SI Appendix

Zhang et al.

Protocols:

Formation of individual DNA origami raft. The mixture of staple strands, modified strands (leg sticky ends, biotinylated staples for function rafts only, horizontal sticky ends) and single-stranded M13mp18 DNA genome was diluted in 1× TAE/ Mg^{2+} buffer (40 mM Tris-HCl, pH 8.0, 20 mM acetic acid, 2.5 mM EDTA and 12.5 mM magnesium acetate). The final concentration of M13mp18 DNA genome in the solution was 10 nM. The molar ratio of the M13mp18 DNA genome to each staple strand was ~ 1: 10. The sample was cooled from 70 °C to 20 °C in an incubator over the course of 10 hours. The DNA origami rafts were purified afterwards using Amicon Ultra 0.5 mL centrifugal filters (100K, Millipore). The concentration of the DNA origami rafts was measured using NanoPhotometer P300 (Implen GmbH).

Patch sticky-end functionalization on DNA origami rafts. The biotinylated DNA origami rafts (functional raft a/b) and fluorescent streptavidin (Alexa Fluor®, Thermo Fisher) were mixed at a ratio of 1: 32 in 1× TAE/ Mg^{2+} buffer, and incubated at room temperature for 5 hours. Then, the streptavidin functionalized DNA origami rafts were purified using Amicon Ultra 0.5 mL centrifugal filters (100K, Millipore) to remove extra streptavidin. The biotinylated strand (S36-biotin) was mixed with an equal molar amount of each arm sticky end (from cS36-Arm-a11 to cS36-Arm-c'20) in 1× TAE/ Mg^{2+} buffer, and incubated at room temperature for 30 minutes to form duplexes. The six pre-annealed duplexes were then added to six batches of streptavidin functionalized DNA origami rafts at a ratio of 128: 1. The mixtures were incubated at room temperature for 3 hours, and purified using Amicon Ultra 0.5 mL centrifugal filters (100K, Millipore). The concentrations of the final products were measured using NanoPhotometer P300 (Implen GmbH).

DNA functionalization on emulsion droplets. Biotinylated emulsion droplets were fabricated using the technique from L. Feng, et. al.'s paper (35). The biotinylated strand S36-biotin, strand cS36-cLeg20 and fluorescent streptavidin were mixed stoichiometrically and diluted to 0.5 μM in 100 μl TMS buffer (10mM Tris-HCl, pH 8.0, 3mM $MgCl_2$, 1mM SDS, 0.1mM NaN_3). The sample was annealed at room temperature for 30 minutes. In the folding experiment, 5 pmol of the linker strands (Linker-a20/b20) were added to the DNA/streptavidin solution. Then, 2 μl of creamed emulsion droplets was added to the DNA/streptavidin solution. The mixture was gently tumbled at room temperature for an hour and then washed three times to remove the excess DNA strands and streptavidin. The number density of DNA coated droplets was measured by fluorescent confocal microscopy (Leica TCS SP5).

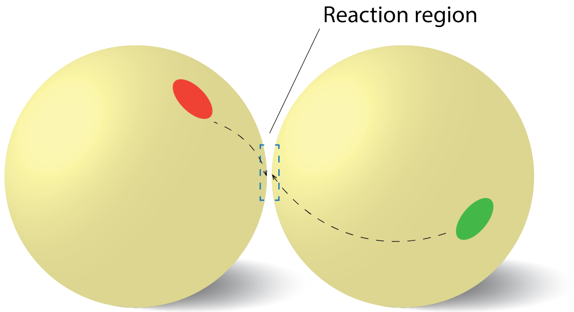
DNA origami patch formation on emulsion droplets. The functional rafts were diluted to 100 pM in TMS buffer, and mixed with emulsion droplets at a ratio of 400: 1. The mixture was then diluted with TMS buffer to keep the DNA origami concentration ~ 50 pM. The sample was gently tumbled at room temperature for 24 hours, and then washed to remove the unattached functional rafts. The shepherding rafts were diluted to 100 pM in TMS buffer, mixed with emulsion droplets at a ratio of 400: 1, and gently tumbled at room temperature for 12 hours. The above step was repeated twice. Then the sample was washed to remove the free DNA origami rafts in the solution. The patches on the droplets were characterized using fluorescence confocal microscopy and atomic force microscopy. To obtain 3D reconstruction from confocal Z stack, the patchy droplets were mixed with 20% polyacrylamide gel solution (50 μl 40% acrylamide/bis-acrylamide, 50 μl 2×TMS buffer, 1 μl 10% ammonium persulfate, 0.1 μl N,N,N',N'-Tetramethylethylenediamine) at a volume ratio of 1:1. Z-stack scanning was carried out immediately after gel polymerization.

Self-assembly of emulsion droplets with DNA origami patches. The droplets with various patch functionalities were mixed to form dimers (monovalent droplets), polymers (divalent droplets), or oligomers (monovalent and divalent droplets). The mixture was loaded into a capillary (#8100, VitroCom, Inc.), sealed with UV adhesives (NOA 68, Norland Products, Inc.) on a glass slide, and incubated at room temperature. The sample cell was tilted at an angle of ~ 70°. This allows for the packing of the droplets in the corner of the capillary, which significantly increases the kinetics for patch-mediated, inter-droplet binding. The sample was flipped every six hours to an opposite direction, enabling packing rearrangement. Data was collected after 6, 18, 36, 54, 78, 108 and 192 hours of incubation. During each collection, regions of interest were randomly selected and the numbers of droplets used for further statistical analysis were ~ 5 – 10% of the whole population.

Folding and unfolding of the trimers. Folding: The complementary linker strands (cLinker-a'20 and cLinker-b'10) were mixed at a 1: 1 ratio in TMS buffer (~ 0.5 μM) and incubated for 30 minutes to form duplexes. The mixture was then diluted to 10 nM in TMS buffer for further use. With the capillary containing the trimers unsealed, the linkers were injected such that the molar ratio of the linker duplexes to the sticky ends on the emulsion droplets (cLinker-a'20/b'10) ranges from 10:1 to 100:1. The capillary was re-sealed and incubated at room temperature for folding to happen. After a 4-hour incubation, the folding movies were taken using confocal fluorescent microscopy. Unfolding: After most trimers had folded, the capillary was unsealed again and an excess amount of the displacing strand DS17 (the ratio of DS17 to the linker duplex is ~ 10000: 1) was injected. The capillary was re-sealed and incubated at room temperature for another 4 hours before movie acquisition.

Calculation and Simulation:

Kinetics for patch binding. We first calculated the time that it takes for two droplets with complementary patches to bind, given they are held next to each other, as shown in the figure below. Then, we calculated the average inter-droplet binding time under different conditions, demonstrating that the densely packing of droplets will speed up the binding kinetics from several months to a few hours.



SI Fig. 1. A pair of droplets with complementary DNA origami patches held in contact. The two patches can diffuse on the surface. The red patch on the left droplet can bind to the green patch on the right droplet via DNA hybridization upon meeting in the reaction region.

a) When the two droplets with complementary patches are held in contact with each other, the average reaction time τ_r is calculated as (1):

$$\tau_r = \tau_s(1 + \frac{\tau_b}{\tau_t}), \tag{1}$$

where τ_s is the rotational search time (the average time for both patches to appear in the reaction region simultaneously); τ_b is the binding time of two complementary patches staying in their reaction region (the area A_i can be approximated as the patch's size ~ 0.5 μm^2), and τ_t is the translational transit time of both patches staying in the reaction region before diffusing away. τ_b can be written as $\tau_b = \tau_h/N_G$, where τ_h is the hybridization time for one pair of DNA sticky ends (from 1 s to 10 s), and N_G is the average number of DNA bonds that could form when two patches stay in the reaction region. The average number of rafts within one patch is ~ 50, among which half are functional rafts (with arm sticky ends for inter-droplet binding). There are 6 to 8 vertical sticky ends on each functional raft. Thus, the total number of the vertical sticky ends is 150 to 200. From this, we can estimate that N_G is ~ 100, and consequently, τ_b ~ 0.01 s to 0.1 s. τ_t can be calculated from a simple equation:

$$\tau_t \sim A_i / D_{patch}. \qquad (2)$$

Using $D_{patch} \sim 0.1\ \mu m^2/s$, we get τ_t ~ 1 s. The ratio of τ_b to τ_t is much less than 1. The average reaction time of the two droplets with complementary patches, when held in contact with each other, is just τ_s, indicating that the process is diffusion limited aggregation (DLA). A previous study (2) has shown the calculation of τ_s as:

$$\tau_s = D_{rot}^{-1} \frac{2ln2}{\alpha}, \qquad (3)$$

where D_{rot} is the rotational diffusion coefficient, and α is defined as $A_p/4\pi R_d^2$, the ratio of the patch's area (~ 0.5 μm^2, estimated from AFM images) to the droplet's surface area. In our case, the translational diffusion coefficient of DNA origami patch D_{patch} on droplet surface is on the order of $0.1\ \mu m^2/s$ and the radius of droplet R = 2.35 μm, leading to D_{rot} ~ 0.02 s^{-1}. $D_{origami}$ is obtained from previous work (3), in which they measured the diffusion constant ~ 0.2 μm^2/s for a 0.5 μm particle attached to an O/W droplet. Since the patch size in our system is slightly bigger than that, we simply took 0.1 μm^2/s as an estimation of the patch's diffusivity. Qualitatively this checks with our observations that a patch roughly diffuses its own diameter (0.5 μm) in a few seconds. Plugging all of these numbers into equation (2), we get $\tau_s \approx$ 3 hours, which is also the "conditional" reaction time τ_r.

b) In this section we calculated the "real" reaction time τ of two droplets with complementary patches under a more general condition (being held in contact with each other is **not** required). Considering a monolayer of droplets with a typical number density C_0 ~ 1500 /mm^2 (dilute case), the reaction time can be written as the following equation:

$$\tau = \tau_d + \frac{\tau_r}{4\pi R L C_0}, \qquad (4)$$

where L (~ 20 nm) is the thickness of the contact region, where patch sticky ends on the opposing droplets can reach each other. Using the measured 2D diffusion coefficient of droplets D_{2D} ~ 0.037 μm^2/s, we get a rough estimation of τ_d ~ 3 minutes. Plugging all the parameters into equation (4), we found the generalized reaction time is incredibly long ~ 4 months.

c) To speed up the reaction, we considered an "extreme" incubation protocol: the droplets are densely packed. In this scenario, the diffusion of droplets can be neglected, and the reaction time τ is equal to τ_r as calculated above. Close packing, however, hinders the rearrangement of droplets, and thus, a "flipping" technique is applied.

Monte Carlo simulation of the polymerization process. In the experiment, complementary divalent emulsion droplets were mixed so that they bind alternately following a step growth polymerization fashion. A Monte Carlo approach was implemented to simulate this process. In the simulation, the aggregation can be written by the following equation:

$$A_i + A_j \overset{k_{ij}}{\rightarrow} A_{ij}. \qquad (4)$$

Here, a constant reaction rate k was used for each k_{ij}. Thus, the characteristic aggregation time τ can be calculated as $1/kC_0$, where C_0 is the droplet concentration. In the experiment, τ can be extracted from the data, which is around 20 hours. The time step of each iteration is defined as $\Delta t = \frac{1}{kC_0 N_{1/2}} = \frac{1}{\tau N_{1/2}}$, where $N_{1/2}$ is half of the total number of emulsion droplets in one sample. In a typical sample cell, the length of the capillary is around *2 cm* and there are about 2 to 3 layers of droplets at the corner. Thus $N_{1/2} \approx 0.5 \times \frac{2\ cm}{5\ \mu m} \times 2.5 = 5000$, and $\Delta t \approx 14.4\, s$. The "checking points" in the experiment are: 6 hours, 18 hours, 54 hours, 108 hours and 192 hours, corresponding to 1500, 4500, 13500, 27000, 40000 iterations in the simulation, respectively.

