# Supplementary information

#### Fast synthesis of DNA origami single crystals at room temperature

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#### **Table of Contents**

#### **Section 1. Experimental Materials and Methods**

a. DNA functionalization of gold nanoparticles

- b. Design and synthesis of regular-octahedral DNA origami frames
- c. Encaging functionalized gold nanoparticles inside DNA origami frames
- d. Fabrication of DNA origami single crystals at room temperature
- e. Fabrication of DNA origami single crystals at outdoor temperature
- f. Linear fitting of  $T_{\rm m}$  variation curve with urea concentration
- g. Silica-coated DNA origami crystals.

#### Section 2. Electron Microscopy and Confocal Laser Scanning Microscope

- a. Sample preparation and transmission electron microscope (TEM)
- b. Sample preparation and scanning electron microscope (SEM)
- c. Sample preparation and confocal laser scanning microscope (CLSM)

Section 3. Small Angle X-ray Scattering (SAXS)

a. Experimental method and data analysis

#### **Section 4. Supplementary Figures**

#### Section 5. DNA Sequence

a. Staples of regular-octahedral DNA origami frames (PAGE purified)

b. Sticky ends of regular-octahedral DNA origami frames (ULTRAPAGE purified)

c. Inner strands of regular-octahedral DNA origami frames (ULTRAPAGE purified)

d. Thiolated DNA for AuNPs function (HPLC purified)

e. Sticky ends of regular-octahedral DNA origami frames-universality verification-1st system

f. Staples of elongated-octahedral DNA origami frames-universality verification-2<sup>nd</sup> system

g. Inner strands of elongated-octahedral DNA origami frames-universality verification-2<sup>nd</sup> system

h. Sticky ends of elongated-octahedral DNA origami frames-universality verification-2<sup>nd</sup> system

i. Sticky ends with hairpin DNA conformation of regular-octahedral DNA origami framesuniversality verification-3<sup>rd</sup> system (ULTRAPAGE purified)

**Section 6. Supplementary References** 

#### **Section 1. Experimental Materials and Methods**

#### a. DNA functionalization of gold nanoparticles

The thiol-modified single-strand DNA (ssDNA) sequences were purchased from Shanghai Sangon Biotech Co. Ltd, and the 10 nm gold nanoparticles were purchased from Ted Pella Inc. Prior to modifying the thiol-modified ssDNA on the surface of nanoparticles, we first reduced the thiol-modified ssDNA using TCEP (tris[2-carboxyethyl] phosphine) in an ice bath for 1.5 hour and then purified it using a size exclusion column (G-25, GE Healthcare). Then, we mixed the 10 nm gold nanoparticles with the purified ssDNA at a molar ratio of 1:300 and adjusted the phosphate buffer concentration in solution to 10 mM (pH = 7). The mixed solution was firstly rotated on a rotator for 1.5 hour. Afterward, we gradually introduced NaCl solution (2 M NaCl, 10 mM phosphate) into the mixture in batches until the final NaCl concentration reached 0.1 M. The mixed solution was then rotated on the rotator for 18 hours. After that, we removed excess reduced thiol-modified ssDNA from the mixed solution by centrifugation at 14500 rpm for 1 hour, and replaced the supernatant with buffer (10 mM phosphate, 0.1 M NaCl), repeating this centrifugation and replacement operation three times. The functionalized gold nanoparticles were stored in the refrigerator at 4 °C.

#### b. Design and synthesis of regular-octahedral DNA origami frames

Regular-octahedral DNA origami frame contained twelve edges, and each edge was composed of a 6-helix bundle (6HB) that had a length of 84 base pairs (~28.56 nm). Detailed design and synthesis process can be found in previous literature<sup>1</sup>.To synthesize the regular-octahedral DNA origami frames, we mixed M13mp18 scaffold DNA (Bayou Biolabs, LLC), DNA staples, sticky ends, and inner strands at a molar ratio of 1:10:10:7.5 in the buffer solution containing 1 mM EDTA, 12.5 mM magnesium acetate and 40 mM tris acetate. Thus, the final solution was composed of 10 nM M13mp18 scaffold DNA, 100 nM DNA staples, 100 nM sticky ends and 75 nM inner strands. This solution was firstly heated to 90 °C and then slowly cooled to room temperature over a period of approximately 23 hours. The synthesized regular-octahedral DNA origami frames were stored at room temperature.

