Supporting Information for:

Synthesis of Highly Transparent Diblock Copolymer Vesicles *via* RAFT Dispersion Polymerization of 2,2,2-Trifluoroethyl Methacrylate in *n*-Alkanes

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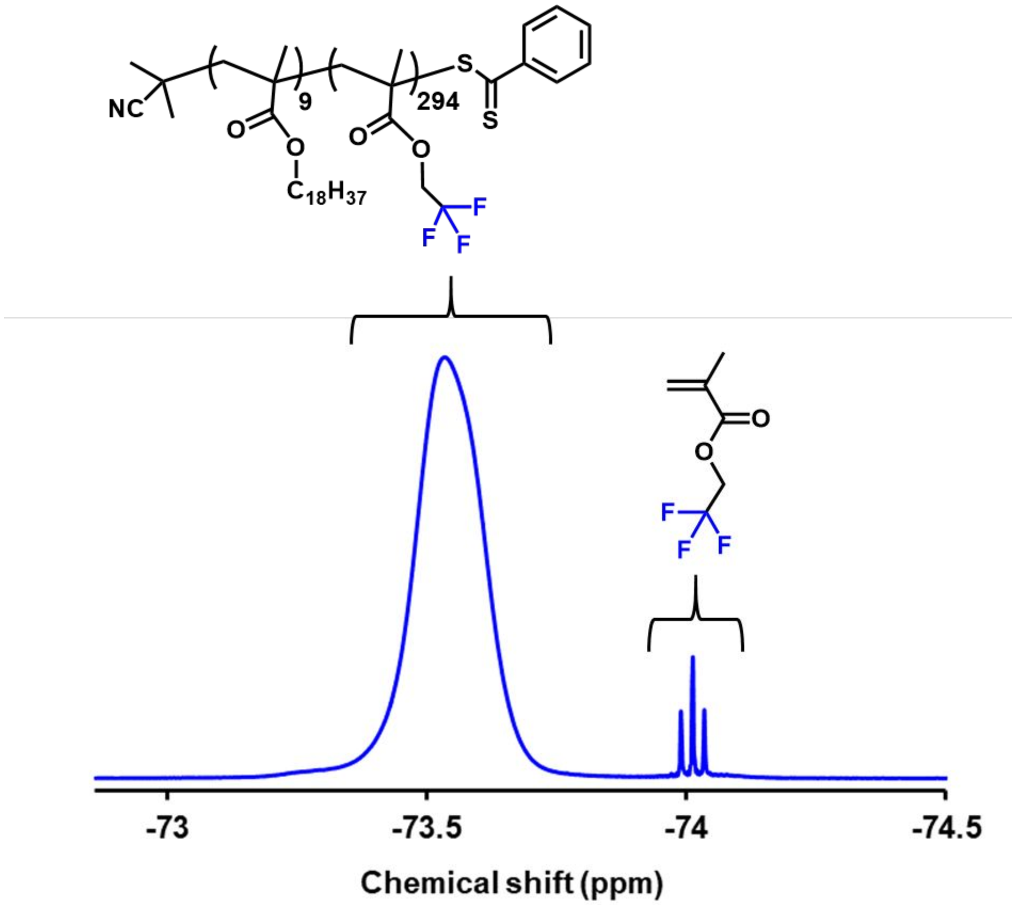

Figure S1. Assigned ^{19}F NMR spectrum recorded in $CDCl_3$ for the $PSMA_9$-$PTFEMA_{294}$ diblock copolymer vesicles prepared *via* RAFT dispersion polymerization of TFEMA at 25% w/w solids in *n*-dodecane at 90 °C. In this case, the target diblock copolymer composition was $PSMA_9$-$PTFEMA_{300}$ and the final TFEMA monomer conversion was 98%.

