Supplemental Information to generating forces in confinement via polymerization

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Table of Parameters used in Study

The table below lists all physical parameters in the manuscript, as well as the values or ranges we study them.

Constant	Symbol	Values Studied
Temperature	$k_b T$	1
Toy model polymer spring constant	k	20
Toy model polymer bending constant	κ	4-1028
Elastic Shell spring constant	k_s	20-100
Elastic Shell bending constant	b	0.5-32
Monomer size (shell and interior)	σ	1
Translational Diffusion coefficient	D	$1/3\pi$
Rotational Diffusion coefficient	D_R	$1/\pi$
Patchy particle attraction	ϵ	20-100
Patchy particle angle	$ heta_m$	0.5-0.8
Rate of Monomer release	R	$50/\tau - 5000/\tau$

Table 1:	Parameter	Ranges	studied	in	manuscript
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Deformations of empty shells



Figure S1: Response of the elastic shell to applied force on two monomers on opposite ends of the shell. We apply a force of the given magnitude to each monomer, pushing in opposite directions. We measure the axial ratios under different strengths of the pulling force, across different elastic parameters for the shell. Increasing the stretching and bending moduli of the shell increases its resistence to pulling (higher effective spring constants)

Bundling transitions in toy models

We plot the bundling transition in the toy model for different values of the shell bending potential.



Figure S2: The bundling parameter (defined in the main text) for the toy model of polymers growing in shells with $k_s = 20$ and different bending moduli, or in a completely hard shell. In the hard shell, there is no bundling transition.

Persistence length of free polymers

The main text used polymers with different microscopic parameters for the binding energy and binding angles of our monomeric subunits. Below, we present what the persistence lengths of polymers in free space are for different choices of microscopic parameters δE and θ_m (the binding energy and angular aperture of the binding site, respectively). The persistence length is calculated as the correlation distance of tangent vectors to the polymer:

$$\langle \cos(\theta) \rangle = e^{-L/P} \tag{1}$$

where L is the distance along the contour of the chain and P is the persistence length.

We perform these simulations by beginning with all the monomers in free space and letting them grow naturally, then measuring the average persistence length of the resultant polymers after equilibration. The persistence length is presented in Figure S3.



Figure S3: The persistence lengths of polymers with different microscopic parameters, measured in terms of the hard sphere diameter. The persistence length increases with increasing binding energy, and decreases with greater patch size.

As can be observed, the persistence lengths of all the polymers in free space are larger than the sizes of the vesicles we are embedding them in, leading to the polymers exerting elastic forces on the vesicle. The presence of the vesicle will affect measured persistence lengths, by forcing the polymers to bend, but at an energetic penalty. Phase diagrams of self assembly polymer models against microscopic shell and monomer parameters



Figure S4: The deformation score for the self assembling polymer system at the given time when the shell parameters are varied for $\epsilon = 100, \theta_m = 0.6$. As with the toy model, the weaker shells have more axial deformation.



Figure S5: The deformation score at a given time when the monomer parameters are varied. For $k_s = 20, b = 4, R\tau = 50$. Stiffer polymers always do better, despite being more difficult to assemble.

Numbers of Monomers inside the Shell over time



Figure S6: (Top) number of monomers in the interior of shell over time for toy model (top) and the patchy particle model (bottom). (Top) Growth in the number of monomers over time for a strong polymer $k_s = 20, b = 32, \kappa = 1024$ and a weak polymer $k_s = 20, b = 1/2, \kappa = 4$. The in the case of the weak shell, the polymers in the toy model. Due to how the model is defined, fluctuations of the shell suppress further growth leading to saturation effects on the number of monomers in the strong polymers. As the weak polymers can wrap around the interior their growth is not suppressed. (Bottom) Monomers over time for patchy particle model (corresponding to main text??C). Monomers are released into the interior at some predefined rate R so the total number of monomers grows linearly with some rate. Suppression effects are less pronounced in such a model as the monomers are introduced into the interior before making their way to the growing tips rather than being added to the end directly where steric hindrance can reduce the probability of monomer addition. We limit the total amount of monomers that can enter the shell to 2000 so the shell doesn't become overfull with monomers in an unrealistic manner.

Additional results for different multivalent schemes



Figure S7: The average deformation scores against monomer numbers as the total number of crosslinkers are varied for a variant of main text scheme 4D) where each monomer has two potential crosslinking sites on the surface as indicated. The results are not qualitatively different to the main text scheme. $k_s = 20, b = 1, R\tau = 50, \epsilon = 50, \theta_m = 0.8$



Figure S8: The average deformation scores against monomer numbers as the total number of crosslinkers are varied for a variant of main text scheme 4D) where crosslinkers have the ability to bind to one another. One can still observe deformation, but the relative proportion of crosslinkers to achieve optimal performance is modified. $k_s = 20, b = 1, R\tau = 50, \epsilon = 50, \theta_m = 0.8$

Assembly under steric hindrance

In the main text, we introduced a model of assembly where monomers are added to the interior of the shell. One could imagine an alternative scheme where all the monomers are already present in the shell and are "activated" (i.e., the binding sites are turned on) at some rate R. While we do not expect this model to be too different to our main model, potential steric interactions with unactivated monomers could lead to potential differences in results. We show here that this model variant still produces axial deformation.



Figure S9: Model variant where the monomers are already present in the shell, but need to be activated at some rate R. We still observe axial deformation in this case, the steric hindrance of the already present monomers does not modify the ability of the polymerization mechanism to generate force. $k_s = 20, b = 4, R\tau = 50, \epsilon = 100, \theta_m = 0.6$