With the system parameters set, the simulation was conducted following the steps below:

1. Assign indices to each particle (1, 2, 3, ... , N).
2. Set the degree of polymerization (DOP) of each particle to 1 (monomer)
3. Randomly pick two particles with different indices.
4. If the DOPs of both particles are non-zero, add the DOPs, and assign the updated value to particle i. Set the DOP of particle j to be zero (particle vanished). Otherwise, do not update any information (no reaction).
5. Iterate: step = step + 1, and go back to step 3
6. At each "checking point", calculate the weight fractions of chains with various lengths.

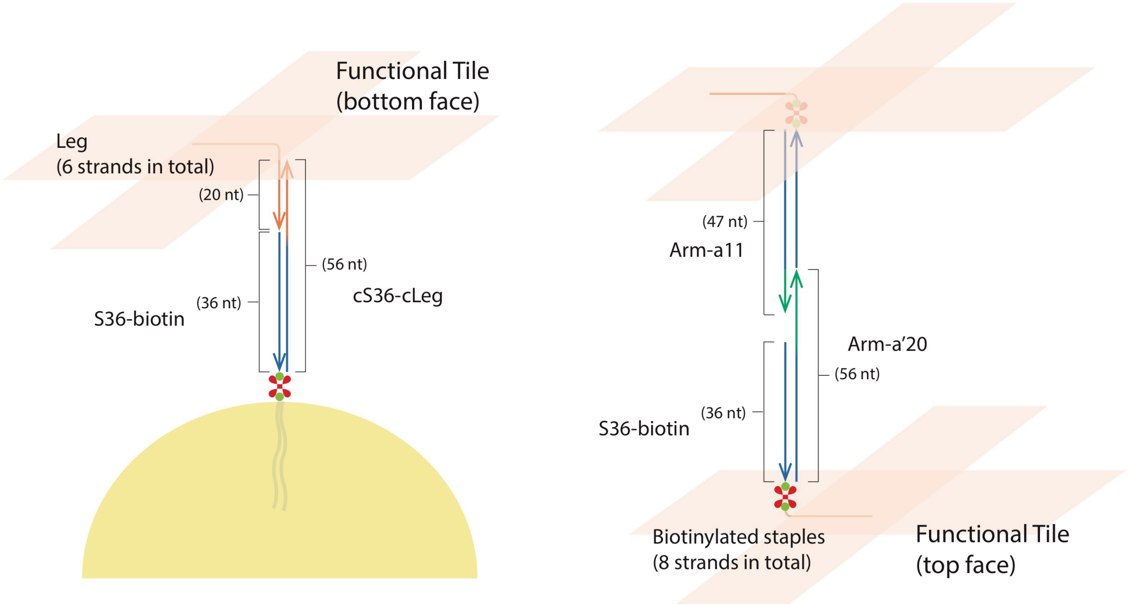


Fig. S1. Schematic diagram of functional DNA origami raft. Left panel: there are 6 single-stranded extensions (DNA "legs" of 20 bases) on the bottom face of the functional raft. These "legs" serve to anchor the functional raft to the complementary DNA coated emulsion droplet surface. Right panel: on the top face of the raft, fluorescent streptavidin molecules first bind to the biotin groups (8 in total) extended from the staples. Each "arm" contains two segments: a 36-nt stem that can hybridize with S36-biotin (attachment to the streptavidin molecules on the top face), and a 20-nt sticky end for patch binding.

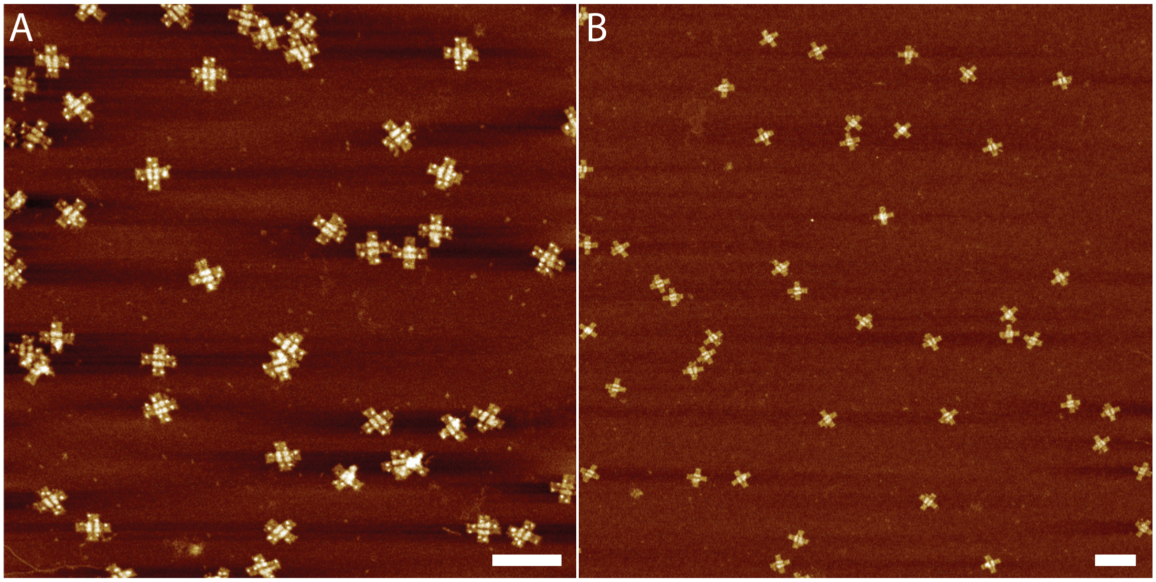


Fig. S2. AFM images of functional DNA origami rafts **(A)** and shepherding DNA origami rafts **(B)**. Scale bars are 200 nm.

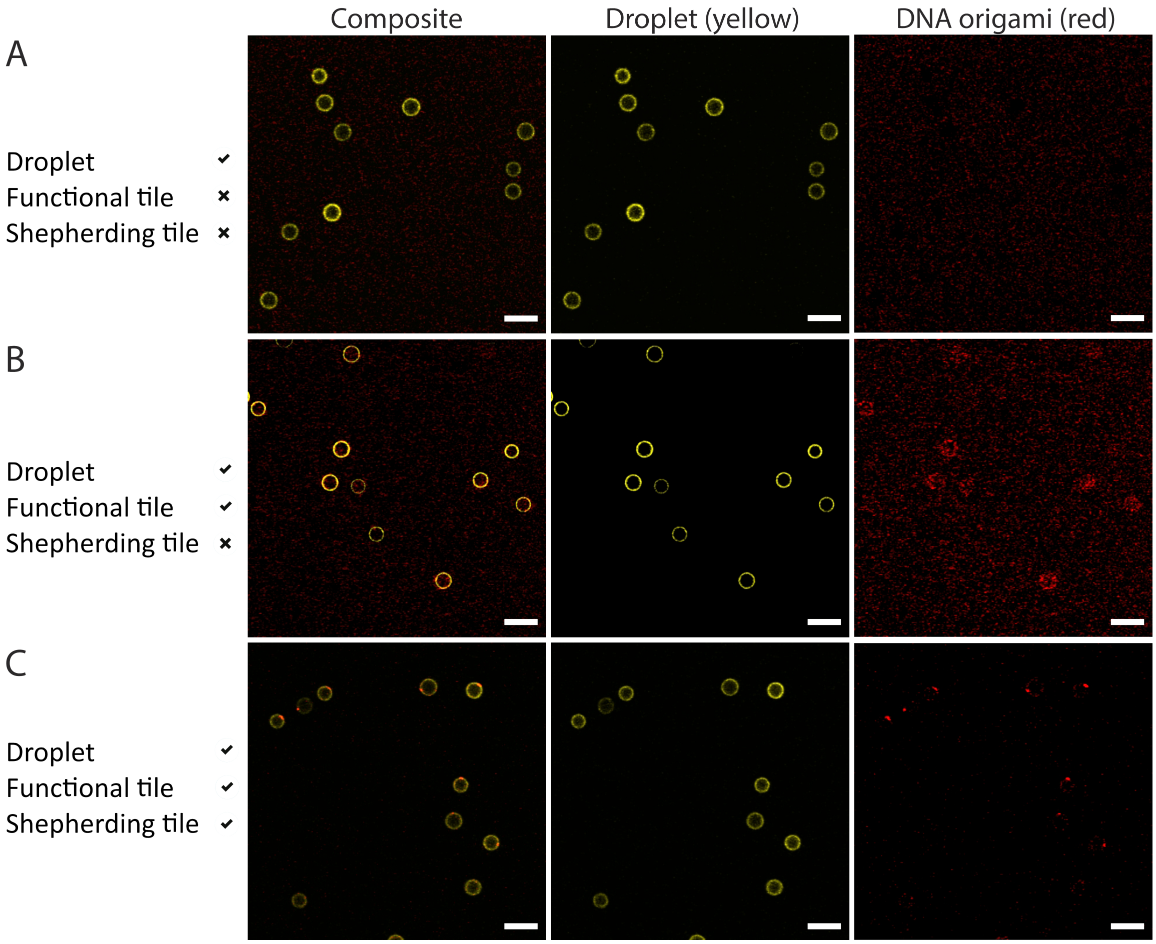


Fig. S3. Patch formation on DNA coated emulsion droplet surface. **(A)** Confocal images of the DNA coated droplets labeled with streptavidin Alexa Fluor® 546 (SA-AF 546) (yellow). **(B)** Confocal images of the functional rafts labeled with SA-AF 647 on droplet surface. The functional rafts (red) are attached and homogeneously distributed on the droplets as shown in the DNA origami channel. **(C)** Confocal images of patch formation on the droplets after shepherding rafts are added. The DNA origami channel shows the assembly of the functional rafts into one patch on each droplet. All scale bars are 10 µm.

