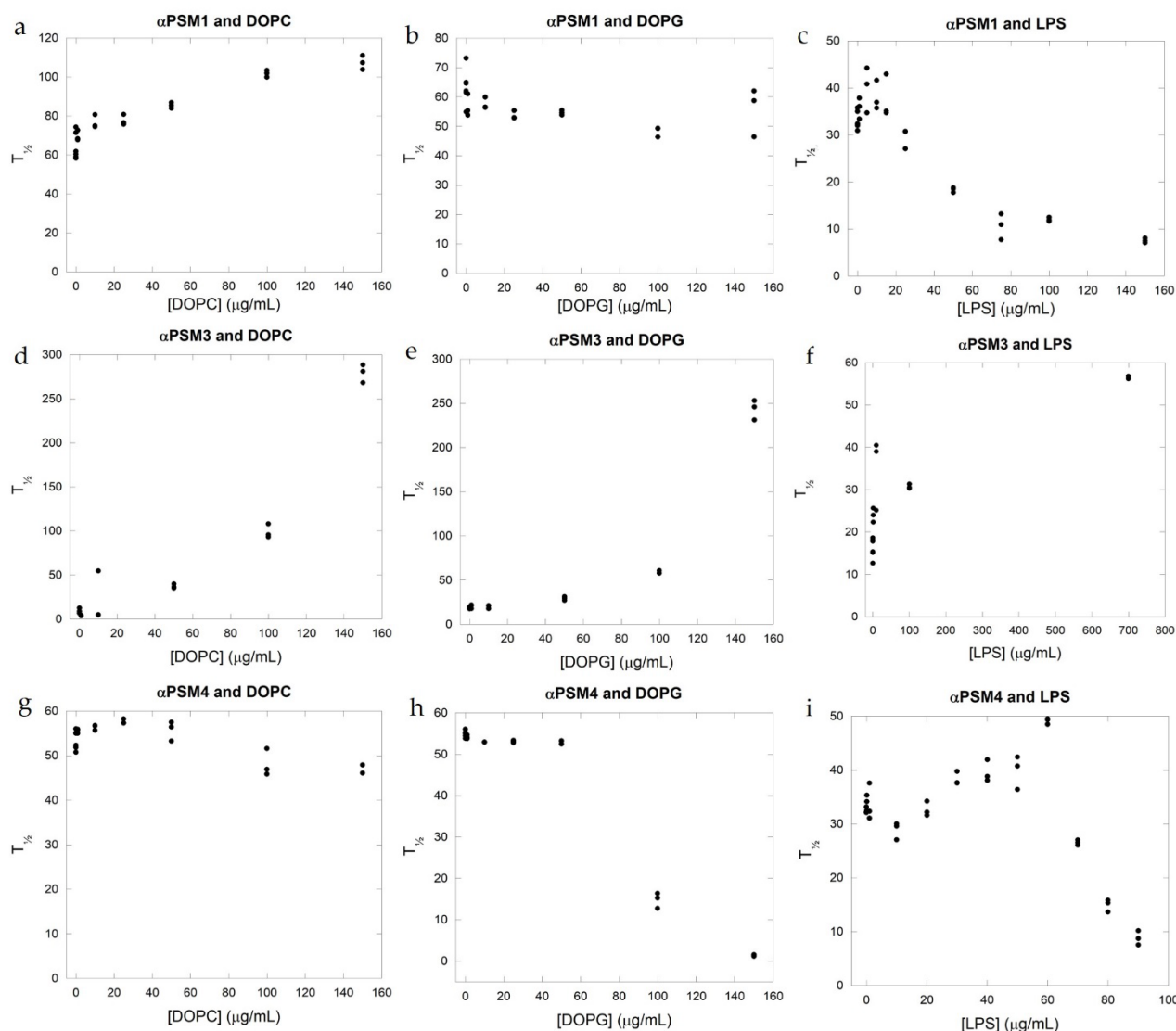


Figure S3. Half-time plots for αPSM1 , αPSM3 and αPSM4 when added various concentrations of lipids. The half-times are a function of lipid concentration in $\mu\text{g/mL}$ obtained from the three repeats of the aggregation experiments for each peptide and lipid combinations. (a) αPSM1 and DOPC (b) αPSM1 and DOPG (c) αPSM1 and LPS (d) αPSM3 and DOPC (e) αPSM3 and DOPG (f) αPSM3 and LPS (g) αPSM4 and DOPC (h) αPSM4 and DOPG (i) αPSM4 and LPS. Please note the differences in the y-axis.

