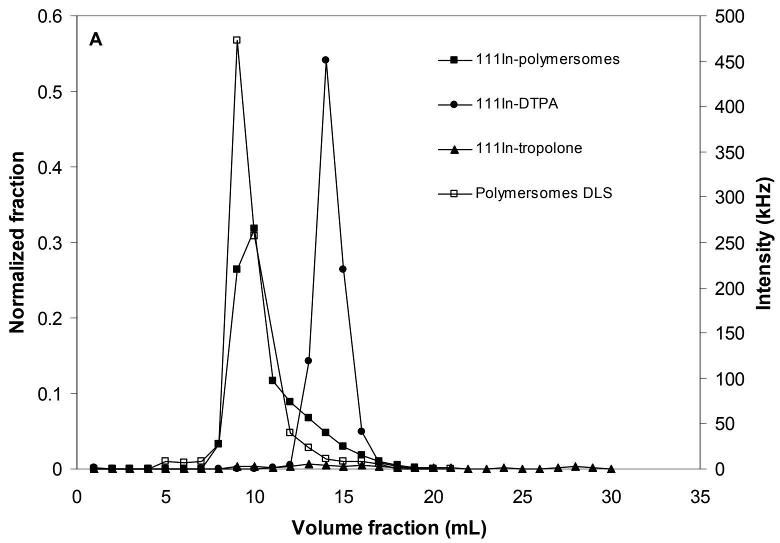
Supporting information



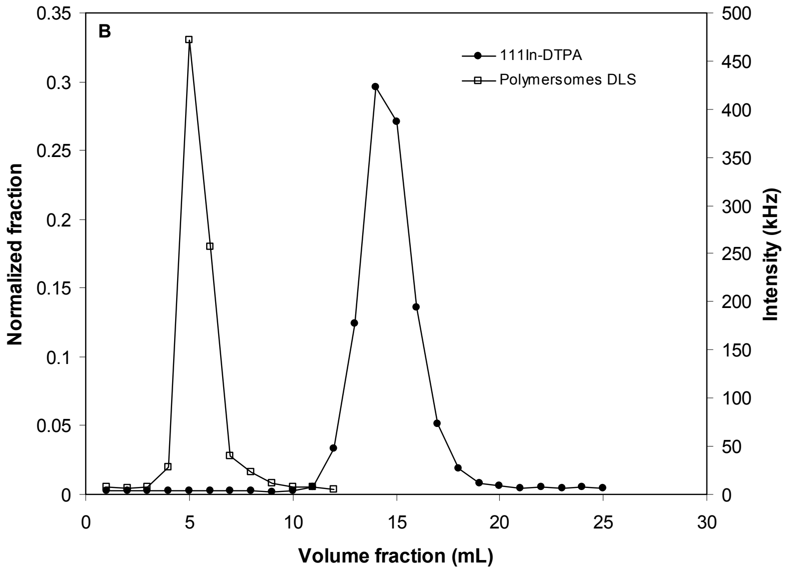


Fig. 1A) *Elution profiles of different species corresponding to separation using Sephadex G-25 size exclusion column with dimensions of DxL=1x30 cm. The empty BE1 polymersomes (□) elution profile has been determined by DLS and given in kHz and BE1 polymersomes containing ^{111}In (■), pure ^{111}In-DTPA (●) and pure ^{111}In-tropolone (▲)have been measured with a High Purity Germanium detecto. 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 detecto. The normalized fraction corresponds to the ratio of radioactivity in each volume fraction and the total radioactivity before separation.*