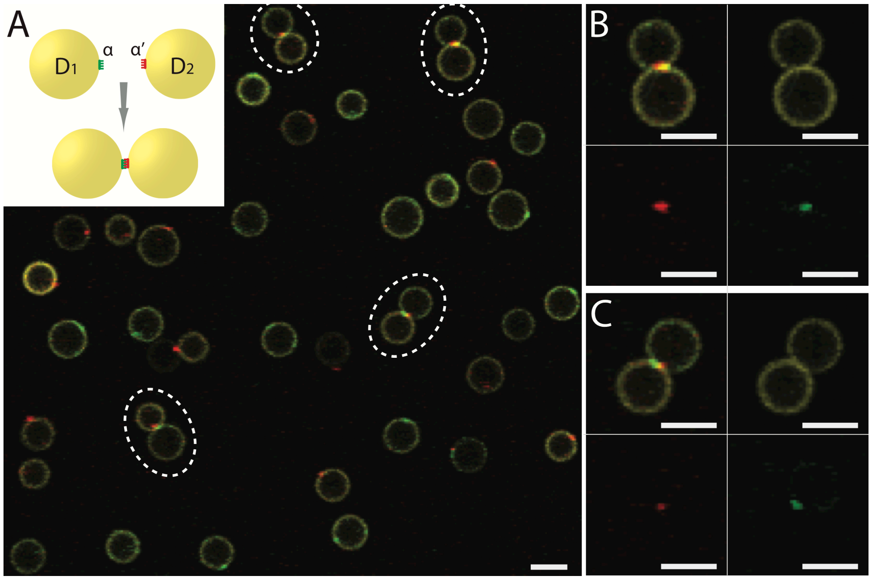


Fig. S4. (A) Low magnification confocal image of the dimer assembly after 12 hours. The droplets and the two complementary patches are labeled with SA-AF 546 (yellow), 488 (green) and 647 (red), respectively. Inset: schematic of dimer assembly from two monovalent droplets functionalized with complementary 'arm' sticky ends (α on D_1 and α' on D_2). The zoom-in images **(B)** and **(C)** show that each dimer is formed via the binding of complementary DNA origami patches. The top left, top right, bottom left and bottom right panel represents overlaid image, droplet channel, patch α' channel and patch α channel, respectively. All scale bars are 5 μm.

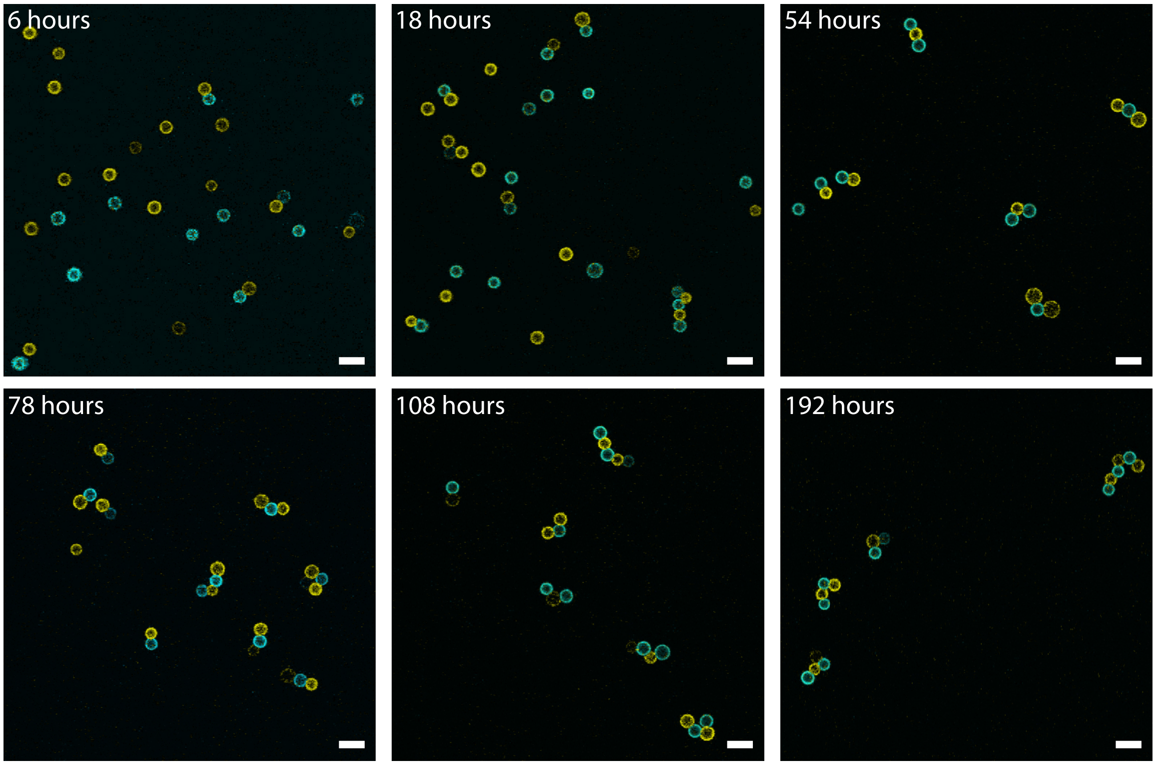


Fig. S5. Representative confocal images showing the growth of the polymers assembled from complementary divalent droplets. The two types of droplets are labeled either with SA-AF 488 (yellow) or with 546 (cyan). All scale bars are 10 μm.

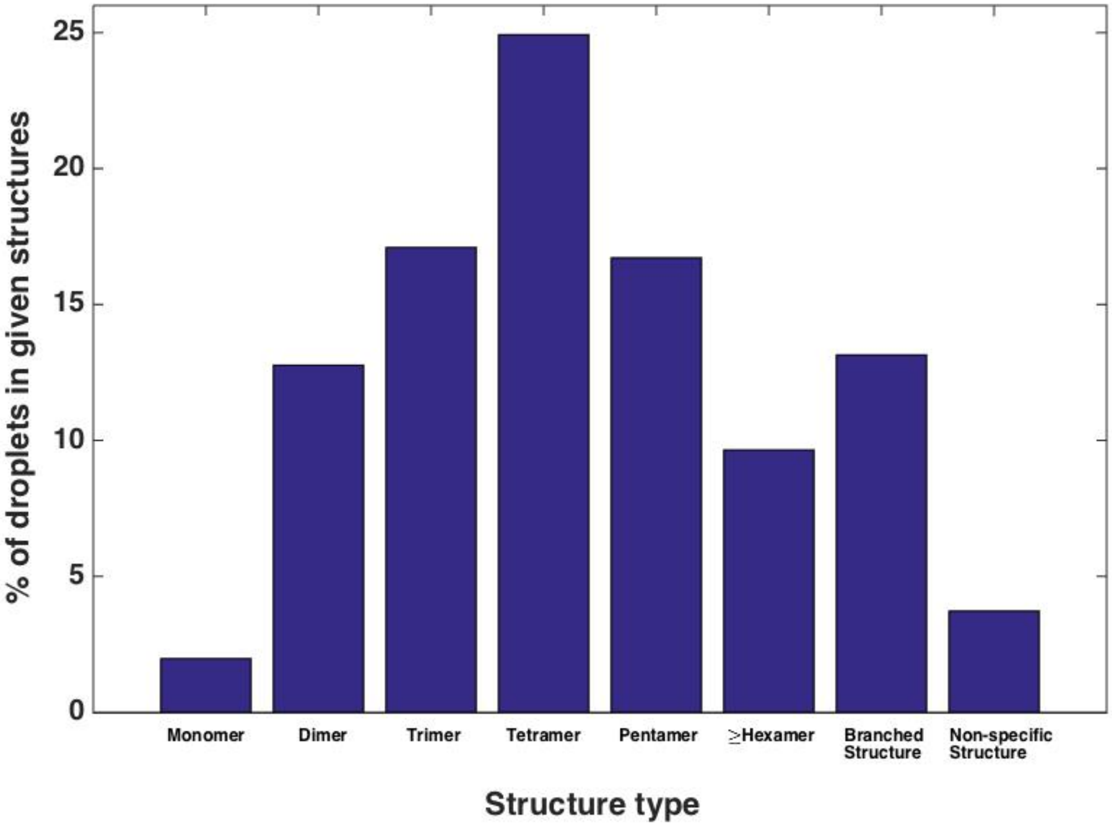


Fig. S6. Fractions of droplets in given structures after 192 hours of incubation.

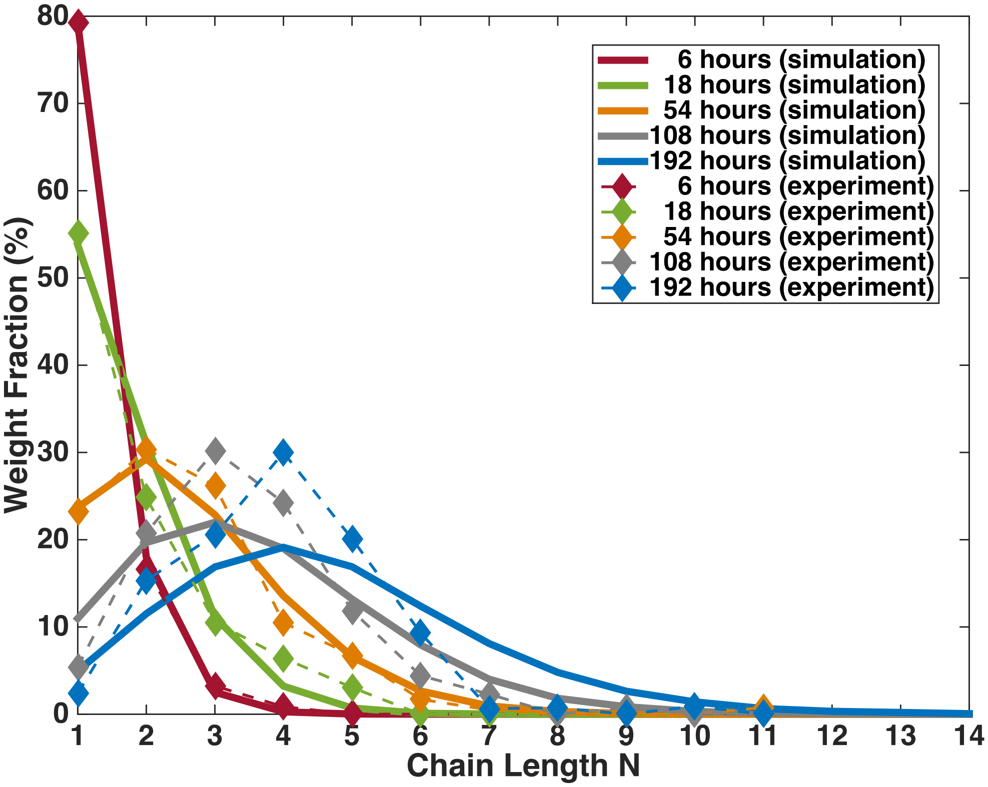


Fig. S7. A comparison between Monte Carlo simulation of the chain length distributions and experimental results at various time points. At each time point, the numbers of droplets in chains of various lengths are used to calculate the weight fractions. Note in experiments there is a significant reduction of long chains at long times as compared to the Monte Carlo simulation.