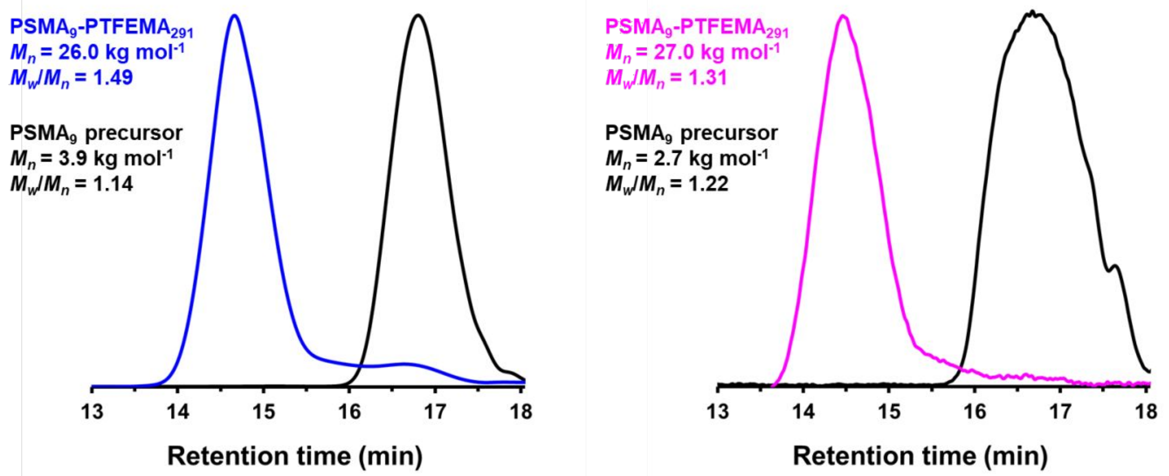

Figure S2. THF GPC curves recorded for the $PSMA_9$-$PTFEMA_{291}$ diblock copolymer vesicles prepared at 20% w/w solids and the corresponding $PSMA_9$ macro-CTA using (a) a refractive index detector *vs*. poly(methyl methacrylate) standards and (b) a UV detector calibrated at a fixed wavelength of 260 nm against polystyrene standards.

Table S1. Summary of the GPC, DLS, and TEM data obtained for a series of $PSMA_9$-$PTFEMA_x$ diblock copolymer nano-objects prepared at 15% w/w in *n*-dodecane. The $PSMA_9$ precursor block is also included as a reference.

Target Composition	**Solids Content (% w/w)**	**TFEMA Conversion (%)**	**THF GPC**		**DLS**		**TEM Morphology**
			M_n (g mol^{-1})	**M_w/M_n**	**D_h (nm)**	**PDI**	
$PSMA_9$ macro-CTA	-	-	2,700	1.22	-	-	-
$PSMA_9$-$PTFEMA_{20}$	15	95	4,200	1.17	-	-	No particles
$PSMA_9$-$PTFEMA_{40}$	15	95	5,900	1.19	19	0.12	Spheres
$PSMA_9$-$PTFEMA_{50}$	15	97	8,800	1.21	138	0.22	Worms
$PSMA_9$-$PTFEMA_{60}$	15	95	9,900	1.17	140	0.23	Worms
$PSMA_9$-$PTFEMA_{65}$	15	95	10,300	1.27	246	0.37	Worms
$PSMA_9$-$PTFEMA_{70}$	15	96	10,100	1.27	143	0.34	Worms
$PSMA_9$-$PTFEMA_{75}$	15	95	10,600	1.18	90	0.18	Mixed phase
$PSMA_9$-$PTFEMA_{80}$	15	96	11,200	1.18	92	0.19	Mixed phase
$PSMA_9$-$PTFEMA_{90}$	15	97	11,500	1.29	95	0.07	Mixed phase
$PSMA_9$-$PTFEMA_{100}$	15	96	12,900	1.24	100	0.08	Vesicles
$PSMA_9$-$PTFEMA_{120}$	15	96	14,500	1.24	108	0.09	Vesicles
$PSMA_9$-$PTFEMA_{140}$	15	95	16,200	1.19	125	0.22	Vesicles
$PSMA_9$-$PTFEMA_{160}$	15	96	18,400	1.32	188	0.24	Vesicles
$PSMA_9$-$PTFEMA_{180}$	15	96	17,600	1.29	165	0.24	Vesicles
$PSMA_9$-$PTFEMA_{200}$	15	96	21,200	1.28	169	0.16	Vesicles
$PSMA_9$-$PTFEMA_{250}$	15	97	23,200	1.29	153	0.04	Vesicles
$PSMA_9$-$PTFEMA_{300}$	15	97	28,500	1.32	158	0.05	Vesicles

Table S2. Summary of the GPC, DLS, and TEM data obtained for a series of PSMA$_9$-PTFEMA$_x$ diblock copolymer nano-objects prepared at 20% and 25% w/w in *n*-dodecane.

