Supporting Information

Self-assembled morphologies of polyelectrolyte-grafted nanoparticles
directed by opposite charged polymer matrices

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S1. Model of PE grafted nanoparticles and matrix PE chains

In order to demonstrate our simulation model clearly, the polymer grafted nanoparticles and matrix PE chain are plotted, which is shown in Fig. S1.

![Schematic for our simulation system](image)

**Fig. S1**  Schematic for our simulation system. For clarity, only two PE grafted nanoparticles and some matrix chains are provided.

S2. The autocorrelation functions of radius of gyration

The autocorrelation functions of radius of gyration for all systems are calculated, which are plotted in Fig. S2. As shown in Fig. S2, the autocorrelation functions for the highest $\lambda_B = 20.0$ (left panels) take longer to equilibrate.
**Fig. S2** Autocorrelation functions of radius of gyration of the grafted chains with different matrix chain lengths. The upper panels (a ~ c) are for $\lambda_B = 3.0$, the middle panels (d ~ f) are for $\lambda_B = 10.0$, and the upper panels (g ~ i) are for $\lambda_B = 20.0$. The panels arranged vertically correspond to the same matrix chain rigidity, from left to right, (a, d, g) $k_\theta = 0$, (b, e, h) $k_\theta = 5.0$, (c, f, i) $k_\theta = 100.0$.

**S3. The density distributions of counterions (matrix monomers at $N_m = 1$)**

The density distributions of counterions along the radial direction of nanoparticles are plotted in Fig. S3. The vertical lines are employed to display the thickness ($H$) of the grafted nanoparticles. It is observed that the density of counterions $\rho_c(r) > 0$ when $r < H$, which indicates that part of counterions are confined in the brush layer.
Fig. S3 The density distributions of counterions along the radial direction of nanoparticles at $N_m = 1$. The vertical lines are employed to display the thickness of the grafted nanoparticles, and the color of vertical lines are the same as those of density distribution profiles.

S4. Supplementary snapshots at $N_m = 48$

To understand why the $R_g$ for $N_m = 48$ is lowest for $\lambda_B = 20$ at $k_0 = 0$ (see Fig. 5a) and it is the highest for other $k_0$ values (see Fig. 5b and 5c), we plotted the snapshots of one PE grafted nanoparticle and matrix chains for $N_m = 48$ for different $\lambda_B$ values, which are shown in Fig. S4.

**Fig. S4.** Snapshots of one polyelectrolyte grafted nanoparticles and matrix chains at $N_m = 48$. The green bead is the nanoparticle, and the golden and purple beads refer to the grafted and matrix...
monomers respectively.

**S5. The location of the primary peak in pair correlation functions**

The location of the peak-position and the number of peaks in the pair correlation function profiles is employed to characterize the local structure of self-assembled nanoparticles. In light of this, the position of the primary peak in pair correlation function as a function of $N_m$ for different $\lambda_B$ are shown in Fig. S5.

![Graph](image.png)

**Fig. S5** The primary peak position of the pair correlation functions as a function of matrix chain length for different matrix chain rigidness. (a) $k_\theta = 0$, (b) $k_\theta = 5.0$, (c) $k_\theta = 100.0$. 

$\lambda_B = 3.0$, $\lambda_B = 10.0$, $\lambda_B = 20.0$.