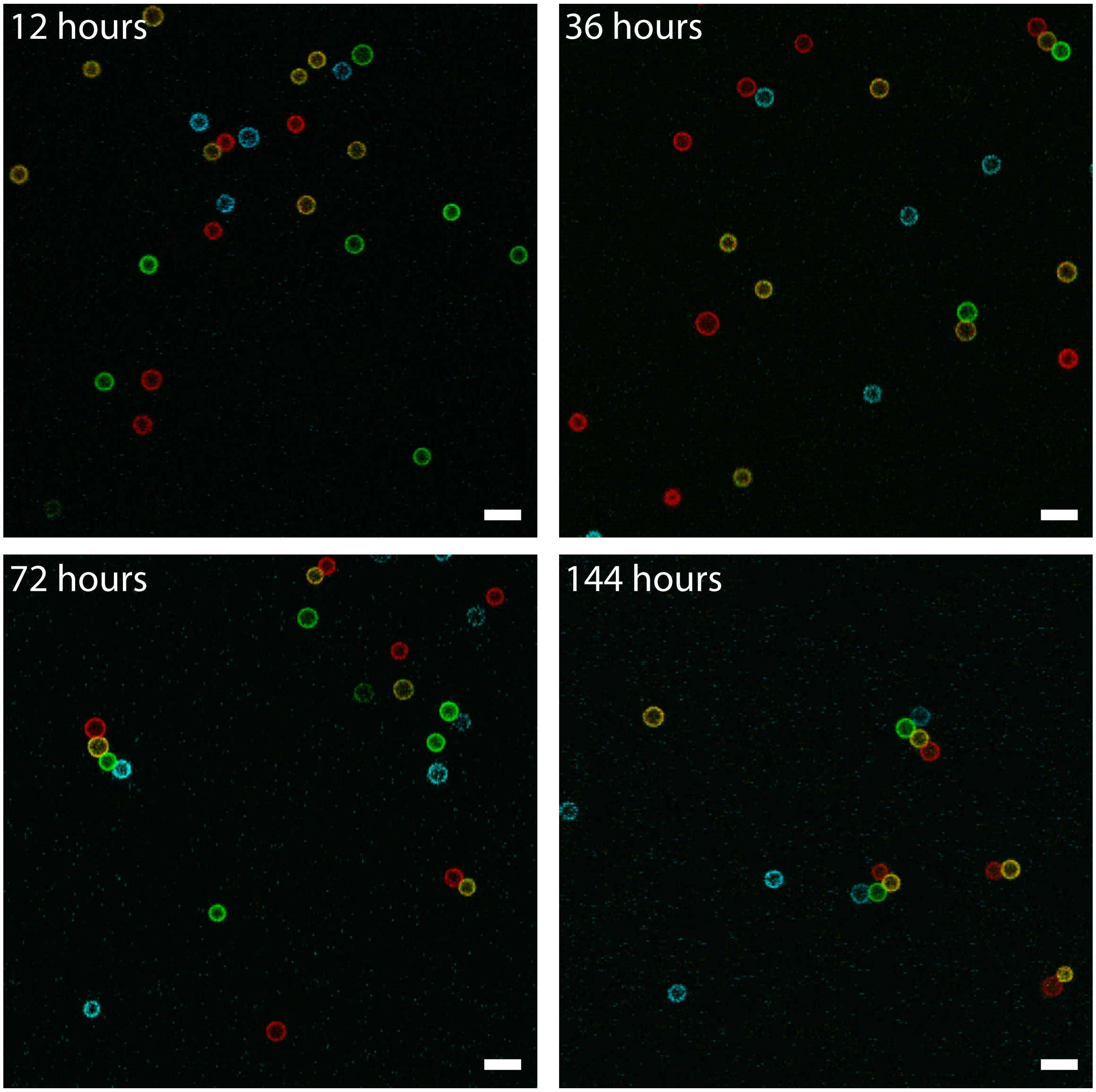


Fig. S8. Representative confocal images showing the growth of tetramers assembled from four types of droplets with patches bearing pre-programmed binding specificity. The four types of droplets are: red monovalent droplets equipped with one patch α, yellow divalent droplets equipped with one patch α’ and one patch β, green divalent droplets equipped with one patch β’ and one patch γ and blue monovalent droplets equipped with one patch γ’, respectively. All scale bars are 10 μm.

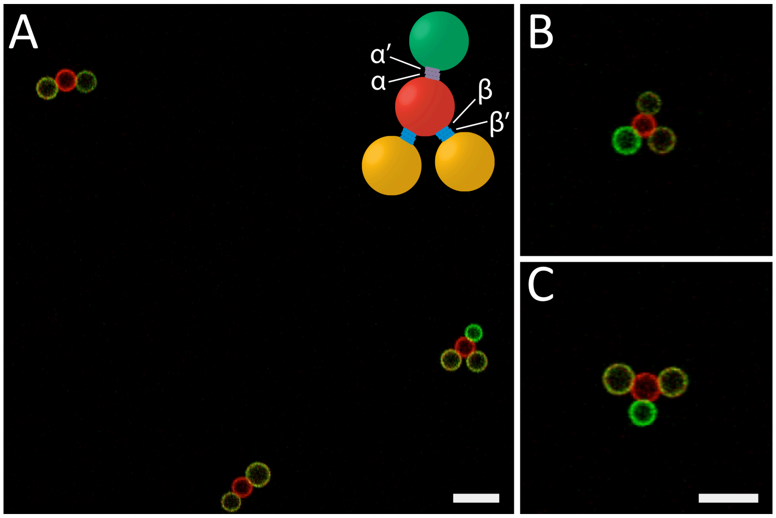


Fig. S9. Representative confocal images showing the hetero trivalent clusters assembled from three types of droplets with patches bearing pre-programmed binding specificity after 102 hours of incubation. The three types of droplets are: red (core) trivalent droplets equipped with one patch α and two patch β, green monovalent droplets equipped with one patch α' and yellow monovalent droplets equipped with patch β', respectively. All scale bars are 10 µm.

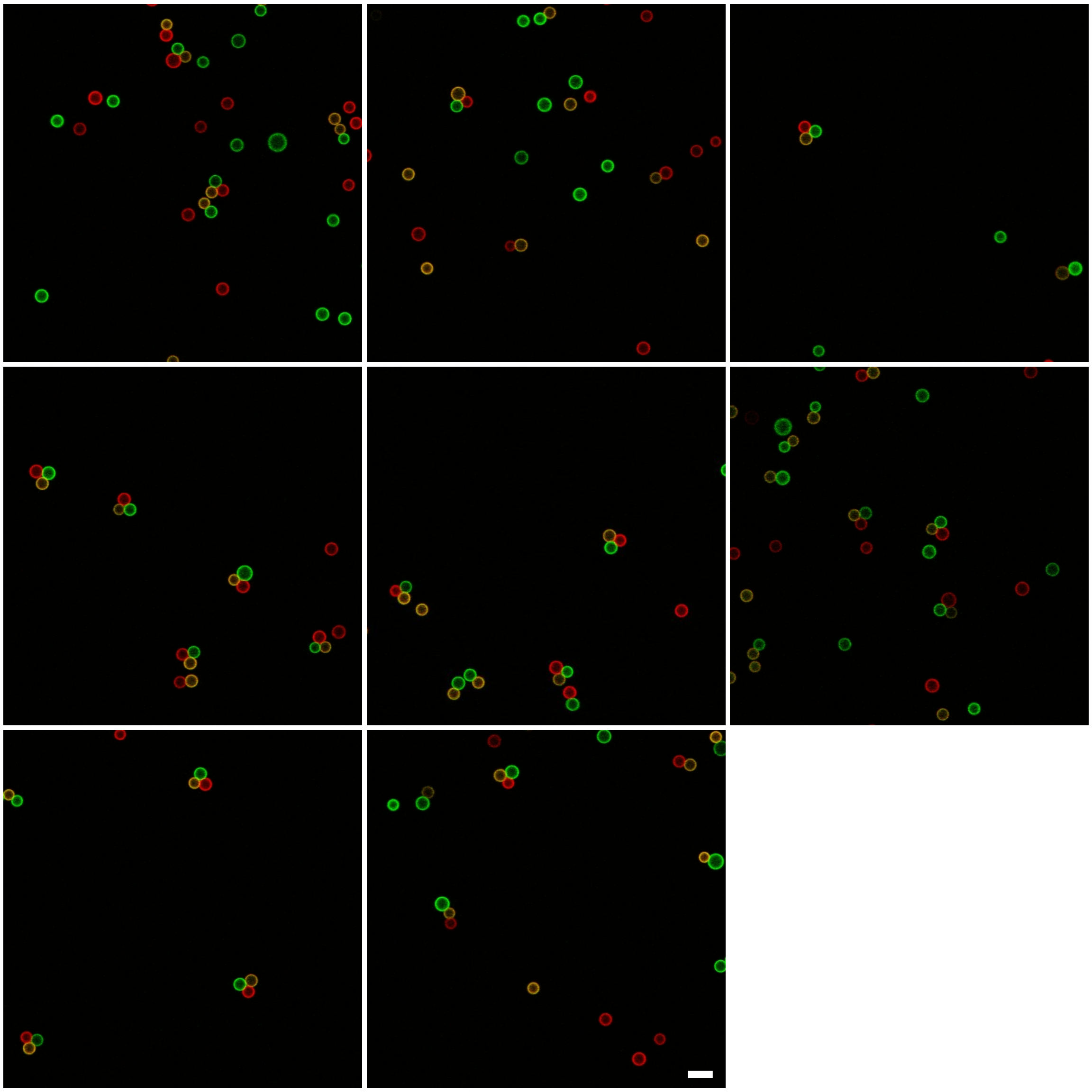


Fig. S10. Representative confocal images of the 'triangles' formed after adding the linker strands. 19 'triangles' were counted with 1 linear trimer failed to fold, which could be caused by the fact that the sticky ends on "head" and "tail" of a trimer are both covered by linker strands. Scale bar is 10 µm.

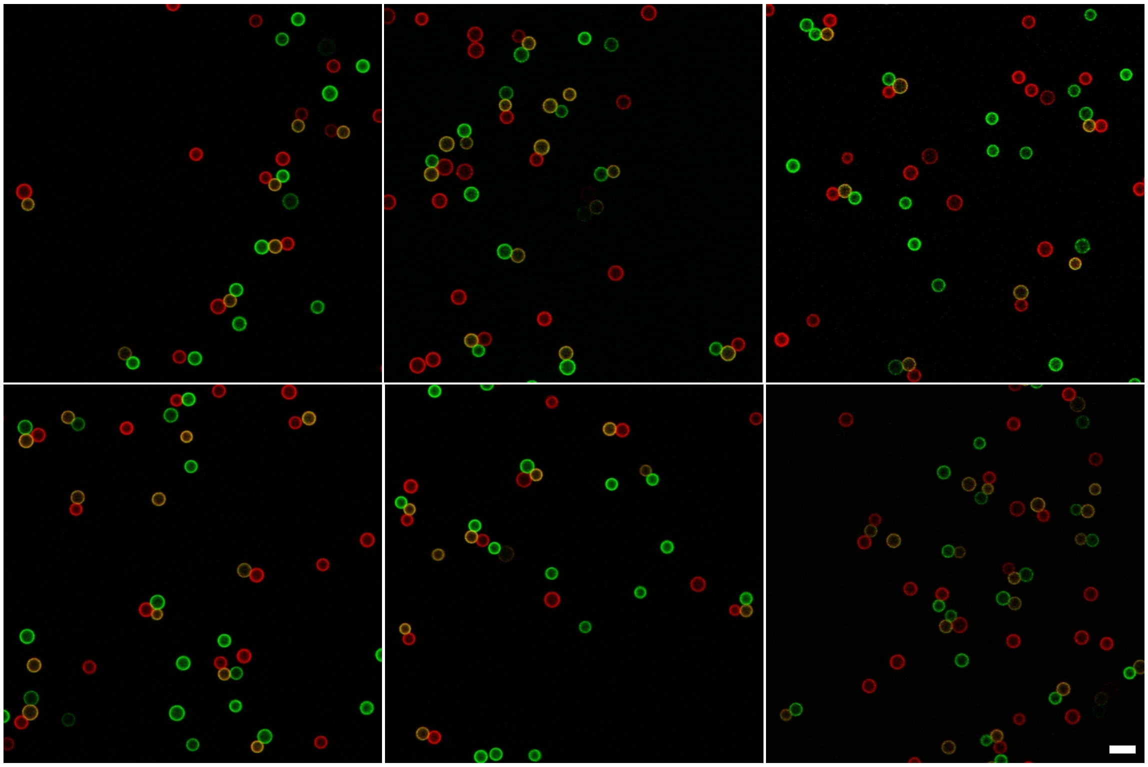


Fig. S11. Representative confocal images of the unfolded linear trimers formed 4 hours after adding the displacing strands. 16 linear trimers were counted with 4 'triangles' stayed unfold, which might be due to the slow kinetics for all the linkers to be unzipped via toehold strand displacement reaction. Scale bar is 10 µm.