Target Composition	**Solids Content (% w/w)**	**TFEMA Conversion (%)**	**THF GPC**		**DLS**		**TEM Morphology**
			M_n **(g mol^{-1})**	M_w/M_n	***D*** **(nm)**	**PDI**	
PSMA$_9$-PTFEMA$_{20}$	20	95	4,600	1.19	-	-	No particles
PSMA$_9$-PTFEMA$_{40}$	20	95	6,400	1.20	21	0.33	Spheres
PSMA$_9$-PTFEMA$_{50}$	20	97	8,800	1.23	55	0.22	Worms
PSMA$_9$-PTFEMA$_{60}$	20	96	9,000	1.20	171	0.24	Worms
PSMA$_9$-PTFEMA$_{65}$	20	97	9,200	1.21	162	0.29	Worms
PSMA$_9$-PTFEMA$_{70}$	20	97	10,100	1.24	257	0.39	Worms
PSMA$_9$-PTFEMA$_{75}$	20	96	10,400	1.19	141	0.15	Mixed phase
PSMA$_9$-PTFEMA$_{80}$	20	97	10,600	1.22	128	0.16	Mixed phase
PSMA$_9$-PTFEMA$_{100}$	20	97	11,800	1.24	98	0.07	Mixed phase
PSMA$_9$-PTFEMA$_{110}$	20	97	12,200	1.31	309	0.41	Mixed phase
PSMA$_9$-PTFEMA$_{120}$	20	95	13,800	1.27	110	0.07	Vesicles
PSMA$_9$-PTFEMA$_{140}$	20	95	15,200	1.31	136	0.08	Vesicles
PSMA$_9$-PTFEMA$_{160}$	20	97	17,700	1.23	178	0.14	Vesicles
PSMA$_9$-PTFEMA$_{180}$	20	97	18,300	1.31	186	0.12	Vesicles
PSMA$_9$-PTFEMA$_{200}$	20	97	19,600	1.25	206	0.13	Vesicles
PSMA$_9$-PTFEMA$_{250}$	20	97	24,600	1.24	222	0.19	Vesicles
PSMA$_9$-PTFEMA$_{300}$	20	97	27,000	1.31	216	0.06	Vesicles
PSMA$_9$-PTFEMA$_{20}$	25	95	4,500	1.19	-	-	No particles
PSMA$_9$-PTFEMA$_{40}$	25	96	6,800	1.20	18	0.11	Spheres
PSMA$_9$-PTFEMA$_{50}$	25	97	6,900	1.34	1170	0.53	Worms
PSMA$_9$-PTFEMA$_{60}$	25	97	7,300	1.26	58	0.19	Worms
PSMA$_9$-PTFEMA$_{65}$	25	95	8,900	1.23	403	0.47	Worms
PSMA$_9$-PTFEMA$_{70}$	25	95	9,600	1.25	298	0.23	Mixed phase
PSMA$_9$-PTFEMA$_{75}$	25	95	10,400	1.21	158	0.21	Mixed phase
PSMA$_9$-PTFEMA$_{80}$	25	97	10,200	1.20	345	0.23	Mixed phase
PSMA$_9$-PTFEMA$_{100}$	25	97	11,200	1.21	115	0.07	Mixed phase
PSMA$_9$-PTFEMA$_{120}$	25	95	14,600	1.31	245	0.46	Mixed phase
PSMA$_9$-PTFEMA$_{130}$	25	98	15,700	1.20	756	0.41	Mixed phase
PSMA$_9$-PTFEMA$_{140}$	25	95	16,200	1.19	185	0.13	Vesicles
PSMA$_9$-PTFEMA$_{160}$	25	96	17,000	1.26	192	0.28	Vesicles
PSMA$_9$-PTFEMA$_{180}$	25	96	18,300	1.17	186	0.11	Vesicles
PSMA$_9$-PTFEMA$_{200}$	25	96	20,300	1.21	194	0.28	Vesicles
PSMA$_9$-PTFEMA$_{250}$	25	98	24,200	1.25	214	0.13	Vesicles
PSMA$_9$-PTFEMA$_{300}$	25	98	26,500	1.31	237	0.10	Vesicles

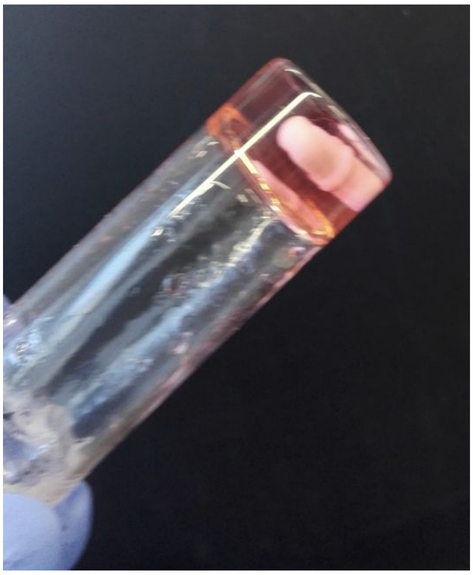

Figure S3. Digital photograph recorded for a 20% w/w dispersion of PSMA$_9$–PTFEMA$_{63}$ worms prepared *via* RAFT dispersion polymerization of TFEMA in *n*-dodecane at 90 °C. A highly transparent, free-standing gel is obtained for this PISA formulation.

Table S3. Summary of structural parameters obtained from fitting small-angle X-ray scattering patterns recorded for a series of PSMA$_9$-PTFEMA$_x$ nano-objects using either a spherical micelle,[1] worm-like micelle[1], or vesicle model.[2] D_{sphere} is the overall sphere diameter such that $D_{\mathrm{sphere}} = 2R_s + 4R_g$, where R_s is the mean core radius and R_g is the radius of gyration of the stabilizer chains. T_{worm} is the overall worm thickness ($T_{\mathrm{worm}} = 2R_{\mathrm{wc}} + 4R_g$, where R_{wc} is the mean worm core radius) and L_{worm} is the mean worm contour length. D_{vesicle} is the overall vesicle diameter ($D_{\mathrm{vesicle}} = 2R_m + T_{\mathrm{membrane}} + 4R_g$, where R_m is the distance from the center of the vesicle to the center of the vesicle membrane, and T_{membrane} is the vesicle membrane thickness). N_{agg} is the mean aggregation number (*i.e.*, the mean number of copolymer chains per nano-object).