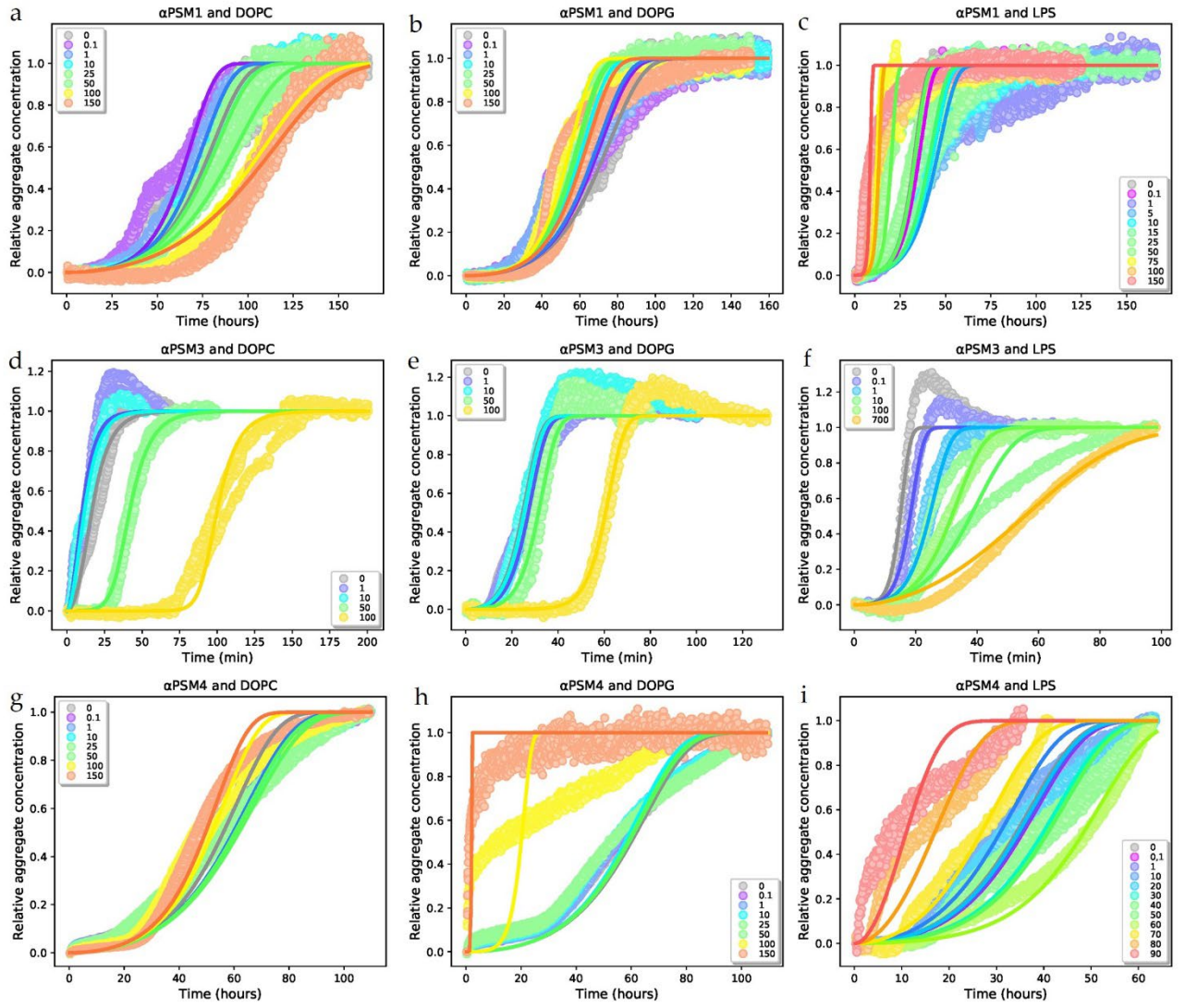


Figure S4. Fitting of aggregation kinetic data for PSM peptides in the presence of lipids using Amylofit. All kinetic data is fitted to a secondary nucleation-dominated model. **(a)** α PSM1 in the presence of DOPC lipid vesicles fitted to a secondary nucleation model ($k+k_2$ as fitting parameter and $k+k_n$ as a global fit). **(b)** α PSM1 in the presence of DOPG lipid vesicles fitted to a secondary nucleation model ($k+k_2$ as fitting parameter and $k+k_n$ as a global fit). **(c)** α PSM1 in the presence of LPS lipid vesicles fitted to a secondary nucleation model ($k+k_2$ as fitting parameter). **(d)** α PSM3 in the presence of DOPC lipid vesicles fitted to a secondary nucleation model ($k+k_n$ as fitting parameter). **(e)** α PSM3 in the presence of DOPG lipid vesicles fitted to a secondary nucleation model ($k+k_n$ as fitting parameter and $k+k_2$ as a global fit). **(f)** α PSM3 in the presence of LPS lipid vesicles fitted to a secondary nucleation model ($k+k_2$ as fitting parameter). **(g)** α PSM4 in the presence of DOPC lipid vesicles fitted to a secondary nucleation model ($k+k_2$ as fitting parameter). **(h)** α PSM4 in the presence of DOPG lipid vesicles fitted to a secondary nucleation model ($k+k_2$ as fitting parameter). **(i)** α PSM4 in the presence of LPS lipid vesicles fitted to a secondary nucleation model ($k+k_n$ as fitting parameter). Parameters from the data fitting are shown in **Table S1**.

Table S1: Kinetic parameters from fitting of kinetic data of PSM peptide in the presence of DOPC, DOPG and LPS lipid vesicles.