#### c. Encaging functionalized gold nanoparticles inside DNA origami frames

The synthesized regular-octahedral DNA origami frames were firstly mixed with functionalized 10 nm gold nanoparticles at a molar ratio of 1:0.9. Then, the mixed solution was heated to 50 °C and slowly cooled to 20 °C at the rate of 2.4 °C /h, resulting in an efficiency of nearly 99.0% for loading the nanoparticles (Fig. S1b). The DNA origami frames encaged with functionalized gold nanoparticles were uniformly called DNA origami building blocks in this work.

#### d. Fabrication of DNA origami single crystals at room temperature

Firstly, we mixed two types of DNA origami building blocks at a molar ratio of 1:1, and then use urea solution (9.85 M urea, 1 mM EDTA, 12.5 mM magnesium acetate, 40 mM tris acetate) to adjust the urea concentration of the mixture to our requirement. The required urea concentration was determined based on the fitting curve of  $T_m$  (we firstly queried the room temperature from the thermometer, and then regarded this temperature as Tm to calculate the required urea concentration). The mixture was placed directly at room temperature for incubation. During the incubation, no any other operations were required. After the incubation, we used the urea-free buffer (1 mM EDTA, 12.5 mM magnesium acetate and 40 mM tris acetate) to replace the supernatant, and stored the samples at 4 °C.

#### e. Fabrication of DNA origami single crystals at outdoor temperature

Firstly, we mixed two types of DNA origami building blocks at a molar ratio of 1:1, and then use urea solution (9.85 M urea, 1 mM EDTA, 12.5 mM magnesium acetate, 40 mM tris acetate) to adjust the urea concentration of the mixture to our requirement. The required urea concentration was determined based on the fitting curve of  $T_m$  (we firstly queried the temperature fluctuations in the next two days from the weather forecast and determined the average temperature, and then regarded this average temperature as Tm to calculate the required urea concentration). The mixture was placed directly at outdoor temperature for incubation. During the incubation, no any other operations were required. After the incubation, we used the urea-free buffer (1 mM EDTA, 12.5 mM magnesium acetate and 40 mM tris acetate) to replace the supernatant, and stored the samples at 4 °C.

#### f. Linear fitting of T<sub>m</sub> variation curve with urea concentration

We used the linear equation to fit the curve of  $T_m$  changing with urea concentration, and the fitting formula is:

$$T_{m} = 40.8 \text{ }^{\circ}\text{C} + 1 - 4.64 \text{ }^{\circ}\text{C}/\text{M} \times C_{urea}$$

where  $T_m$  is the melting temperature,  $C_{urea}$  is the urea concentration, and 40.8 °C is the orignal  $T_m$  of the system without urea. The R<sup>2</sup> of this fitting curve is 0.997, confirming the fine fitting. This fitting curve can be used to calculate the required urea concentration for the crystallization of DNA building blocks in a specific environment by substituting the temperature parameters into  $T_m$  (for room temperature: taking the current temperature; for outdoor temperature: taking the average temperature).

#### g. Silica-coated DNA origami crystals

To intuitively observe the morphology and packing modes of the assemblies, we had to remove them from the salt solution and expose them to high-energy electron beam irradiation that unfortunately would cause the collapse of structure. Therefore, we encapsulated the bundles of regular-octahedral DNA origami frames and the connection part by a thin layer of silica to preserve their structural integrity. This silica-coated approach was referring to previous work<sup>2</sup>. Firstly, we diluted the concentration of magnesium ion in the solution to 7 mM, followed by adding 0.5-0.8  $\mu$ L TMAPS (N-trimethoxysilylpropyl-N, N, N-trimethylammonium chloride) at room temperature. Then, this mixture was shaken for 20 minutes at 400 rpm, added 0.4-0.7  $\mu$ L TEOS (tetraethyl orthosilicate), shaken for another 30 minutes at 500 rpm. Finally, after holding overnight, we replaced the supernatant of the sample with deionized water to interrupt the encapsulation process. The silica-coated samples were stored at room temperature.