Block copolymer	**Nanoparticle Morphology**	D_{sphere} **(nm)**	T_{worm} **(nm)**	L_{worm} **(nm)**	D_{vesicle} **(nm)**	T_{membrane} **(nm)**	N_{agg}
PSMA$_9$-PTFEMA$_{38}$	Spheres	14.6 ± 1.7	-	-	-	-	110
PSMA$_9$-PTFEMA$_{49}$	Worms	-	15.8 ± 2.0	905	-	-	13,700
PSMA$_9$-PTFEMA$_{63}$	Worms	-	16.4 ± 2.1	1040	-	-	13,400
PSMA$_9$-PTFEMA$_{194}$	Vesicles	-	-	-	195 ± 66	20.6 ± 4.4	50,700
PSMA$_9$-PTFEMA$_{291}$	Vesicles	-	-	-	190 ± 48	28.8 ± 4.4	41,100

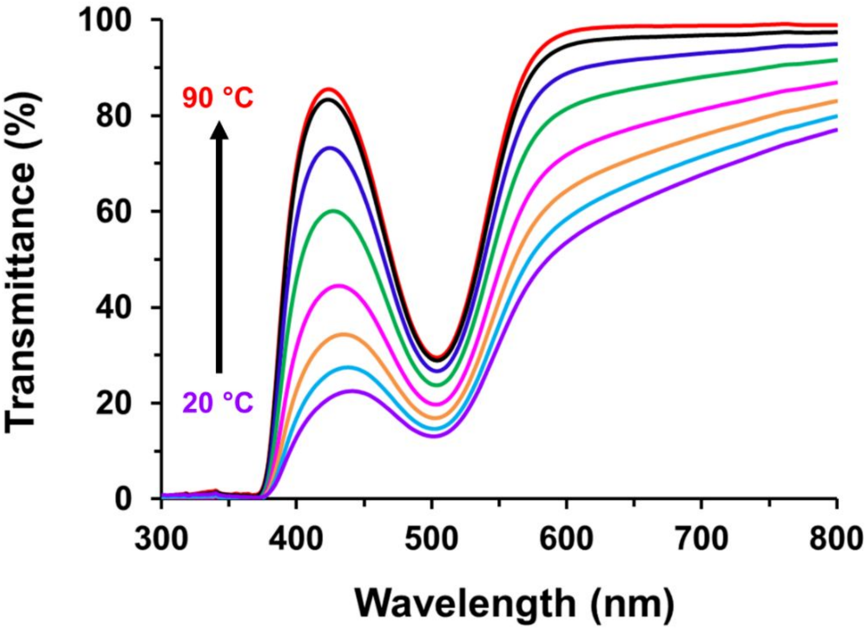

Figure S4. Transmittance *vs*. wavelength plots recorded between 20 and 90 °C for 25% w/w dispersions of $PSMA_9$-$PTFEMA_{294}$ vesicles in *n*-dodecane.

Table S4. Summary of the GPC and DLS data obtained for $PSMA_{16}$-$PTFEMA_{86}$ spherical nanoparticles prepared at 15% w/w solids in *n*-hexadecane during the *in situ* visible absorption spectroscopy study and the kinetic experiments conducted using ^{19}F NMR spectroscopy.

	Target Composition	**Solids content (% w/w)**	**M_n (kg mol^{-1})**	**M_w/M_n**	**DLS diameter (nm)**	**PDI**
Precursor block	$PSMA_{16}$	-	5.5	1.16	-	-
In situ visible absorption spectroscopy	$PSMA_{16}$ – $PTFEMA_{86}$ spheres	15	17.7	1.13	26	0.05
Kinetics by ^{19}F NMR spectroscopy	$PSMA_{16}$ – $PTFEMA_{86}$ spheres	15	17.1	1.17	25	0.02

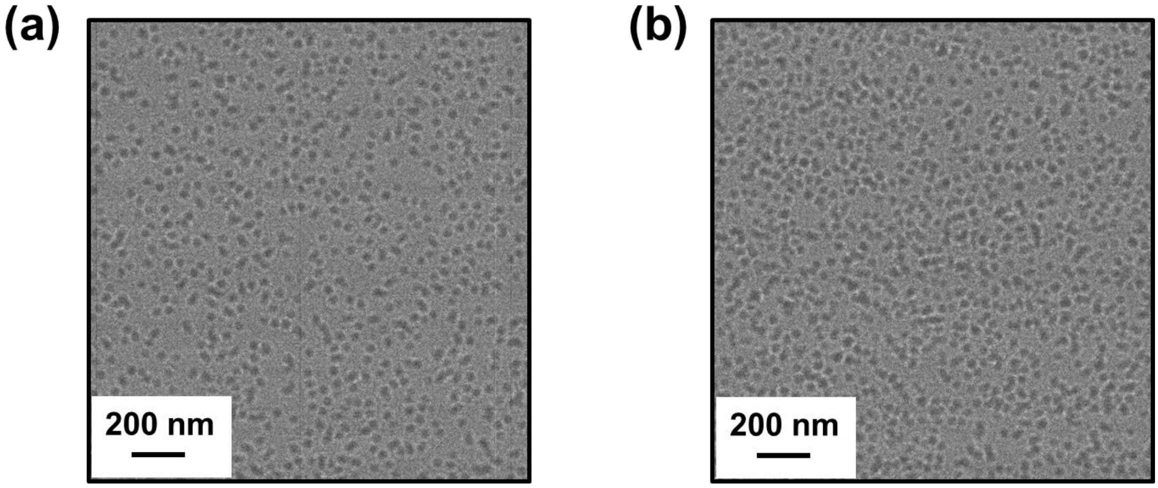

Figure S5. Representative TEM images recorded for $PSMA_{16}$–$PTFEMA_{86}$ spherical nanoparticles prepared at 15% w/w solids in *n*-hexadecane during (a) the *in situ* visible absorption spectroscopy study and (b) the kinetic experiments conducted using ^{19}F NMR spectroscopy. The very similar particle size indicated by these two images is consistent with the DLS data reported in **Table S4**.