α PSM1					
[DOPC] ($\mu\text{g/mL}$)	$k+k_2 (M^{-n_c-1} h^{-2})$	[DOPG] ($\mu\text{g/mL}$)	$k+k_2 (M^{-n_c-1} h^{-2})$	[LPS] ($\mu\text{g/mL}$)	$k+k_2 (M^{-n_c-1} h^{-2})$
0	15.4	0	14.9	0	154
0.1	26.5	0.1	18.4	0.1	123
1	19.3	1	19.7	1	67.8
10	13.9	10	29.3	5	55.8
25	13.8	25	34.6	10	68.3
50	8.78	50	33.2	15	75.8
100	4.72	100	42.0	25	148
150	3.74	150	26.4	50	617
				75	2.31×10^3
				100	1.84×10^3
				150	5.91×10^3
m_o	111 μM	m_o	111 μM	m_o	111 μM
$k+k_n$	$3.03 \times 10^{-5} (M^{-n_c} h^{-2})$ global fit	$k+k_n$	$4.32 \times 10^{-5} (M^{-n_c} h^{-2})$ global fit	$k+k_n$	$6.98 \times 10^{-5} (M^{-n_c} h^{-2})$
n_c	7.84×10^{-6}	n_c	7.84×10^{-6}	n_c	7.84×10^{-6}
n_2	1.66×10^{-3}	n_2	1.66×10^{-3}	n_2	1.66×10^{-3}
MRE	5.23×10^{-3}	MRE	6.05×10^{-3}	MRE	1.19×10^{-2}
α PSM3					
[DOPC] ($\mu\text{g/mL}$)	$k+k_n (M^{-n_c} \text{min}^{-2})$	[DOPG] ($\mu\text{g/mL}$)	$k+k_n (M^{-n_c} \text{min}^{-2})$	[LPS] ($\mu\text{g/mL}$)	$k+k_2 (M^{-n_c-1} \text{min}^{-2})$
0	1.86×10^{12}	0	1.44	0	1.52×10^3
1	9.16×10^{12}	1	0.915	0.1	868
10	6.13×10^{12}	10	1.29	1	352
50	6.51×10^9	50	0.401	10	75.8
100	1.64×10^4	100	1.87×10^{-3}	100	147
				700	11.2
m_o	192 μM	m_o	192 μM	m_o	192 μM
$k+k_n$	-	$k+k_n$	-	$k+k_n$	$3.01 \times 10^{-1} (M^{-n_c} \text{min}^{-2})$ global fit
$k+k_2$	$1.80 \times 10^{13} (M^{-n_c-1} \text{min}^{-2})$	$k+k_2$	$255 (M^{-n_c-1} \text{min}^{-2})$ global fit	$k+k_2$	-
n_c	4.00	n_c	1.00	n_c	0.600
n_2	3.00	n_2	0.123	n_2	0.123
MRE	6.38×10^{-3}	MRE	5.36×10^{-3}	MRE	7.55×10^{-3}
α PSM4					
[DOPC] ($\mu\text{g/mL}$)	$k+k_2 (M^{-n_c-1} h^{-2})$	[DOPG] ($\mu\text{g/mL}$)	$k+k_2 (M^{-n_c-1} h^{-2})$	[LPS] ($\mu\text{g/mL}$)	$k+k_n (M^{-n_c} h^{-2})$
