Supplementary Material

Molecular simulation study of the assembly of DNA-grafted nanoparticles: Effect of bidispersity in DNA strand length

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1. Schematic of coarse grained model and types of single stranded DNA to single stranded DNA hybridization

Figure S1. (a) Schematic of our coarse grain model. In this model, a nucleotide on each strand is represented with one coarse-grained bead. Each nanoparticle is grafted with N_g number of DNA strands ($N_g=8$ in this figure) with an equal number of short strands and long strands of length N_{short} and N_{long} , respectively. (b) Types of ssDNA:ssDNA hybridization: short-short (s:s), short-long (s:l), long-long (l:l).

2. Effect of bidispersity in DNA strand length on relative shape anisotropy

Figure S2. Relative shape anisotropy, $\langle RSA \rangle$, as a function of ensemble average number of nanoparticles per cluster, $\langle N \rangle$, with changing reduced temperature for 10 DNA grafted nanoparticles, in a $(100 \text{ nm})^3$ box, with each particle of diameter $D=4 \text{ nm}$ and grafted with $N_g=8$ strands with bidispersity in DNA strand length ratio ($N_{short}=4$) shown in legend and sequences listed in Table 1 for $N_{short}/D=1$. Also shown are the monodisperse cases (1:1) for N_{short} (GGCC) and N_{long} (AAAAAAAAGGCC). Error bars are calculated from the average of 10 trials.

3. Effect of bidispersity in DNA strand length at varying number of G/Cs on thermodynamics and structure of assembly

Figure S3. (a-c) Number of free nanoparticles, (d-f) ensemble average number of nanoparticles per cluster, $\langle N \rangle$, (g-i) average coordination number, $\langle Z \rangle$, and (j-l) radius of gyration of cluster as a function of reduced temperature, T^* , for 10 particles, in a $(100 \text{ nm})^3$ box, each of diameter $D=4 \text{ nm}$ and grafted with $N_g=8$ strands with bidisperse strand length ratio (a, d, g, j) 1:1.5, 1:2 (b, e, h, k), and (c, f, i, l) 1:3 and number of G/Cs per strand = 4 (dotted line), 6 (dashed line), or 8 (solid line). Error bars are calculated from the average of 10 trials.

4. Effect of bidispersity in DNA strand length on inter-particle distance of assembled clusters at $N_{short}:N_{long}=1:2$

Figure S4. (a-c) Nanoparticle-nanoparticle number profile, $N(r)$, versus $r-D$ and (d-f) ssDNA:ssDNA hybridization histogram at $T_{low}=0.08$ for 10 particles, in a $(100 \text{ nm})^3$ box, each of diameter $D=4 \text{ nm}$ and grafted with $N_g=8$ strands with bidispersity in DNA strand length ratio $N_{short}:N_{long}=1:2$ and number of G/Cs per strand (a and d) =4, (b and e) =6, or (c and f) =8 (sequences listed in Table 1). For (d-f), x-axis s:s denotes short strand to short strand hybridization, s:l denotes short strand to long strand hybridization and l:l denotes a long strand to long strand hybridization. Error bars are calculated from the average of 10 trials.

5. Effect of bidispersity in DNA strand length on inter-particle distance of assembled clusters at $N_{short}:N_{long}=1:3$

Figure S5. (a-c) Nanoparticle-nanoparticle number profile, $N(r)$, versus $r-D$ and (d-f) ssDNA:ssDNA hybridization histogram at $T_{low}=0.08$ for 10 particles, in a $(100 \text{ nm})^3$ box, each of diameter $D=4 \text{ nm}$ and grafted with $N_g=8$ strands with bidispersity in DNA strand length ratio $N_{short}:N_{long}=1:3$ and number of G/Cs per strand (a and d) =4, (b and e) =6, or (c and f) =8 (sequences listed in Table 1). For (d-f), x-axis s:s denotes short strand to short strand hybridization, s:l denotes short strand to long strand hybridization and l:l denotes a long strand to long strand hybridization. Error bars are calculated from the average of 10 trials.

6. Effect of bidispersity in DNA strand length at $N_g=16$ number of grafted strands on thermodynamics and structure of assembly

Figure S6. (a-c) Number of free nanoparticles, (d-f) ensemble average number of nanoparticles per cluster, $\langle N \rangle$, (g-i) average coordination number, $\langle Z \rangle$, and (j-l) radius of gyration of cluster as a function of reduced temperature, T^* , for 10 particles, in a $(100 \text{ nm})^3$ box, each of diameter $D=4 \text{ nm}$ and grafted with $N_g=16$ strands with bidisperse strand length ratio (a, d, g, j) 1:1.5, 1:2 (b, e, h, k), and (c, f, i, l) 1:3 and number of G/Cs per strand = 4 (dotted line), 6 (dashed line), or 8 (solid line). Error bars are calculated from the average of 10 trials.

7. Schematic of loops within final cluster

Figure S7. Representative snapshots of cluster formation at $T_{low}=0.08$ for 10 particles, in a $(100 \text{ nm})^3$ box, each of diameter $D=4 \text{ nm}$ and grafted with $N_g=8$ strands with bidispersity in DNA strand length ratio $N_{short}:N_{long} =$ (a) 4:4 and (b) 4:6 and number of G/Cs per strand = 4 (sequences listed in Table 1). Indicated in bright green are loops of size 3 and in magenta are loops of size 4 formed from the centers of clustered nanoparticles.

8. Effect of bidispersity in DNA strand length at number of G/Cs=4 on thermodynamics and structure of assembly

Figure S8. (a) Number of free nanoparticles, (b) ensemble average number of nanoparticles per cluster, $\langle N \rangle$, (c) average coordination number, $\langle Z \rangle$, and (d) radius of gyration of cluster as a function of reduced temperature, T^* , for 10 particles, in a $(100 \text{ nm})^3$ box, each of diameter $D=4$ nm and grafted with $N_g=8$ strands (dashed lines) and $N_g=16$ (solid lines) of with bidispersity in DNA strand length ratio shown in legend (sequences listed in Table 1 for $N_{short}/D=1, 1.5$, and 2). Error bars are calculated from the average of 10 trials.

9. Effect of bidispersity in DNA strand length at varying number of G/Cs on inter-particle distance of assembled clusters

Figure S9. (a-b) Nanoparticle-nanoparticle number profile, $N(r)$, versus $r-D$ and (c-d) ssDNA:ssDNA hybridization histogram at $T_{low}=0.08$ for 10 particles, in a $(100 \text{ nm})^3$ box, each of diameter $D=4 \text{ nm}$ and grafted with (a, c) $N_g=8$ or (b, d) $N_g=16$ strands with bidispersity in DNA strand length ratio $N_{short}:N_{long}=4:8$ and number of G/Cs per strand=4 (sequences listed in Table 1 for $N_{short}/D=1$). For (c-d), x-axis s:s denotes short strand to short strand hybridization, s:l denotes short strand to long strand hybridization and l:l denotes a long strand to long strand hybridization. Error bars are calculated from the average of 10 trials.