Supporting information

![Graph A](image1.png)

**Fig. 1A)** Elution profiles of different species corresponding to separation using Sephadex G-25 size exclusion column with dimensions of DxL=1x30 cm. The empty BE1 polymersomes (□) elution profile has been determined by DLS and given in kHz and BE1 polymersomes containing $^{111}$In (●), pure $^{111}$In-DTPA (●) and pure $^{111}$In-tropolone (▲) have been measured with a High Purity Germanium detector. The normalized fraction corresponds to the ratio of radioactivity in each volume fraction and the total radioactivity before separation. N.B. 95% of all $^{111}$In-tropolone does not come off the column.

**B)** Elution profiles of $^{111}$In-DTPA and empty polymersomes corresponding to separation using Sephadex G-25 size exclusion
column with dimensions of DxL=1x21 cm. The empty BE1 polymersomes (○) elution profile has been determined by DLS and is given in kHz and pure $^{111}$In-DTPA (●) has been measured with a High Purity Germanium detector. The normalized fraction corresponds to the ratio of radioactivity in each volume fraction and the total radioactivity before separation.

![Loading efficiency vs time graph](image)

**Fig. 2** Effect of the loading time on the loading efficiency of BE1 polymersomes (triangles), BE3 polymersomes (circles) and BE4 polymersomes (squares). The concentration of polymersomes is 0.5 mg/ml, the $^{111}$In$^{3+}$ activity is 0.15 MBq and the tropolone and DTPA concentration are respectively 20 µM and 1 mM.

Calculations encapsulated DTPA amount:

For BE1 polymersomes, D=100 nm

a) Determining the average internal volume per vesicle

The average internal volume per vesicles, $V_{\text{int}}$, is calculated using the diameter of the polymersomes and the thickness of membrane. The membrane thickness $W=7$ nm has
been determined by Cryo-EM. The diameter of polymersomes has been set at 100 nm according to the size measured by Cryo-EM.

\[ V_{int} = \frac{4}{3} \pi \left[ (D_i / 2) - W \right]^3 = \frac{4}{3} \pi \left[ (100 / 2) - 7 \right]^3 = 3.3 \times 10^5 \text{ nm}^3 \]

b) Determining the number of vesicles per gram of polymer

For the block copolymer poly(butadiene-b-ethylene oxide) (PB-PEO) used in this study, the PB content by weight is 66.7%. The weight of PB per gram of polymer is, therefore, equal to ca. 0.667 g/g. The volume of PB per vesicle is calculated using the equation below:

\[ V_{iPB} = \frac{4}{3} \pi \left[ (D_i / 2) - (D_i / 2) - W \right]^3 = \frac{4}{3} \pi \left[ ((100 / 2) - (100 / 2) - 7) \right]^3 = 1.9 \times 10^5 \text{ nm}^3 \]

PB weight of one polymersomes is:

\[ W_{PB} = V_{iPB} \times \rho = 1.9 \times 10^5 \text{ nm}^3 \times 0.93 \text{ g/mL} = 1.77 \times 10^{-16} \text{ g} \]

Assuming that the density of PB equals 0.93 g/mL, the concentration of polymersomes is 0.5 mg/mL. The number of vesicles per milliliter of solution can be expressed as:

\[ N = \frac{\text{Weight of PB}}{\text{Average weight of PB per polymersomes}} = \frac{0.667 \times 0.5 \text{ mg}}{1.77 \times 10^{-16} \text{ g}} = 1.88 \times 10^{12} \]

c) 1 ml DTPA is equal to \(10^{-6}\) mol when 1 mM is used

Number of DTPA:
\[ N_{\text{DTPA}} = 10^{-6} \times 6.02 \times 10^{23} = 6.02 \times 10^{17} \text{ per } 1 \text{ mL} = 6.02 \times 10^{-4} \text{ per nm}^3 \]

One DTPA occupies volume of:

\[ V_{\text{DTPA}} = \frac{1}{N} = 1661 \text{ nm}^3 \]

Total Number of DTPA is:

\[ N_{\text{total DTPA}} = N_{\text{polymersomes}} \times V_{\text{internal}} \times V_{\text{DTPA}} = 1.88 \times 10^{12} \times 3.3 \times 10^5 \times 6.02 \times 10^{-4} = 3.73 \times 10^{14} \]

d) 37 MBq Indium is equal to \(2.15 \times 10^{-11}\) mol.