0	49.3	0	47.6	0	2.50×10^{-4}
0.1	37.9	0.1	50.4	0.1	2.16×10^{-4}
1	38.0	1	52.2	1	2.22×10^{-4}
10	35.7	10	55.3	10	3.60×10^{-4}
25	32.5	25	51.6	20	2.39×10^{-4}
50	36.0	50	50.4	30	1.40×10^{-4}
100	66.3	100	1.61×10^3	40	1.27×10^{-4}
150	79.5	150	5.32×10^5	50	1.26×10^{-4}
				60	5.92×10^{-5}

				70	5.58×10^{-4}
				80	1.92×10^{-3}
				90	5.26×10^{-3}
m_o	45.4 μM	m_o	40.2 μM	m_o	45.4 μM
$k+k_n$	$7.79 \times 10^{-5} (M^{-n_c} h^{-2})$	$k+k_n$	$7.00 \times 10^{-5} (M^{-n_c} h^{-2})$	$k+k_n$	-
$k+k_2$	-	$k+k_2$	-	$k+k_2$	$130 (M^{-n_c-1} h^{-2})$
n_c	8.00×10^{-6}	n_c	8.00×10^{-6}	n_c	8.00×10^{-5}
n_2	1.70×10^{-3}	n_2	1.70×10^{-3}	n_2	2.00×10^{-2}
MRE	4.45×10^{-3}	MRE	1.38×10^{-2}	MRE	7.17×10^{-3}

Table S2. Wavelength minimum for CD spectra of α PSM peptide fibril samples in the absence and presence of lipid vesicles.

Peptide	DOPC	λ_{\min} (nm)	DOPG	λ_{\min} (nm)	LPS	λ_{\min} (nm)
αPSM1	0 $\mu\text{g/mL}$	219	0 $\mu\text{g/mL}$	219	0 $\mu\text{g/mL}$	219
	50 $\mu\text{g/mL}$	218.5	10 $\mu\text{g/mL}$	217.5	50 $\mu\text{g/mL}$	217.5
	150 $\mu\text{g/mL}$	218.5	150 $\mu\text{g/mL}$	218.5	150 $\mu\text{g/mL}$	218
αPSM3	0 $\mu\text{g/mL}$	224	0 $\mu\text{g/mL}$	224	0 $\mu\text{g/mL}$	224
	50 $\mu\text{g/mL}$	226	10 $\mu\text{g/mL}$	226.4	10 $\mu\text{g/mL}$	225.5
	150 $\mu\text{g/mL}$	227.8	150 $\mu\text{g/mL}$	224.6	700 $\mu\text{g/mL}$	213.5
αPSM4	0 $\mu\text{g/mL}$	216	0 $\mu\text{g/mL}$	216	0 $\mu\text{g/mL}$	216
	25 $\mu\text{g/mL}$	127.9	10 $\mu\text{g/mL}$	126.8	10 $\mu\text{g/mL}$	215.5
	150 $\mu\text{g/mL}$	217.9	150 $\mu\text{g/mL}$	226.9	90 $\mu\text{g/mL}$	222.5