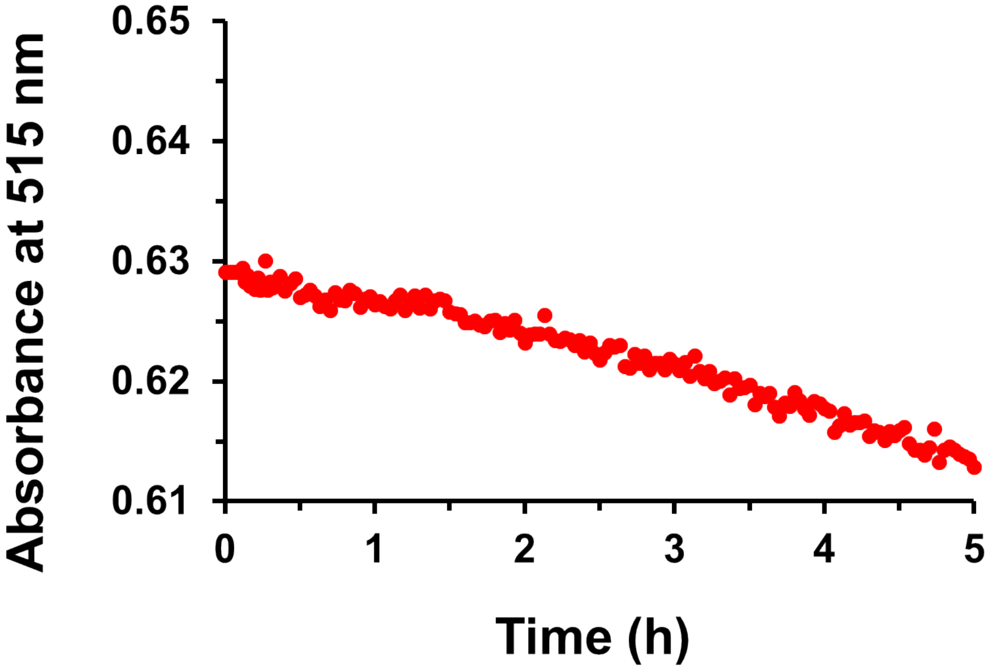

Figure S6. Absorbance *vs*. time curve observed for the $PSMA_{16}$ stabilizer block dissolved in *n*-hexadecane ($[PSMA_{16}]$ = 6.2 mmol dm^{-3} = 35 g dm^{-3}) on heating for 5 h at 90 °C. The monotonic reduction in absorbance under such conditions indicates the gradual loss of dithiobenzoate RAFT end-groups even in the absence of any free radical initiator.

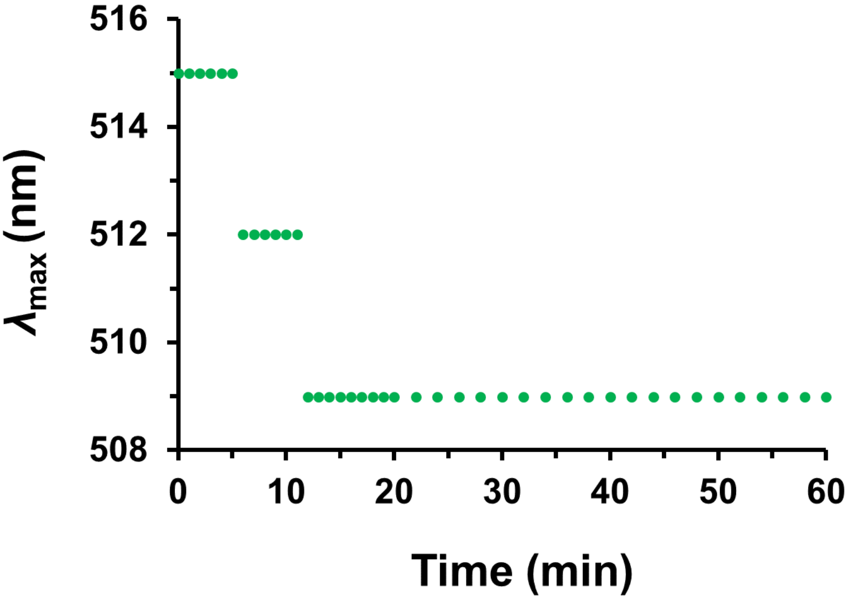

Figure S7. Systematic blue shift in λ_{max} observed for the relatively weak $n \rightarrow \pi^*$ transition of the dithiobenzoate end-group during the synthesis of PSMA$_{16}$-PTFEMA$_{86}$ spherical nanoparticles *via* RAFT dispersion polymerization of TFEMA at 15% w/w solids in *n*-hexadecane at 90 °C. Spectra were recorded between 200 and 800 nm at a resolution of ± 3 nm.