Table S1. Staple sequences of cross DNA origami

Name	Sequence
CO-M-1	AGC TAA TGC AGA ACG CGC CTG TTT TAA TAT CC
CO-M-2	CAT CCT AAT TTG AAG CCT TAA ATC TTT TAT CC
CO-M-3	TGA ATC TTG AGA GAT AAC CCA CAA AAC AAT GA
CO-M-4	AAT AGC AAT AGA TGG GCG CAT CGT ACC GTA TC
CO-M-5	GGC CTC AGC TTG CAT GCC TGC AGG GAA TTC GT
CO-M-6	AAT CAT GGT GGT TTT TCT TTT CAC CCG CCT GG
CO-M-7	CCC TGA GAG AGT TGC AGC AAG CGG GTA TTG GG
CO-M-8	CGC CAG GGT CAT AGC TGT TTC CTG GAC GGC CA
CO-M-9	GTG CCA AGG AAG ATC GAC ATC CAG ATA GGT TA
CO-M-10	CGT TGG TGT AGC TAT CTT ACC GAA TTG AGC GC
CO-M-11	TAA TAT CAA CCT TCG CTA ACG AGC CCG ACT TG
CO-M-12	CGG GAG GTT TTA CGA GCA TGT AGA ACA TGT TC
CO-M-13	CTG TCC AGA CGA CGA CAA TAA ACA AAC CAA TC
CO-M-14	AAT AAT CGC GTT TTA GCG AAC CTC GTC TTT CC
CO-M-15	AGA GCC TAC AAA GTC AGA GGG TAA GCC CTT TT
CO-M-16	TAA GAA AAG ATT GAC CGT AAT GGG CCA GCT TT
CO-M-17	CCG GCA CCC ACG ACG TTG TAA AAC TGT GAA AT
CO-M-18	TGT TAT CCG GGA GAG GCG GTT TGC TCC ACG CT
CO-M-19	GGT TTG CCC CAG CAG GCG AAA ATC AAT CGG CC
CO-M-20	AAC GCG CGG CTC ACA ATT CCA CAC CCA GGG TT
CO-M-21	TTC CCA GTG CTT CTG GTG CCG GAA GTG GGA AC
CO-M-22	AAA CGG CGG TAA GCA GAT AGC CGA AAC TGA AC
CO-M-23	ACC CTG AAA TTT GCC AGT TAC AAA TTC TAA GA
CO-M-24	ACG CGA GGG CTG TCT TTC CTT ATC AAG TAA TT
CO-M-25	AAT ATA AAG TAC CGA CAA AAG GTA ATT CCA AG
CO-M-26	AAC GGG TAG AAG GCT TAT CCG GTA ATA AAC AG
CO-M-27	CCA TAT TAA TTA GAC GGG AGA ATT ACA AAG TTA CC
CO-M-28	GTC GGA TTC TCC ACC AGG CA
CO-M-29	AAG CGC CAA TTA AGT TGG GTA ACG AAC ATA CG
CO-M-31	CCT GTC GTG CAT AAA GTG TAA AGC GAT GTG CT
CO-M-32	GCA AGG CGT TCG CCA TTC AGG CTG CGC AAC TG
CO-M-33	GGA AGC GCT TTA TCC CAA TCC AAA AAG CAA AT
CO-M-35	AGG CAT TTT CGA GCC AGT ACT CAT CG
CO-M-36	AGA ACA AGT ACC GCG CCC AAT AGC TAA GAA AC
CO-M-37	GAT TTT TTA CAG AGA GAA TAA CAT AAA AAC AG
CO-M-39	CCT AAT GAA CTG CCC GCT TTC CAG CCC TTA TA
CO-M-42	TTG CGC TCG TGA GCT AAC TCA CAT GAT AGC CC
CO-M-43	TAT TAC GCG GCG ATC GGT GCG GGC GAG GAT TT
CO-M-44	CAG CCT TTG TTT AAC GTC AAA AAT TTT CAA TT

CO-M-45	GGA ATC ATC AAG CCG TTT TTA TTT GTT ATA TA
CO-M-47	ACT ATA TGC TCC GGC TTA GGT TGG TCA TCG TA
CO-M-48	ACC TGA GCA GAG GCG AAT TAT TCA GAA AAT AG
CO-M-49	AGA AGT ATA ATA GAT AAT ACA TTT CTC TTC GC
CO-M-50	TAA AAC ATC TTT AAT GCG CGA ACT TAA TTG CG
CO-M-51	CTA TTA GTC GCC ATT AAA AAT ACC ATA GAT TA
CO-M-52	GAG CCG TCT AGA CTT TAC AAA CAA TTC GAC AA
CO-M-54	TTT TTA ACT AAA TGC TGA TGC AAA ATT GAG AA
CO-M-56	CAA GAC AAA AAT CAT AGG TCT GAG ACA AAC AT
CO-M-57	CAA GAA AAA TTG CTT TGA ATA CCA AGT TAC AA
CO-M-58	CTC GTA TTG GTG CAC TAA CAA CTA GAA CGA AC
CO-M-60	TGC TGG TAA TAT CCA GAA CAA TAT AAG CGT AA
CO-M-61	GAA TAC GTG AAG ATA AAA CAG AGG ATC TAA AA
CO-M-62	TAT CTT TAA AAT CCT TTG CCC GAA CCG CGA CCT GC
CO-M-63	CGA AAC AAA GTA ATA ACG GA
CO-M-64	TTC GCC TGC AAA ATT AAT TAC ATT AAT AGT GA
CO-M-66	ATA TGC GTT ATA CAA ATT CTT ACC TTT TCA AA
CO-M-67	TAT ATT TTG ACG CTG AGA AGA GTC TAA CAA TT
CO-M-68	TGA TTT GAT ACA TCG GGA GAA ACA CAA CGG AG
CO-M-70	ATT TTA AAG GAA TTG AGG AAG GTT TGA GGC GG
CO-M-71	TCA GTA TTA ACC CTT CTG ACC TGA TAC CGC CA
CO-M-72	GCC ATT GCA ACA GGA AAA ACG CTC TGG CCA AC
CO-M-73	AGA GAT AG AAC ACC GC CTG CAA CA AAA TCA AC
CO-M-74	AGT AGA AAA GTT TGA GTA ACA TTA
CO-M-76	GTA CCT TTA TTA CCT TTT TTA ATG CGA TAG CT
CO-M-77	TAG ATT AAA GTT AAT TCG ATC TTC TTA GTA TC
CO-M-78	TCA TAA TTA CTA GAA AAA GCC TGT TGA CCT AA
CO-M-79	ATT TAA TGA TCC TTG AAA ACA TAG GAA ACA GT
CO-M-80	ACA TAA ATA CGT CAG ATG AAT ATA TGG AAG GA
CO-M-81	ATT GAA CCA ATA TAA TCC TGA TTG TCA TTT TG
CO-M-82	CGG AAC AAT ATC TGG TCA GTT GGC GTG CCA CG
CO-M-83	CTG AGA GCA ATA AAA GGG ACA TTC ATG GAA AT
CO-M-84	ACC TAC ATT TTG ACG CTC AAT CGT CAG TCA CA
CO-M-85	CGA CCA GTC AGC AGC AAA TGA AAA TCA AAC CC
CO-M-86	TCA ATC AAA GAA ACC ACC AGA AGG ATG ATG GC
CO-M-87	AAT TCA TCA ACC ATA TCA AAA TTA TAG ATT TT
CO-M-88	CAG GTT TAC AAT ATA TGT GAG TGA TTA ATT TT
CO-M-89	CCC TTA GAG TTT GAA ATA CCG ACC CAC CGG AA
CO-M-90	ATA AGC AAA AAT TCG CGT TAA ATT TTT GTT AA
CO-M-91	CTC ATA TAA AAG ATT CAA AAG GGT AAG ATT GT
CO-M-92	CGA ACG AGA AAT GGT CAA TAA CCT TTA GAA CC

CO-M-93	ATA GTC AGG GAA GCC CGA AAG ACT CAA TTC TG
CO-M-94	ACC ACA TTT TAC GAG GCA TAG TAA TGA CTA TT
CO-M-95	CAA GAG TAA ATC AAC GTA ACA AAG TTA GGA AT
CO-M-96	TCA GTG AAG CGC ATA GGC TGG CTG ACC TTC AT
CO-M-97	CTA TCA TAA TTC ATC AGT TGA GAT CTG CTC AT
CO-M-98	CGC GTT TTA ATC AGG TCT TTA CCC GAG CAA CA
CO-M-99	ATA TTT TCT GTA ACA GTT GAT TCC TCA AAT AT
CO-M-100	CCG GAG ACG CAA GGA TAA AAA TTT GTT TAG CT
CO-M-101	ATC AGC TCA AGC CCC AAA AAC AGG GAG AAA GG
CO-M-102	AAT CAG AAA TTT TTT AAC CAA TAG GAA CGC CA
CO-M-103	ATT TCA ACA GTC AAA TCA CCA TCA CGG TTG AT
CO-M-104	TCA TTC CAA TTT GGG GCG CGA GCT AAG CCT TT
CO-M-105	AAA TCA AAA ATT CGA GCT TCA AAG TGG AAG TT
CO-M-106	GTA GAA AGA CCC TCG TTT ACC AGA ATG ACC AT
CO-M-107	CAG ACC AGT AAG GCT TGC CCT GAC TAT TAC AG
CO-M-108	CAG AAC GAG AAA GAG GAC AGA TGA ACG GTG TA
CO-M-109	AAA ACC AAA CTA ACG GAA CAA CAT GAG AAC AC
CO-M-110	AAC CGG AAG AGT TCA GAA AAC GAG ACG ACG ATA
CO-M-111	GGC ATC AAA CTA AAG TAC GGT GTC CGA ACC AG
CO-M-112	TTC AAC CGA ATA CTT TTG CGG GAG GAA AAG GT
CO-M-113	TCA AAA ATT CAA TCA TAT GTA CCC ATA TGA TA
CO-M-115	GAC CCT GTT TCT AGC TGA TAA ATT TCG TAA AA
CO-M-116	AAC AGT TAA CCA GAG CCG CCG CCA GAA CCG CC
CO-M-118	TAA AAC GAA ATA GCG AGA GGC TTT CTC AAA TG
CO-M-119	CCA ACT TTG TAG TAA ATT GGG CTT TAC GTT AA
CO-M-121	AGA GTA CCT ATT CAT TGA ATC CCC TGC AAA AG
CO-M-122	CAT CCA ATG GTG CTG TAG CTC AAC ATG TTT
CO-M-123	AGA GGG TAA ATC GGT TGT ACC AAA AGC ATT AA
CO-M-124	CCA GCT TTA ATC GAT GAA CGG TAA AAT GCC GG
CO-M-125	AAC AAG AGC ATC AAC ATT AAA TGT GAG CGA GTA ACA ACT TAA GGA AAC CGA GGA AA
CO-M-127	GTC ATA AAT TTA ATT GCT CCT TTT CTT AAT TG
CO-M-128	GTC AGG ACC CAG AGG GGG TAA TAG GCG GAA TC
CO-M-129	AAC GAG GCG CAG ACG GAA CTT TAA TCA TTG TGT TAT ACC A
CO-M-130	GCG CCG ACT TTA AGA ACT GGC TCA AAT TAC CT
CO-M-131	CAA CGC CTG ATA GCG TCC AAT ACT TAA AAT GT
CO-M-132	TAT TAT TCT GCG GAT GGC TTA GAG GAT AAG AG
CO-M-133	CCT CAG AGA TTA AGC AAT AAA GCC GCA AAG AA
CO-M-134	CGT CAC CGG TCA TTG CCT GAG AGT CTA CAA AG
CO-M-135	GCT ATC AGA CTT GAG CCA TTT GGG ATT ATC AC
CO-M-136	TTA GCA AAC CAC CAC CCT CAG AGC ACC GCC AC
CO-M-137	GTC ATT TTT GAA ACA TGA AAG TAT TCG GAA CC