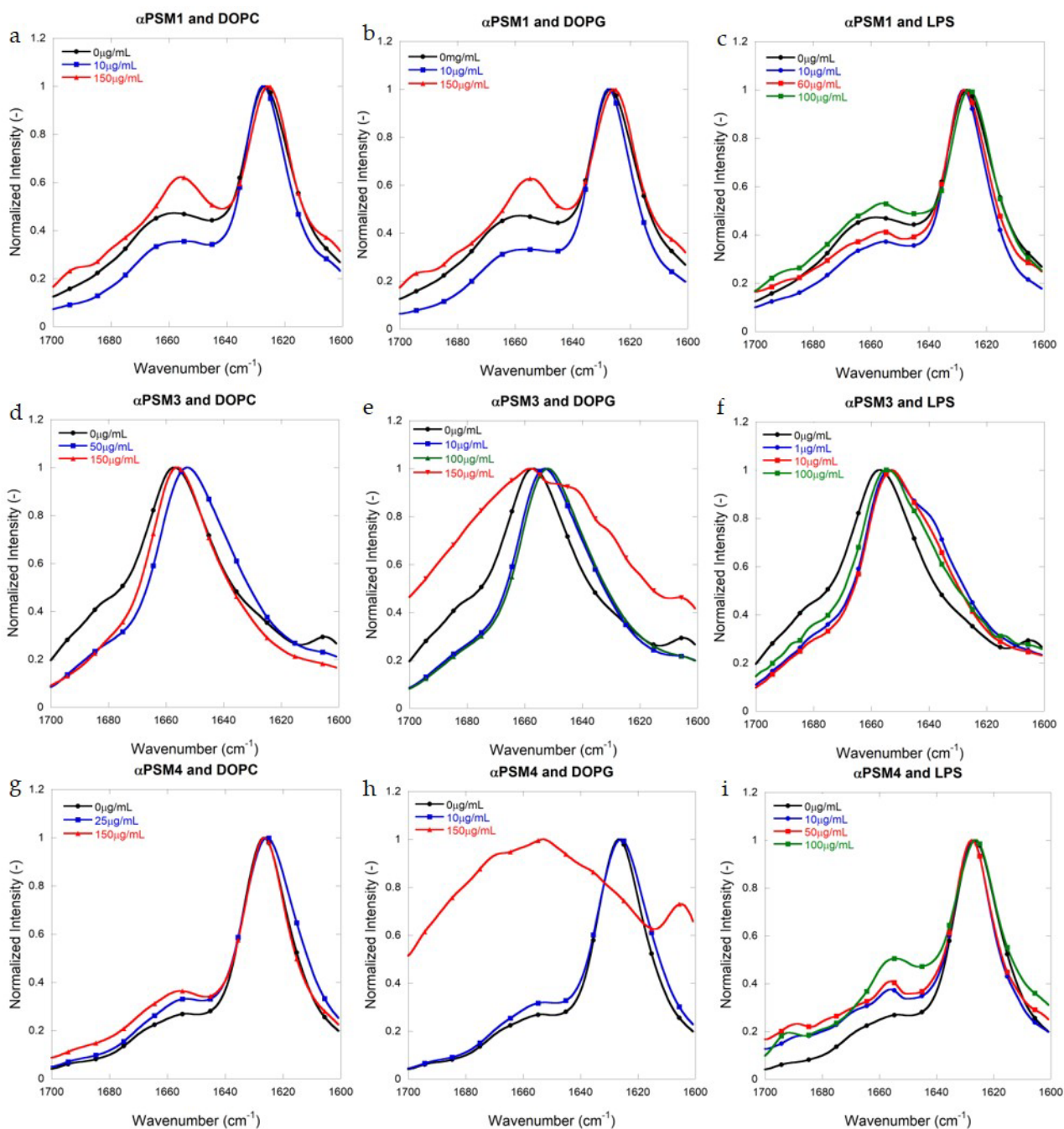


Figure S5. Fourier transform infrared (FTIR) spectroscopy of the amide I' region (1600–1700 cm⁻¹) of fibrils of PSMs variants. (a) PSMα1 and DOPC. (b) PSMα1 and DOPG. (c) PSMα1 and LPS. (d) PSMα3 and DOPC. (e) PSMα3 and DOPG. (f) PSMα3 and LPS. (g) PSMα4 and DOPC. (h) PSMα4 and DOPG. (i) PSMα4 and LPS.