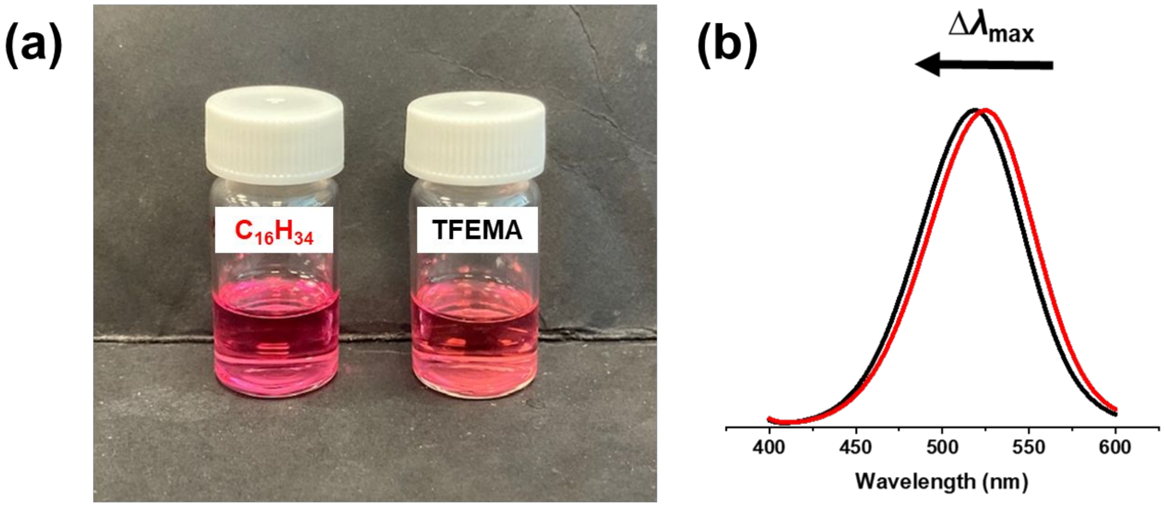

Figure S8. (a) Digital photograph recorded at 25 °C for an 8 mmol dm^{-3} solution of CPDB dissolved in either *n*-hexadecane (λ_{max} = 524 nm) or TFEMA monomer (λ_{max} = 518 nm), respectively. The subtle blue shift in the latter solvent is discernible to the naked eye. (b) Visible absorption spectra recorded at 25 °C for the same two 8 mmol dm^{-3} solutions of CPDB dissolved in either *n*-hexadecane (red data; λ_{max} = 524 nm) or TFEMA monomer (black data; λ_{max} = 518 nm), respectively.

Table S5. Summary of the $\lambda_{\max}$ data obtained at 90 °C for the PSMA$_{16}$ precursor block dissolved in either *n*-hexadecane or TFEMA, and the PSMA$_{16}$-PTFEMA$_9$ and the PSMA$_{16}$-PTFEMA$_{19}$ chains prepared at 15% w/w solids in *n*-hexadecane.

Composition	**Solvent**	**Solids content (% w/w)**	***T* (°C)**	**$\lambda_{\max}$ (nm)**
PSMA$_{16}$	*n*-hexadecane	-	90	516
PSMA$_{16}$	TFEMA	-	90	510
PSMA$_{16}$ – PTFEMA$_9$ chains	*n*-hexadecane	15	90	510
PSMA$_{16}$ – PTFEMA$_{19}$ chains	*n*-hexadecane	15	90	509

SAXS Models Used in this Study

Programming tools within the Irena SAS Igor Pro macros[3] were used to implement the scattering models.

In general, the intensity of X-rays scattered by a dispersion of nano-objects [as represented by the scattering cross-section per unit sample volume, $\frac{d\Sigma}{d\Omega}(q)$] can be expressed as:

$$\frac{d\Sigma}{d\Omega}(q) = NS(q)\int_0^\infty \dots \int_0^\infty F(q,r_1,\dots,r_k)^2\Psi(r_1,\dots,r_k)dr_1,\dots,dr_k \qquad \text{(S1)}$$

where $F(q,r_1,\dots,r_k)$ is the form factor, $r_1,\dots,r_k$ is a set of k parameters describing the structural morphology, $\Psi(r_1,\dots,r_k)$ is the distribution function, $S(q)$ is the structure factor and N is the number density of nano-objects per unit volume expressed as:

$$N = \frac{\varphi}{\int_0^\infty \dots \int_0^\infty V(r_1,\dots,r_k)\Psi(r_1,\dots,r_k)dr_1,\dots,dr_k} \qquad \text{(S2)}$$

where $V(r_1,\dots,r_k)$ is the volume of the nano-object and φ is its volume fraction within the dispersion. It is assumed that $S(q) = 1$ at the sufficiently low copolymer concentrations used in this study (1.0% w/w).