CO-M-138	TTA GAC TGG TAG CAT TCC ACA GAC ACA AAC TA
CO-M-139	TAT GCG ATA ATG ACA ACA ACC ATC CGA TAG TT
CO-M-140	ATA ACC GAT CAT CTT TGA CCC CCA GCG ATT ATA CCA AGT TCA TGT TAC TTA GCC GG
CO-M-142	GAA CCA CCA TGC CCC CTG CCT ATT TAA GAG GC
CO-M-143	CCA GCA AAA GCC GCC ACC CTC AGA CGC CAC CA
CO-M-144	CGC AAT AAT AAC GGA ATA TTC ATT AAA GGT GAA ATT AGA G
CO-M-146	GTA ACA CTC TCA AGA GAA GGA TTA GGA TTA
CO-M-147	AGA ATT TCG TAA CGA TCT AAA GTT CAT GTA CC
CO-M-148	TAA AAC ACT ATA TTC GGT CGC TGA TTT CGA GG
CO-M-150	TTT CCA GAC GGT TTA TCA GCT TGC GGC TTG CA
CO-M-152	AGC AAG GCA CCA GAG CCA CCA CCG GCA TTG AC
CO-M-153	AGA CTC CTT TGA GGG AGG GAA GGT TTA CCA TT
CO-M-154	TCA ACC GAT ATT ACG CAG TAT GTT AGC AAA CG
CO-M-155	TCA CCG GAC GGA AAC GTC ACC AAT GGC GAC AT
CO-M-156	GGG TCA GTG AGG CAG GTC AGA CGA AAT CAA AA
CO-M-157	GGG ATA GCG CTC AGT ACC AGG CGG TTT TAA CG
CO-M-158	AAT TGT ATC GTT AGT AAA TGA ATT CAT TTT CA
CO-M-159	CAA CCT AAA AGG CCG CTT TTG CGG GAG CCT TT
CO-M-160	CCC TCA GCT ACG TAA TGC CAC TAC GAA GGC AC
CO-M-161	GGG ATT TTA AAA AGG CTC CAA AAG GAT CGT CA
CO-M-162	CGT CGA GAT CAG AGC CAC CAC CCT TTC TGT AT
CO-M-163	GAT ATT CAG TGT ACT GGT AAT AAG ATA AGT GC
CO-M-164	CGA TAG CAT TTG CCA TCT TTT CAT TTG GCC TT
CO-M-165	TAG AAA ATG CGC CAA AGA CAA AAG GAA ACC AT
CO-M-166	GTT TAC CAA CAT ACA TAA AGG TGG CAA CAT AT
CO-M-167	TAT TAG CGG CAC CGT AAT CAG TAG TTC ATA TG
CO-M-168	ATA CAG GAC AAA CGA ATG GAT CTT AGC CCC CT
CO-M-169	CGC CAC CCG GGT TGA TAT AAG TAT TTT TGA TG
CO-M-170	TCT CCA AAG CTA AAC AAC TTT CAA CTC AGA AC
CO-M-171	GGG TAA AAA GCG AAA GAC AGC ATC GTT GAA AA
CO-M-172	GGT AGC AAT TCA TGA GGA AGT TTC CAT TAA AC
CO-M-173	GCG GAG TGA TAA TAA TTT TTT CAC GGA ACG AG
CO-M-174	ATA GGT GTC CTC AGA ACC GCC ACC CAG TTT CA
CO-M-175	CCA GAA TGA AGC GTC ATA CAT GGC AGC CCG GA
CO-M-176	TCA AGT TTC GGC ATT TTC GGT CAT CAT TAA AG
CO-M-177	AAA AGA AAC ACA ATC AAT AGA AAA CGA CAG AA
CO-M-30*(1)	AGC CGG AAG CCA GCT GCA TTA ATG CTG TTT GAT GGT GTC TTC CTG TAG
CO-M-114*(1)	CTA GCA TGA ATT CGC GTC TGG CTG TTC CGA AAT CGG CAA AAT TCG GGA AA
CO-M-38*(2)	TTG GGA AGC AGC TGG CTT AAA GCT AGC TAT TTT TGA GAG ATC TGG AGC A

CO-M-126*(2)	CTG AAT CTA AAT CAT ACA GGC AAG TCA GAG CAT GAA AGG GGC TGG GGT G
CO-M-40*(3)	AAT CAA AAG AAT AGC CCT TTA AAT ATG CAT TCT ACT AAT AGT AGT AAC ATT AT
CO-M-41*(3)	GAG ATA GGG TTG TCA GGA TTA G
CO-M-117*(3)	CTT TAA ACC AAA CTC CAA CAG TTG AGT GTT GTT CGT AGA AGA ACT CAA ACT TTG AAT GG
CO-M-59*(4)	CAC CAG CAG GCA CAG ATT TAA TTT CTC AAT CAT AAG GGA ACC GAA CTG A
CO-M-120*(4)	AAG TTT TGG TTG GGA AGA AAA ATC GAG ATG GTT CAA TAT TTA TCG GCC T
CO-M-53*(5)	AAT CGC GCA AAA GAA GTT AGT TAG CTT AAA CAG CTT GAT ACG CCC ACG C
CO-M-141*(5)	TGA GAC TCG AGT TTC GTC ACC AGT AGC CCT CAT ATG ATG AAA GAC TAC C
CO-M-65*(5)	ATT TAT CAA GAA CGC GAG AAA ACT AGT ATA AAG CCA ATA AAG AAT ACA C
CO-M-149*(6)	GGG AGT TAA ACG AAA GAG GCG TCG CTC AAC AGT AGG GCT TAT CCA ATC G
CO-M-55*(7)	TCG CCA TAT TTA ACA ACG TTG CGG GGT TTT AAG CCC AAT AGG AAC CTT GTC GTC
CO-M-46*(7)	CCA ACA GTG TGT GCC CGT ATA
CO-M-151*(7)	AGG AGG TTG CCT TGA GTA ACA TAA TTT AGG CAG
CO-M-34*(8)	CAG ATA TAT TAA ACC ATA CGG AAA TTA CCC AAA AGA ACT GGC ATG ATT A
CO-M-145*(8)	TCC CTC AGA TCA CCA GTA GCA CCA AAA TAT TGT AGT ACC GCA ATA AGA G
CO-M-69#	TTT GGA TTA TAC CTG ATA AAT TGT GTC GAA ATC GTT ATT A
CO-M-75#	ATT TGT ATC ATC GCT TCT GAA TTA CAG TAA CA
FT-L1-Blunt (AT-U1)	TCC TGA ACA AGA AAA AAT CAA CAA TAG ATA AG
FT-L2-Blunt (AT-U2)	TTG CAC CCA GCT ACA AAA GAT TAG TTG CTA TT
FT-L3-Blunt (AT-U3)	AAT AAT AAG AGC AAG AGA ATT GAG TTA AGC CC
FT-L4-Blunt (AT-U4)	GTT TGA GGG GAC GAC GAA CCG TGC ATC TGC CA
FT-L5-Blunt (AT-U5)	CCC GGG TAC CGA GGT CTC GAC TCT AGA GGA TC
FT-L6-Blunt (AT-U6)	AGC TGA TTG CCC TTC ACA GTG AGA CGG GCA AC
FT-R1-Blunt (AT-D1)	GTT AAA TAA GAA TAA AGT GTG ATA AAT AAG GC
FT-R2-Blunt (AT-D2)	AAA TCG TCG CTA TTA AAT AAC CTT GCT TCT GT
FT-R3-Blunt (AT-D3)	AAA TAA AGA AAT TGC GTT AGC ACG TAA AAC AG
FT-R4-Blunt (AT-D4)	TAT TCC TGA TTA TCA GAG CGG AAT TAT CAT CA
FT-R5-Blunt (AT-D5)	TGC TGA ACC TCA AAT AAT CTA AAG CAT CAC CT
FT-R6-Blunt (AT-D6)	ACA TTG GCA GAT TCA CCT GAA ATG GAT TAT TT
FT-U1-Blunt (AT-R1)	AAT AAG TTT ATT TTG TCG CAA AGA CAC CAC GG
FT-U2-Blunt (AT-R2)	TGT AGC GCG TTT TCA TGC CTT TAG CGT CAG AC
FT-U3-Blunt (AT-R3)	AAT TTA CCG TTC CAG TGA AAG CGC AGT CTC TG
FT-U4-Blunt (AT-R4)	GGT TTA GTA CCG CCA CAT CAC CGT ACT CAG GA
FT-U5-Blunt (AT-R5)	ACT AAA GGA ATT GCG AAG AAT AGA AAG GAA CA
FT-U6-Blunt (AT-R6)	GAG GAC TAA AGA CTT TCG GCT ACA GAG GCT TT
FT-D1-Blunt (AT-L1)	CGT TAA TAT TTT GTT AAT ATT TAA ATT GTA AA
FT-D2-Blunt (AT-L2)	TGA GTA ATG TGT AGG TTT TTA AAT GCA ATG CC
FT-D3-Blunt (AT-L3)	ATT AGA TAC ATT TCG CTA GAT TTA GTT TGA CC

FT-D4-Blunt (AT-L4)	ATC AAA AAG ATT AAG AAA GCA AAG CGG ATT GC
FT-D5-Blunt (AT-L5)	ATA ACG CCA AAA GGA ACA ACT AAT GCA GAT AC
FT-D6-Blunt (AT-L6)	GGA TAT TCA TTA CCC AAT CTT CGA CAA GAA CC

Table S2. Biotinylated staples, leg sticky ends and arm sticky ends on DNA origami