The maximum loading capacity is then:

\[ A = \frac{N_{\text{DTPA}} \times 37 \text{ MBq}}{N_{\text{37 MBq}}} = \frac{3.73 \times 10^{14}}{1.2 \times 10^{13}} \times 37 \text{ MBq} = 1151 \text{ MBq} \]

For BE2 polymersomes, 100 nm,

a) Determining the average internal volume per vesicle

The average internal volume per vesicles, \(V_{\text{int}}\), is calculated using the diameter of the polymersomes and the thickness of membrane. The membrane thickness has been set at \(W=13\) nm measured by Cryo-EM. The diameter of polymersomes (D) has been set at 100 nm according to the Cryo-EM measurements.

\[ V_{\text{int}} = \frac{4}{3} \pi \left( \frac{D}{2} - W \right)^3 = \frac{4}{3} \pi \left( \frac{100}{2} - 13 \right)^3 = 2.1 \times 10^5 \text{ nm}^3 \]

b) Determining the number of vesicles per gram of polymer
For the block copolymer poly(butadiene-b-ethylene oxide) (PB-PEO), the PB content by weight is 62.5%. The weight of PB per gram of polymer is, therefore, equal to ca. 0.625 g/g. The volume of PB per vesicle is calculated using equation below:

\[ V_{PB}^{PB} = \frac{4}{3} \pi [(D_i / 2)^3 - (D / 2 - W)] = \frac{4}{3} \pi [(100 / 2)^3 - ((100 / 2) - 13)^3] = 3.11 \times 10^5 \text{ nm}^3 \]

PB weight of one polymersomes is:

\[ W_{PB} = V_{PB}^{PB} \times \rho = 3.11 \times 10^5 \times 0.93 \text{ g/mL} = 2.89 \times 10^{-16} \text{ g} \]

Assuming that the density of PB is equal 0.93 g/ml and the concentration of polymersomes is 0.5 mg/ml. The number of vesicles per milliliter of solution can be expressed as:

\[ N = \frac{\text{Weight of PB}}{\text{Average weight of PB per polymersomes}} = \frac{0.625 \times 0.5 \text{ mg}}{2.89 \times 10^{-16} \text{ g}} = 1.1 \times 10^{12} \]

c) 1 ml DTPA is equal to $10^{-6}$ mol when 1 mM is used.

Number of DTPA:

\[ N_{DTPA} = 10^{-6} \times 6.02 \times 10^{23} = 6.02 \times 10^{17} \text{ per } 1 \text{ mL} = 6.02 \times 10^{-4} \text{ per } \text{nm}^3 \]

One DTPA occupy volume is:

\[ V_{DTPA} = \frac{1}{N} = 1661 \text{ nm}^3 \]

Total Number of DTPA is:
\[ N_{total\ DTPA} = N_{polymersomes} \times V_{internal} \times V_{DTPA} = 1.0 \times 10^{12} \times 2.1 \times 10^5 \times 6.02 \times 10^{-4} = 1.26 \times 10^{14} \]

d) 37 MBq Indium is equal to \(2.15 \times 10^{-11}\) mol.

The maximum loading capacity is then:

\[ A = \frac{N_{DTPA}}{N_{37\ MBq}} \times 37 \ MBq = \frac{1.17 \times 10^{14}}{1.29 \times 10^{13}} \times 37 \ MBq = 335 \ MBq \]