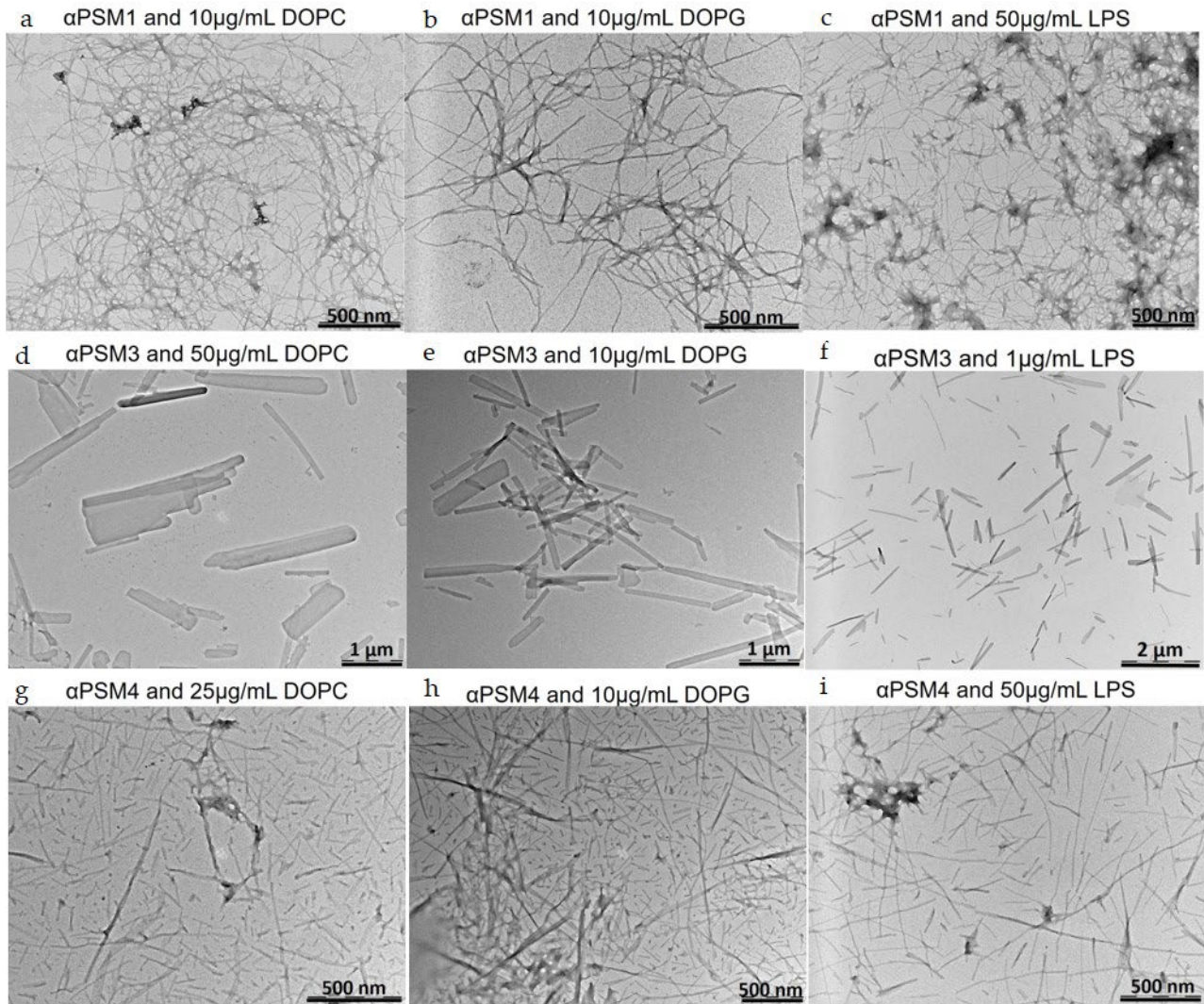


Figure S6. Morphology of aggregates of α PSM peptides with low concentrations of lipids. Transmission electron microscopic images of the end state of PSM peptide fibril samples with DOPC, DOPG and LPS lipid vesicles. (a) α PSM1 and 10 $\mu\text{g/mL}$ DOPC. (b) α PSM1 and 10 $\mu\text{g/mL}$ DOPG. (c) α PSM1 and 50 $\mu\text{g/mL}$ LPS. (d) α PSM3 and 50 $\mu\text{g/mL}$ DOPC. (e) α