Name	Sequence
Biotin4T-CO-M-1	AGC TAA TGC AGA ACG CGC CTG TTT TAA TAT CCC ATC CTA ATT TGA AGC TTT T/3BioTEG/
Biotin4T-CO-M-16	TAA GAA AAG ATT GAC CGT AAT GGG CCA GCT TTC CGG CAC CCA CGA CGT TTT T/3BioTEG/
Biotin4T-CO-M-72	GCC ATT GCA ACA GGA AAA ACG CTC TGG CCA ACG AGA TAG AAC ACC GC TTT T/3BioTEG/
Biotin4T-CO-M-87	AAT TCA TCA ACC ATA TCA AAA TTA TAG ATT TTC AGG TTT ACA ATA TAT TTT T/3BioTEG/
Biotin4T-CO-M-92	CGA ACG AGA AAT GGT CAA TAA CCT TTA GAA CCT TTT /3BioTEG/
Biotin4T-CO-M-107	CAG ACC AGT AAG GCT TGC CCT GAC TAT TAC AG TTTT /3BioTEG/
Biotin4T-CO-M-165	TAG AAA ATG CGC CAA AGA CAA AAG GAA ACC ATT TTT /3BioTEG/
Biotin4T-CO-M-174	ATA GGT GTC CTC AGA ACC GCC ACC CAG TTT CAT TTT /3BioTEG/
Leg20-CO-M-19	GGT TTG CCC CAG CAG GCG AAA ATC AAT CGG CCT TTT TTT TTT CGT AAG TGG TGT TCC AAC TG
Leg20-CO-M-21	TTC CCA GTG CTT CTG GTG CCG GAA GTG GGA ACT TTT TTT TTT CGT AAG TGG TGT TCC AAC TG
Leg20-CO-M-23	ACC CTG AAA TTT GCC AGT TAC AAA TTC TAA GAT TTT TTT TTT CGT AAG TGG TGT TCC AAC TG
Leg20-CO-M-66	ATA TGC GTT ATA CAA ATT CTT ACC TTT TCA AAT TTT TTT TTT CGT AAG TGG TGT TCC AAC TG
Leg20-CO-M-68	TGA TTT GAT ACA TCG GGA GAA ACA CAA CGG AGT TTT TTT TTT CGT AAG TGG TGT TCC AAC TG
Leg20-CO-M-70	ATT TTA AAG GAA TTG AGG AAG GTT TGA GGC GGT TTT TTT TTT CGT AAG TGG TGT TCC AAC TG
S36-biotin	CAT CGA ACA ATC CGG TCG AGT GCC ATG ATT TGT GAG/3BioTEG/
cS36-Arm-a11	CTC ACA AAT CAT GGC ACT CGA CCG GAT TGT TCG ATG GAG ATT CCA GC
cS36-Arm-a’20	CTC ACA AAT CAT GGC ACT CGA CCG GAT TGT TCG ATG AGC AGC AGA GCT GGA ATC TC
cS36-Arm-b11	CTC ACA AAT CAT GGC ACT CGA CCG GAT TGT TCG ATG TCG TAC ATA AT
cS36-Arm-b’20	CTC ACA AAT CAT GGC ACT CGA CCG GAT TGT TCG ATG ACG TCT GAT ATT ATG TAC GA
cS36-Arm-c11	CTC ACA AAT CAT GGC ACT CGA CCG GAT TGT TCG ATG CCT TAG TGC TC
cS36-Arm-c’20	CTC ACA AAT CAT GGC ACT CGA CCG GAT TGT TCG ATG CTA GGT CAC GAG CAC TAA GG

Table S3. Horizontal sticky ends on cross DNA origami

Name	Sequence
FT-A-L1	TCC TGA ACA AGA AAA AAT CAA CAA TAG ATA AGA GCA T
FT-A-L2	TTG CAC CCA GCT ACA AAA GAT TAG TTG CTA TTG CAA A
FT-A-L3	ATC CTA ATA ATA AGA GCA AGA GAA TTG AGT TAA GCC CTA TGG
FT-A-L4	GTC TTG TTT GAG GGG ACG ACG AAC CGT GCA TCT GCC AAA GGT
FT-A-L5	CGA ATC CCG GGT ACC GAG GTC TCG ACT CTA GAG GAT C
FT-A-L6	CTG TTA GCT GAT TGC CCT TCA CAG TGA GAC GGG CAA C
FT-A-R1	CTG TTG TTA AAT AAG AAT AAA GTG TGA TAA ATA AGG C
FT-A-R2	CGA ATA AAT CGT CGC TAT TAA ATA ACC TTG CTT CTG T
FT-A-R3	GTC TTA AAT AAA GAA ATT GCG TTA GCA CGT AAA ACA GAA GGT
FT-A-R4	ATC CTT ATT CCT GAT TAT CAG AGC GGA ATT ATC ATC ATA TGG
FT-A-R5	TGC TGA ACC TCA AAT AAT CTA AAG CAT CAC CTG CAA A
FT-A-R6	ACA TTG GCA GAT TCA CCT GAA ATG GAT TAT TTA GCA T
FT-A-U1	AAT AAG TTT ATT TTG TCG CAA AGA CAC CAC GGA GTG T
FT-A-U2	TGT AGC GCG TTT TCA TGC CTT TAG CGT CAG ACG TTC A
FT-A-U3	TGA GTA ATT TAC CGT TCC AGT GAA AGC GCA GTC TCT GTC TAC
FT-A-U4	CTA TCG GTT TAG TAC CGC CAC ATC ACC GTA CTC AGG AAC TTG
FT-A-U5	GAC ATA CTA AAG GAA TTG CGA AGA ATA GAA AGG AAC A
FT-A-U6	CGT AAG AGG ACT AAA GAC TTT CGG CTA CAG AGG CTT T
FT-A-D1	CGT AAC GTT AAT ATT TTG TTA ATA TTT AAA TTG TAA A
FT-A-D2	GAC ATT GAG TAA TGT GTA GGT TTT TAA ATG CAA TGC C
FT-A-D3	CTA TCA TTA GAT ACA TTT CGC TAG ATT TAG TTT GAC CAC TTG
FT-A-D4	TGA GTA TCA AAA AGA TTA AGA AAG CAA AGC GGA TTG CTC TAC
FT-A-D5	ATA ACG CCA AAA GGA ACA ACT AAT GCA GAT ACG TTC A
FT-A-D6	GGA TAT TCA TTA CCC AAT CTT CGA CAA GAA CCA GTG T
AT-A-U1	TCC TGA ACA AGA AAA AAT CAA CAA TAG ATA AGA CAC T
AT-A-U2	TTG CAC CCA GCT ACA AAA GAT TAG TTG CTA TTT GAA C
AT-A-U3	ACT CAA ATA ATA AGA GCA AGA GAA TTG AGT TAA GCC CGT AGA
AT-A-U4	GAT AGG TTT GAG GGG ACG ACG AAC CGT GCA TCT GCC ACA AGT
AT-A-U5	ATG TCC CCG GGT ACC GAG GTC TCG ACT CTA GAG GAT C
AT-A-U6	TTA CGA GCT GAT TGC CCT TCA CAG TGA GAC GGG CAA C
AT-A-D1	TTA CGG TTA AAT AAG AAT AAA GTG TGA TAA ATA AGG C
AT-A-D2	ATG TCA AAT CGT CGC TAT TAA ATA ACC TTG CTT CTG T
AT-A-D3	GAT AGA AAT AAA GAA ATT GCG TTA GCA CGT AAA ACA GCA AGT
AT-A-D4	ACT CAT ATT CCT GAT TAT CAG AGC GGA ATT ATC ATC AGT AGA
AT-A-D5	TGC TGA ACC TCA AAT AAT CTA AAG CAT CAC CTT GAA C
AT-A-D6	ACA TTG GCA GAT TCA CCT GAA ATG GAT TAT TTA CAC T
AT-A-R1	AAT AAG TTT ATT TTG TCG CAA AGA CAC CAC GGA TGC T
AT-A-R2	TGT AGC GCG TTT TCA TGC CTT TAG CGT CAG ACT TTG C
AT-A-R3	AGG ATA ATT TAC CGT TCC AGT GAA AGC GCA GTC TCT GCC ATA

AT-A-R4	AAG ACG GTT TAG TAC CGC CAC ATC ACC GTA CTC AGG AAC CTT
AT-A-R5	ATT CGA CTA AAG GAA TTG CGA AGA ATA GAA AGG AAC A
AT-A-R6	AAC AGG AGG ACT AAA GAC TTT CGG CTA CAG AGG CTT T
AT-A-L1	AAC AGC GTT AAT ATT TTG TTA ATA TTT AAA TTG TAA A
AT-A-L2	ATT CGT GAG TAA TGT GTA GGT TTT TAA ATG CAA TGC C
AT-A-L3	AAG ACA TTA GAT ACA TTT CGC TAG ATT TAG TTT GAC CAC CTT
AT-A-L4	AGG ATA TCA AAA AGA TTA AGA AAG CAA AGC GGA TTG CCC ATA
AT-A-L5	ATA ACG CCA AAA GGA ACA ACT AAT GCA GAT ACT TTG C
AT-A-L6	GGA TAT TCA TTA CCC AAT CTT CGA CAA GAA CCA TGC T
FT-B-L1	TCC TGA ACA AGA AAA AAT CAA CAA TAG ATA AG AGCAA
FT-B-L2	TTG CAC CCA GCT ACA AAA GAT TAG TTG CTA TT AACTA
FT-B-L3	CTGAG A ATA ATA AGA GCA AGA GAA TTG AGT TAA GCC C CATGC
FT-B-L4	ATCTA G TTT GAG GGG ACG ACG AAC CGT GCA TCT GCC A TTTCA
FT-B-L5	AATCA C CCG GGT ACC GAG GTC TCG ACT CTA GAG GAT C
FT-B-L6	CATAC A GCT GAT TGC CCT TCA CAG TGA GAC GGG CAA C
FT-B-R1	CATAC G TTA AAT AAG AAT AAA GTG TGA TAA ATA AGG C
FT-B-R2	AATCA A AAT CGT CGC TAT TAA ATA ACC TTG CTT CTG T
FT-B-R3	ATCTA A AAT AAA GAA ATT GCG TTA GCA CGT AAA ACA G TTTCA
FT-B-R4	CTGAG T ATT CCT GAT TAT CAG AGC GGA ATT ATC ATC A CATGC
FT-B-R5	TGC TGA ACC TCA AAT AAT CTA AAG CAT CAC CT AACTA
FT-B-R6	ACA TTG GCA GAT TCA CCT GAA ATG GAT TAT TT AGCAA
FT-B-U1	AAT AAG TTT ATT TTG TCG CAA AGA CAC CAC GG GGTAG
FT-B-U2	TGT AGC GCG TTT TCA TGC CTT TAG CGT CAG AC TTCAG
FT-B-U3	TCAGC A ATT TAC CGT TCC AGT GAA AGC GCA GTC TCT G AGAAT
FT-B-U4	TGATG G GTT TAG TAC CGC CAC ATC ACC GTA CTC AGG A TACAA
FT-B-U5	GTGCA A CTA AAG GAA TTG CGA AGA ATA GAA AGG AAC A
FT-B-U6	CGTCG G AGG ACT AAA GAC TTT CGG CTA CAG AGG CTT T
FT-B-D1	CGTCG C GTT AAT ATT TTG TTA ATA TTT AAA TTG TAA A
FT-B-D2	GTGCA T GAG TAA TGT GTA GGT TTT TAA ATG CAA TGC C
FT-B-D3	TGATG A TTA GAT ACA TTT CGC TAG ATT TAG TTT GAC C TACAA
FT-B-D4	TCAGC A TCA AAA AGA TTA AGA AAG CAA AGC GGA TTG C AGAAT
FT-B-D5	ATA ACG CCA AAA GGA ACA ACT AAT GCA GAT AC TTCAG
FT-B-D6	GGA TAT TCA TTA CCC AAT CTT CGA CAA GAA CC GGTAG
AT-B-R1	AAT AAG TTT ATT TTG TCG CAA AGA CAC CAC GG TTGCT
AT-B-R2	TGT AGC GCG TTT TCA TGC CTT TAG CGT CAG AC TAGTT
AT-B-R3	CTCAG A ATT TAC CGT TCC AGT GAA AGC GCA GTC TCT G GCATG
AT-B-R4	TAGAT G GTT TAG TAC CGC CAC ATC ACC GTA CTC AGG A TGAAA
AT-B-R5	TGATT A CTA AAG GAA TTG CGA AGA ATA GAA AGG AAC A
AT-B-R6	GTATG G AGG ACT AAA GAC TTT CGG CTA CAG AGG CTT T
AT-B-L1	GTATG C GTT AAT ATT TTG TTA ATA TTT AAA TTG TAA A
AT-B-L2	TGATT T GAG TAA TGT GTA GGT TTT TAA ATG CAA TGC C