# Section 2. Electron Microscopy and Confocal Laser Scanning Microscope a. Sample preparation and transmission electron microscope (TEM)

The carbon-coated grids were firstly discharged in a 0.26 mbar air atmosphere for 30 seconds using PELCO easiGlow (Ted Pella, Inc.). After shaking the sample solution evenly, 5  $\mu$ L solution was dropped onto this discharged carbon-coated grid. Five minutes later, residual solution was removed from the grid by filter papers, and then 5  $\mu$ l deionized water was dropped and removed immediately by filter papers to dissolve the deposited salt. This process was repeated twice. Next, 5  $\mu$ L 2% (w/v) uranyl acetate solution was dropped onto the grid to stain the sample for 10 seconds, and then removed by filter papers. This grid was then held to air dry for several minutes. The prepared carbon-coated grids covered by samples were then observed using JEOL JEM-1400 operated at 120 kV.

#### b. Sample preparation and scanning electron microscope (SEM)

The silica-coated samples were shaken evenly and then 5  $\mu$ L solution was taken out to be dropped onto the alcohol-washed silicon slice. To accelerate the drying process, we baked the droplet under infrared light until it was completely dry. This silicon slice was glued to the SEM sample stage using copper conductive tape without any metal deposition. The prepared samples were then observed using HITACHI Regulus 8100 operated at 1.5 kV with current of 2  $\mu$ A.

#### c. Sample preparation and confocal laser scanning microscope (CLSM)

Firstly, we shook the samples evenly, and then extracted 2  $\mu$ L mixture to drop into a confocal dish. This dish was then placed onto the sample stage of CLSM (Leica TCS SP8 STED) for subsequent observation. For higher image resolution, we utilized the bright field mode of CLSM (Leica TCS SP8 STED) to observe the mesoscopic morphology of our samples. The crystals did not exhibit their true colors under CLSM which highlighted the contrast, because the imaging principles was different from that of the ordinary optical microscope which can retain the authentic color (dark red) of the samples. However, this did not affect our data collection as we required the size and shape information of samples, and the advantages of CLSM was that it could help us to obtain data with more accurate size.

#### Section 3. Small Angle X-ray Scattering (SAXS)

#### a. Experimental method and data analysis

The SAXS experiments were carried out at the BL19U2 beamline at Shanghai Synchrotron Radiation Facility (SSRF). Before characterization, the samples were transformed from the tube into a glass capillary (inner diameter 1 mm), and then positioned them between the X-ray source and detector. The distance between the detector and samples was about 2-3 m. The 2D SAXS patterns were collected by detecting X-ray exposure, and the 1D scattering curves (scattering intensity I(q) as a function of scattering vector (q) were derived from the integration of 2D SAXS patterns. The scattering vector  $q = 4\frac{\pi}{\lambda}\sin\left(\frac{\theta}{2}\right)$ , where  $\lambda$  represents the wavelength of incident Xray and  $\theta$  is the scattering angle of the X-ray. To highlight the structural information of assembly more effectively, we extracted the structure factor S(q) from the scattering curve I(q) by deducting the corresponding particle form factor P(q), based on the formula  $I(q) = P(q) \times S(q)$ . The fitting of the 1D scattering curves was carried out with the PowderCell software by comparing the position of experimental scattering peaks with those of the corresponding standard model until obtaining the optimize result. Specifically, we first calculated the unit cell parameters based on the calculation formula for crystal plane spacing ( $(d = \frac{2\pi}{q}, d_{hkl} = \frac{1}{\sqrt{\frac{h^2 + k^2 + l^2}{a^2}}}$  (simple cubic system)), and then utilized software to obtain the 1D standard scattering spectra of the theoretical unit cell according to calculated lattice parameters. After that, we compared the positions of the standard scattering peaks with our experimental scattering curves to identify the ultimate structure and parameters. If

the theoretical model was not fit well, we finely adjusted the unit cell parameter appropriately to match the scattering peak position and height with those of the experimental 1D SAXS curves as closely as possible.

**Section 4. Supplementary Figures** 



**Fig. S1** Schematic illustration of DNA origami building block and the corresponding TEM characterization. (a) 6-helix-bundle edges of the regular-octahedron DNA origami were simplified into single-helix-bundle edge, and the four sticky ends extending from the vertices were simplified into a small color ball for the convenience of description. The inserted 10 nm gold nanoparticle was captured by 8 inner strands to fix the nanoparticle in the center of regular-octahedron DNA origami. (b) Statistical analysis of the number of gold nanoparticles inserted into the regular-octahedron DNA origami. (c) (d) TEM images of DNA origami building blocks (scale bars, 100 nm).