PSM3 and 10 $\mu\text{g/mL}$ DOPG. (f) α PSM1 and 1 $\mu\text{g/mL}$ LPS. (g) α PSM4 and 25 $\mu\text{g/mL}$ DOPC. (h) α PSM4 and 10 $\mu\text{g/mL}$ DOPG. (i) α PSM4 and 50 $\mu\text{g/mL}$.

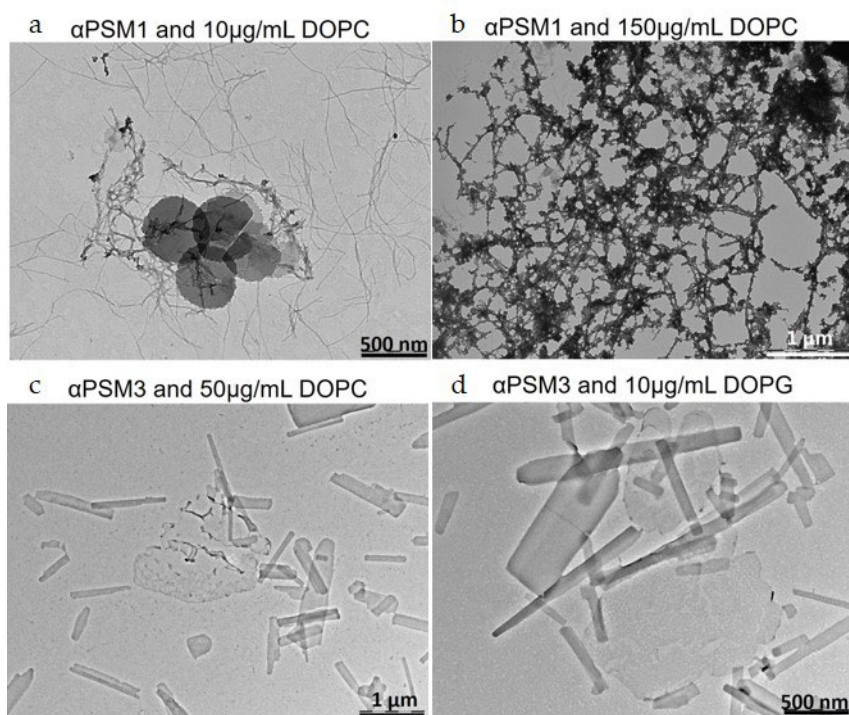


Figure S7. TEM images of lipid vesicles embedded in fibril formation. (a) α PSM1 and 10 μ g/mL DOPC. (b) α PSM1 and 150 μ g/mL DOPC. (c) α PSM3 and 50 μ g/mL DOPC. (d) α PSM3 and 10 μ g/mL DOPG.