AT-B-L3	TAGAT A TTA GAT ACA TTT CGC TAG ATT TAG TTT GAC C TGAAA
AT-B-L4	CTCAG A TCA AAA AGA TTA AGA AAG CAA AGC GGA TTG C GCATG
AT-B-L5	ATA ACG CCA AAA GGA ACA ACT AAT GCA GAT AC TAGTT
AT-B-L6	GGA TAT TCA TTA CCC AAT CTT CGA CAA GAA CC TTGCT
AT-B-U1	TCC TGA ACA AGA AAA AAT CAA CAA TAG ATA AG CTACC
AT-B-U2	TTG CAC CCA GCT ACA AAA GAT TAG TTG CTA TT CTGAA
AT-B-U3	GCTGA A ATA ATA AGA GCA AGA GAA TTG AGT TAA GCC C ATTCT
AT-B-U4	CATCA G TTT GAG GGG ACG ACG AAC CGT GCA TCT GCC A TTGTA
AT-B-U5	TGCAC C CCG GGT ACC GAG GTC TCG ACT CTA GAG GAT C
AT-B-U6	CGACG A GCT GAT TGC CCT TCA CAG TGA GAC GGG CAA C
AT-B-D1	CGACG G TTA AAT AAG AAT AAA GTG TGA TAA ATA AGG C
AT-B-D2	TGCAC A AAT CGT CGC TAT TAA ATA ACC TTG CTT CTG T
AT-B-D3	CATCA A AAT AAA GAA ATT GCG TTA GCA CGT AAA ACA G TTGTA
AT-B-D4	GCTGA T ATT CCT GAT TAT CAG AGC GGA ATT ATC ATC A ATTCT
AT-B-D5	TGC TGA ACC TCA AAT AAT CTA AAG CAT CAC CT CTGAA
AT-B-D6	ACA TTG GCA GAT TCA CCT GAA ATG GAT TAT TT CTACC
FT-C-L1	TCC TGA ACA AGA AAA AAT CAA CAA TAG ATA AG CGTTA
FT-C-L2	TTG CAC CCA GCT ACA AAA GAT TAG TTG CTA TT AAAGT
FT-C-L3	TAGAC A ATA ATA AGA GCA AGA GAA TTG AGT TAA GCC C GAACC
FT-C-L4	CAATG G TTT GAG GGG ACG ACG AAC CGT GCA TCT GCC A AGGTA
FT-C-L5	CCTTG C CCG GGT ACC GAG GTC TCG ACT CTA GAG GAT C
FT-C-L6	TAGGA A GCT GAT TGC CCT TCA CAG TGA GAC GGG CAA C
FT-C-R1	TAGGA G TTA AAT AAG AAT AAA GTG TGA TAA ATA AGG C
FT-C-R2	CCTTG A AAT CGT CGC TAT TAA ATA ACC TTG CTT CTG T
FT-C-R3	CAATG A AAT AAA GAA ATT GCG TTA GCA CGT AAA ACA G AGGTA
FT-C-R4	TAGAC T ATT CCT GAT TAT CAG AGC GGA ATT ATC ATC A GAACC
FT-C-R5	TGC TGA ACC TCA AAT AAT CTA AAG CAT CAC CT AAAGT
FT-C-R6	ACA TTG GCA GAT TCA CCT GAA ATG GAT TAT TT CGTTA
FT-C-U1	AAT AAG TTT ATT TTG TCG CAA AGA CAC CAC GG AGTTT
FT-C-U2	TGT AGC GCG TTT TCA TGC CTT TAG CGT CAG AC GGTTG
FT-C-U3	GGTAC A ATT TAC CGT TCC AGT GAA AGC GCA GTC TCT G GTCTC
FT-C-U4	TCTAT G GTT TAG TAC CGC CAC ATC ACC GTA CTC AGG A AAGCA
FT-C-U5	GCTTA A CTA AAG GAA TTG CGA AGA ATA GAA AGG AAC A
FT-C-U6	CGTGT G AGG ACT AAA GAC TTT CGG CTA CAG AGG CTT T
FT-C-D1	CGTGT C GTT AAT ATT TTG TTA ATA TTT AAA TTG TAA A
FT-C-D2	GCTTA T GAG TAA TGT GTA GGT TTT TAA ATG CAA TGC C
FT-C-D3	TCTAT A TTA GAT ACA TTT CGC TAG ATT TAG TTT GAC C AAGCA
FT-C-D4	GGTAC A TCA AAA AGA TTA AGA AAG CAA AGC GGA TTG C GTCTC
FT-C-D5	ATA ACG CCA AAA GGA ACA ACT AAT GCA GAT AC GGTTG
FT-C-D6	GGA TAT TCA TTA CCC AAT CTT CGA CAA GAA CC AGTTT
AT-C-R1	AAT AAG TTT ATT TTG TCG CAA AGA CAC CAC GG TAACG

AT-C-R2	TGT AGC GCG TTT TCA TGC CTT TAG CGT CAG AC ACTTT
AT-C-R3	GTCTA A ATT TAC CGT TCC AGT GAA AGC GCA GTC TCT G GGTTC
AT-C-R4	CATTG G GTT TAG TAC CGC CAC ATC ACC GTA CTC AGG A TACCT
AT-C-R5	CAAGG A CTA AAG GAA TTG CGA AGA ATA GAA AGG AAC A
AT-C-R6	TCCTA G AGG ACT AAA GAC TTT CGG CTA CAG AGG CTT T
AT-C-L1	TCCTA C GTT AAT ATT TTG TTA ATA TTT AAA TTG TAA A
AT-C-L2	CAAGG T GAG TAA TGT GTA GGT TTT TAA ATG CAA TGC C
AT-C-L3	CATTG A TTA GAT ACA TTT CGC TAG ATT TAG TTT GAC C TACCT
AT-C-L4	GTCTA A TCA AAA AGA TTA AGA AAG CAA AGC GGA TTG C GGTTC
AT-C-L5	ATA ACG CCA AAA GGA ACA ACT AAT GCA GAT AC ACTTT
AT-C-L6	GGA TAT TCA TTA CCC AAT CTT CGA CAA GAA CC TAACG
AT-C-U1	TCC TGA ACA AGA AAA AAT CAA CAA TAG ATA AG AAACT
AT-C-U2	TTG CAC CCA GCT ACA AAA GAT TAG TTG CTA TT CAACC
AT-C-U3	GTACC A ATA ATA AGA GCA AGA GAA TTG AGT TAA GCC C GAGAC
AT-C-U4	ATAGA G TTT GAG GGG ACG ACG AAC CGT GCA TCT GCC A TGCTT
AT-C-U5	TAAGC C CCG GGT ACC GAG GTC TCG ACT CTA GAG GAT C
AT-C-U6	ACACG A GCT GAT TGC CCT TCA CAG TGA GAC GGG CAA C
AT-C-D1	ACACG G TTA AAT AAG AAT AAA GTG TGA TAA ATA AGG C
AT-C-D2	TAAGC A AAT CGT CGC TAT TAA ATA ACC TTG CTT CTG T
AT-C-D3	ATAGA A AAT AAA GAA ATT GCG TTA GCA CGT AAA ACA G TGCTT
AT-C-D4	GTACC T ATT CCT GAT TAT CAG AGC GGA ATT ATC ATC A GAGAC
AT-C-D5	TGC TGA ACC TCA AAT AAT CTA AAG CAT CAC CT CAACC
AT-C-D6	ACA TTG GCA GAT TCA CCT GAA ATG GAT TAT TT AAACT

Table S4. Sticky-end sequences on Emulsion Droplets

Name	Sequence
S36-biotin	CAT CGA ACA ATC CGG TCG AGT GCC ATG ATT TGT GAG/3BioTEG/
cS36-cLeg20	CTC ACA AAT CAT GGC ACT CGA CCG GAT TGT TCG ATG CAG TTG GAA CAC CAC TTA CG
Linker-a20	AGA AAG GAA ACG ATG AGG TT CGT AAG TGG TGT TCC AAC TG
Linker-b20	GGT TTA ACC AGT GAT CCA TG CGT AAG TGG TGT TCC AAC TG
cLinker-a’20	AAC CTC ATC GTT TCC TTT CT CTC ACA AAT CAT GGC ACT CGA CCG GAT TGT TCG ATG
cLinker-b’10	TGG TTA AAC C CAT CGA ACA ATC CGG TCG AGT GCC ATG ATT TGT GAG
DS17	CATGGATCACTGGTTAA

Table S5. Statistics of the divalent droplet polymerization*

	6 hours	18 hours	36 hours	54 hours	78 hours	108 hours	192 hours
Non-Specific structures	4	14	5	7	21	33	49 (18 non-specifically bound droplets)
Branched structures	0	4	28	27	36	77	173 (21 "branching" droplets)
Monomer	735	720	388	327	187	66	26
Dimer	154	318	278	428	268	254	168
Trimer	30	138	165	369	294	369	225
Tetramer	8	80	124	148	200	296	328
Pentamer	0	40	85	95	120	145	220
Hexamer	0	6	24	24	48	54	102
Heptamer	0	0	0	7	28	28	7
Octamer	0	0	8	0	8	0	8
Nonamer	0	0	0	0	9	9	0
Decamer	0	0	0	0	0	0	10
Undecamer	0	0	0	11	0	0	0
Dodecamer	0	0	0	0	0	0	0
Tridecamer	0	0	0	0	0	0	0
Tetradecamer	0	0	0	0	0	0	0
Total number counted	931	1320	1105	1443	1219	1318	1316

* In the statistics, the numbers of droplets in various targeted/unexpected structures were counted.

Table S6. Statistics of linear trimers (ABC structure)

	Unreacted monomers			Specific assemblies			NSA*	Total number			Yield of linear trimers **
	A	B	C	AB	BC	ABC	AA	A	B	C	
Number	33	13	21	7	8	40	1	82	68	69	59%

* Here NSA refers to non-specific assemblies. From our observations, we only found one non-specifically formed product (AA).

** The yield of linear trimers is calculated as: the number of ABC / the total number of B = 59%.

Movie S1.
Low-magnification video of monovalent droplets diffusing at capillary top surface (2D).

Movie S2.
Low-magnification video of divalent droplets diffusing at capillary top surface (2D).

Movie S3.
Low-magnification video of trivalent droplets diffusing at capillary top surface (2D).

Movie S4.
Long ‘alternating copolymers’ wiggling at capillary top surface (2D).

Movie S5.
Real-time capture of a linear trimer folding into a ‘triangle’.

Movie S6.
Real-time capture of a ‘triangle’ unfolding to a linear trimer.

References

1. Wu KT, *et al*. (2013) Kinetics of DNA-coated sticky particles. *Phys. Rev. E* 88:022304.

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