Spherical micelle model

The spherical micelle form factor for Equation S1 is given by[1]:

$$F_{s_{mic}}(q) = {N_s}^2{\beta_s}^2{A_s}^2(q,R_s) + N_s{\beta_c}^2F_c(q,R_g) + N_s(N_s - 1){\beta_c}^2{A_c}^2(q) + 2{N_s}^2\beta A_c(q) \tag{S3}$$

where R_s is the volume-average sphere core radius and R_g is the radius of gyration of the coronal steric stabilizer block (in this case, PSMA$_9$). The X-ray scattering length contrasts for the core and corona blocks are given by $\beta_s = V_s(\xi_s - \xi_{sol})$ and $\beta_c = V_c(\xi_c - \xi_{sol})$ respectively. Here, ξ_s, ξ_c and ξ_{sol} are the X-ray scattering length densities of the core block (ξ_{PTFEMA}= 12.76 x 10^{10} cm^{-2}), corona block (ξ_{PSMA}= 9.24 x 10^{10} cm^{-2}) and *n*-dodecane solvent (ξ_{sol}= 7.32 x 10^{10} cm^{-2}), respectively. V_s and V_c are the volumes of the core block (V_{PTFEMA}) and the corona block (V_{PSMA}), respectively. The sphere form factor amplitude is used for the amplitude of the core self-term:

$$A_c(q,R_s) = \Phi(qR_s)\exp\left(-\frac{q^2\sigma^2}{2}\right) \tag{S4}$$

where $\Phi(qR_s) = \frac{3[\sin(qR_s) - qR_s\cos(qR_s)]}{(qR_s)^3}$. A sigmoidal interface between the two blocks was assumed for the spherical micelle form factor (Equation S3). This is described by the exponent term with a width σ accounting for a decaying scattering length density at the micellar interface. This σ value was fixed at 2.2 during fitting.

The form factor amplitude of the spherical micelle corona is:

$$A_c(q) = \frac{\int_{R_s}^{R_s + 2s}\mu_c(r)\frac{\sin(qr)}{qr}r^2dr}{\int_{R_s}^{R_s + 2s}\mu_c(r)r^2dr}exp\left(-\frac{q^2\sigma^2}{2}\right) \tag{S5}$$

The radial profile, $\mu_c(r)$, can be expressed by a linear combination of two cubic b splines, with two fitting parameters s and a corresponding to the width of the profile and the weight coefficient respectively. This information can be found elsewhere,[4,5] as can the approximate integrated form of Equation S5. The self-correlation term for the coronal block is given by the Debye function:

$$F_c(q,R_g) = \frac{2\left[\exp\left(-q^2{R_g}^2\right) - 1 + q^2{R_g}^2\right]}{q^4{R_g}^4} \tag{S6}$$

where R_g is the radius of gyration of the PSMA coronal block. The aggregation number, N_s, of the spherical micelle is given by:

$$N_s = (1 - x_{sol})\frac{\frac{4}{3}\pi{R_s}^3}{V_s} \tag{S7}$$

where x_{sol} is the volume fraction of solvent within the PTFEMA micelle cores, which was found to be zero in all cases. A polydispersity for one parameter (R_s) is assumed for the micelle model, which is described by a Gaussian distribution. Thus, the polydispersity function in Equation S1 can be represented as:

$$\Psi(r_1) = \frac{1}{\sqrt{2\pi\sigma_{Rs}{}^2}} exp\left(- \frac{(r_1 - R_s)^2}{2\sigma_{Rs}{}^2} \right) \tag{S8}$$

where σ_{Rs} is the standard deviation for R_s. In accordance with Equation S2, the number density per unit volume for the micelle model is expressed as:

$$N = \frac{\varphi}{\int_0^\infty V(r_1)\Psi(r_1)dr_1} \tag{S9}$$

where φ is the total volume fraction of copolymer in the spherical micelles and $V(r_1)$ is the total volume of copolymer within a spherical micelle $[V(r_1) = (V_s + V_c)N_s(r_1)]$.

Worm-like micelle model

The worm-like micelle form factor for Equation S1 is given by:

$$F_{w\_mic}(q) = N_w{}^2\beta_s{}^2F_{sw}(q) + N_w\beta_c{}^2F_c(q,R_g) + N_w(N_w - 1)\beta_c{}^2S_{cc}(q) + 2N_w{}^2\beta_s \tag{S10}$$

where all the parameters are the same as those described in the spherical micelle model (Equation S3), unless stated otherwise.

The self-correlation term for the worm core cross-sectional volume-average radius R_w is:

$$F_{sw}(q) = F_{worm}(q,L_w,b_w)A_{CSworm}{}^2(q,R_{sw}) \tag{S11}$$

where

$$A_{CSworm}{}^2(q,R_{sw}) = \left[2\frac{J_1(qR_{sw})}{qR_{sw}}\right]^2 \tag{S12}$$

and J_1 is the first-order Bessel function of the first kind, and a form factor $F_{worm}(q,L_w,b_w)$ for self-avoiding semi-flexible chains represents the worm-like micelles, where b_w is the Kuhn length and L_w is the mean contour length. A complete expression for the chain form factor can be found elsewhere.[6]

The mean aggregation number of the worm-like micelle, N_w, is given by:

$$N_w = (1 - x_{sol})\frac{\pi R_{sw}{}^2L_w}{V_s} \tag{S13}$$

where x_{sol} is the volume fraction of solvent within the worm-like micelle cores, which was found to be zero in all cases. The possible presence of semi-spherical caps at both ends of each worm is neglected in this form factor.