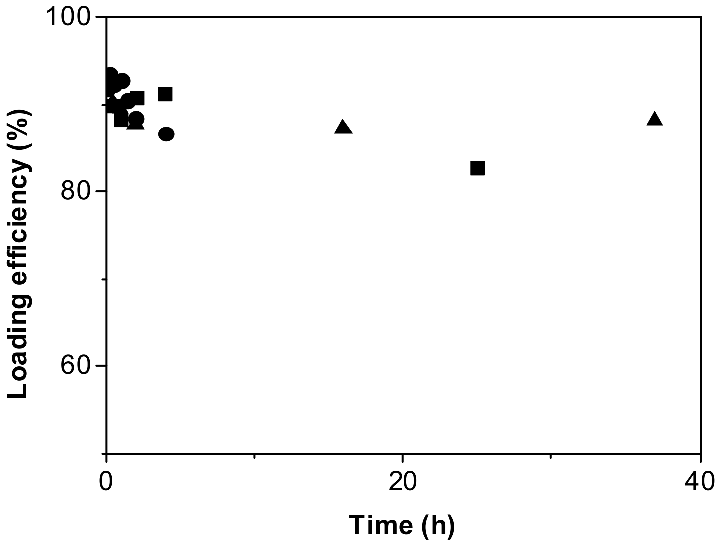


Fig. 2 *Effect of the loading time on the loading efficiency of BE1 polymersomes (triangles), BE3 polymersomes (circles) and BE4 polymersomes (squares). The concentration of polymersomes is 0.5 mg/ml, the $^{111}In^{3+}$ activity is 0.15 MBq and the tropolone and DTPA concentration are respectively 20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

$$V_{\text{int}} = \frac{4}{3}\pi\left[\left(D_i/2\right)-W\right]^3 == \frac{4}{3}\pi\left[\left(100/2\right)-7\right]^3 = 3.3\times10^5\ nm^3$$

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:

$$V_i^{PB} = \frac{4}{3}\pi[(D_i/2)^3 - (D_i/2)-W)^3] = \frac{4}{3}\pi[(100/2)^3 - ((100/2)-7)^3] = 1.9\times10^5\ \text{nm}^3$$

PB weight of one polymersomes is:

$$W_{PB} = V^{PB}\times\rho = 1.9\times10^5 nm^3\times0.93\ g/mL = 1.77\times10^{-16}\ g$$

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:

$$N = \frac{Weight\ of\ PB}{Average\ weight\ of\ PB\ per\ polymersomes} = \frac{0.667\times0.5\ mg}{1.77\times10^{-16}\ g} = 1.88\times10^{12}$$

c) 1 ml DTPA is equal to10^{-6} mol when 1 mM is used

Number of DTPA:

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

One DTPA occupies volume of:

$$V_{DTPA} = \frac{1}{N} = 1661\ nm^3$$

Total Number of DTPA is:

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

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

The maxium loading capacity is then:

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

For BE2 polymersomes, 100 nm,

a) Determining the average internal volume per vesicle

The average internal volume per vesicles, V_{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.

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

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:

$$V_i^{PB} = \frac{4}{3}\pi[(D_i/2)^3 - (D_i/2) - W)^3] = \frac{4}{3}\pi[(100/2)^3 - ((100/2) - 13)^3] = 3.11\times10^5\ \mathrm{nm}^3$$

PB weight of one polymersomes is:

$$W_{PB} = V^{PB} \times \rho = 3.11\times10^5 \times 0.93\, g/mL = 2.89\times10^{-16}\ g$$

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:

$$N = \frac{Weight\ of\ PB}{Average\ weight\ of\ PB\ per\ polymersomes} = \frac{0.625\times0.5\ mg}{2.89\times10^{-16}\ g} = 1.1\times10^{12}$$

c) 1 ml DTPA is equal to10^{-6} mol when 1 mM is used.

Number of DTPA:

$$N_{DTPA} = 10^{-6}\times6.02\times10^{23} = 6.02\times10^{17}\ per\ 1\ mL = 6.02\times10^{-4}\ \mathrm{per\ nm}^3$$

One DTPA occupy volume is:

$$V_{DTPA} = \frac{1}{N} = 1661\ nm^3$$

Total Number of DTPA is:

$$N_{total\ DTPA} = N_{polymersomes} \times V_{internal} \times V_{DTPA} = 1.0\times10^{12} \times 2.1\times10^{5} \times 6.02\times10^{-4} = 1.26\times10^{14}$$

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

The maxium loading capacity is then:

$$A = \frac{N_{DTPA}}{N_{37MBq}} \times 37\ MBq = \frac{1.17\times10^{14}}{1.29\times10^{13}} \times 37\ MBq = 335\ MBq$$