A polydispersity for one parameter (R_{w}) is assumed for the micelle model, which is described by a Gaussian distribution. Thus, the polydispersity function in Equation S1 can be represented as:

$$\Psi(r_1) = \frac{1}{\sqrt{2\pi\sigma_{R_{\mathrm{w}}}{}^2}} exp\left(-\frac{(r_1 - R_{\mathrm{w}})^2}{2\sigma_{R_{\mathrm{w}}}{}^2} \right) \tag{S14}$$

where $\sigma_{R_{\mathrm{w}}}$ is the standard deviation for R_{w}. In accordance with Equation S2, the number density per unit volume for the worm-like micelle model is expressed as:

$$N = \frac{\varphi}{\int_0^{\infty} V(r_1)\Psi(r_1)dr_1} \tag{S15}$$

where φ is the total volume fraction of copolymer in the worm-like micelles and $V(r_1)$ is the total volume of copolymer in a worm-like micelle $[V(r_1) = (V_{\mathrm{s}} + V_{\mathrm{c}})N_{\mathrm{w}}(r_1)]$.

Vesicle model

The vesicle form factor in Equation S1 is expressed as:[2]

$$F_{ves}(q) = N_v{}^2\beta_m{}^2A_m{}^2(q) + N_v\beta_{vc}{}^2F_c(q,R_g) + N_v(N_v - 1)\beta_{vc}{}^2A_{vc}{}^2(q) + 2N \; A_{vc}(q) \tag{S16}$$

where all the parameters are the same as in the spherical micelle model (see Equation S3) unless stated otherwise.

The amplitude of the membrane self-term is:

$$A_m(q) = \frac{V_{out}\varphi(qR_{out}) - V_{in}\varphi(qR_{in})}{V_{out} - V_{in}} exp\left(-\frac{q^2\sigma_{in}{}^2}{2} \right) \tag{S17}$$

where $R_{in} = R_m - \frac{1}{2}T_m$ is the inner radius of the membrane, $R_{out} = R_m + \frac{1}{2}T_m$ is the outer radius of the membrane (R_{m} is the radius from the centre of the vesicle to the centre of the membrane), $V_{in} = \frac{4}{3}\pi R_{in}{}^3$ and $V_{out} = \frac{4}{3}\pi R_{out}{}^3$. It should be noted that Equation S16 differs subtly from the original work in which it was first described.[2] The exponent term in Equation S17 represents a sigmoidal interface between the blocks, with a width σ_{in} accounting for a decaying scattering length density at the membrane surface. The value of σ_{in} was fixed at 2.5 during fitting. The mean vesicle aggregation number, N_v, is given by:

$$N_v = (1 - x_{sol})\frac{V_{out} - V_{in}}{V_m} \tag{S18}$$

where x_{sol} is the volume fraction of solvent within the vesicle membrane, which was found to be zero in all cases. Assuming that there is no penetration of the solvophilic coronal blocks into the solvophobic membrane, the amplitude of the vesicle corona self-term is expressed as:

$$A_{vc}(q) = \Psi(qR_g)\frac{1}{2}\left[\frac{sin[q(R_{out} + R_g)]}{q(R_{out} + R_g)} + \frac{\sin[q(R_{in} - R_g)]}{q(R_{in} - R_g)}\right] \tag{S19}$$

where the term outside the square brackets is the factor amplitude of the corona block polymer chain such that:

$$\Psi(qR_g) = \frac{1 - \exp(-qR_g)}{(qR_g)^2} \tag{S20}$$

For the vesicle model, it was assumed that two parameters are polydisperse: the radius from the centre of the vesicles to the centre of the membrane and the membrane thickness (denoted R_m and T_m, respectively). Each parameter is considered to have a Gaussian distribution of values, so the polydispersity function in Equation S1 can be expressed in each case as:

$$\Psi(r_1 r_2) = \frac{1}{\sqrt{2\pi\sigma_{Rs}{}^2}}exp\left(-\frac{(r_1 - R_m)^2}{2\sigma_{Rm}{}^2}\right)\frac{1}{\sqrt{2\pi\sigma_{Tm}{}^2}}exp\left(-\frac{(r_1 - T_m)^2}{2\sigma_{Tm}{}^2}\right) \tag{S21}$$

where σ_{Rm} and σ_{Tm} are the standard deviations for R_m and T_m, respectively. Following Equation S2, the number density per unit volume for the vesicle model is expressed as:

$$N = \frac{\varphi}{\int_0^\infty \int_0^\infty V(r_1,r_2)\Psi(r_1,r_2)dr_1 dr_2} \tag{S22}$$

where φ is the total volume fraction of copolymer in the vesicles and $V(r_1,r_2)$ is the total volume of copolymers in a vesicle $[V(r_1,r_2) = (V_m + V_{vc})N_v(r_1,r_2)]$.

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