Fig. S2 Large-scaled CLSM images of the assemblies grown at room temperature for 48 h without urea.



**Fig. S3** (a) Large-scaled CLSM images of the assemblies grown at room temperature for 48 h with 3.8 M urea. (b) Crystal size distribution histogram of the crystals incubated at room temperature for 48 h with 3.8 M urea.



Fig. S4 The fitting result of SAXS curves of crystals incubated at room temperature.



**Fig. S5** The development of crystal size over time. (a) Representative CLSM images of crystals with different growth time and the responding crystal size frequency distribution. Scale bars, 10  $\mu$ m. Since the crystal with growth time less than six hours cannot distinguish the sides of cubic shape, we did not make size statistics, and we started to make size statistics from the eight hours. At each time point, the size of more than 70 crystals was counted. (b) development trend of average crystal size measured from CLSM images over time.



**Fig. S6** Comparison of DNA origami building blocks crystallized in 4.3 M, 3.5 M, and 2.8 M urea solution respectively at room temperature. (a) SAXS curves of samples after incubating with 4.3 M, 3.5 M, and 2.8 M urea respectively for 4 h at room temperature. From these SAXS curves, The DNA origami building blocks crystallized in 3.5 M urea solution, the DNA origami building blocks formed amorphous aggregates in 2.7 M urea solution, and the DNA origami building blocks were not assembled in 4.3 M urea solution. (b) SAXS curves of DNA origami building block assembling with time in 2.7 M urea solution. DNA origami building blocks cannot crystallize into ordered assemblies in 2.7 M urea solution. (c) SAXS curves of DNA origami building block assembling with time in 4.3 M urea solution. DNA origami building blocks remained monodisperse state within 18 hours.



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**Fig. S7** 1D SAXS curves of assemblies grown isothermally at fixed temperature with different urea concentration for 48 h.



**Fig. S8** SEM images of the silica-coated DNA origami crystal incubated isothermally at 24 °C with 3.8 M urea concentration for 48 h. (a) (b) Large-scaled SEM image showed that the crystals had cubic Wulff shape. (c) (f) Representative SEM images of the single DNA origami crystals (scale bars, 2  $\mu$ m). (d) (g) Enlarged SEM images of the region framed by red rectangle respectively in c and f, and the relative orientations between adjacent DOBs were identical (scale bars, 50 nm). (e) (h) Assembly models respectively corresponding to (d) and (g), in which the vertices of DNA origami building blocks are fully utilized for assembly.



**Fig. S9** SEM images of the silica-coated amorphous aggregation incubated isothermally at 24 °C with 2.0 M urea concentration for 48 h. (a) (b) Large-scaled SEM image showed that the assembly were amorphous with no distinguishable shape. (c) Representative SEM image of the assembly details showed that the arrangement of DOBs was disordered and the relative orientation between the adjacent DNA origami building blocks was chaotic (scale bar, 200 nm). (d) Enlarged SEM image of the region framed by red rectangle in (c) (scale bar, 50 nm). (e) Assembly model corresponding to the aggregate shown in (d), in which the vertices were not fully utilized for assembly.



**Fig. S10** Characterization of the liquid phase incubated isothermally at 24 °C with 5.0 M urea concentration for 48 h. (a) Images of the samples before and after 48-hour incubation, and there was no precipitation at the bottom of the tube after incubation. (b) Large-scaled TEM image of samples after 48-hour incubation, which showed that there was no connection between most of DNA origami building blocks (scale bar, 100 nm). (c) Closed-up TEM images of samples after 48-hour incubation (scale bar, 50 nm). (d) Characterization results of dynamic light scattering for samples before and after incubation (samples before incubation: blue rectangle; samples after incubation: red rectangle), which was a distribution histogram of number (y axis, %) versus aggregate size (x axis, nm).



Fig. S11 Universal validation of room-temperature synthesis approach. (a) The first system changed the sequence of the sticky ends extending from the vertices of regular-octahedral DNA origami framework. We anticipate that this system can be crystallized at room temperature (24 °C) with 4.0 M urea. (b) The original  $T_m$  of the first system measured by DLS, and the calculation of urea concentration required for crystallization at 24 °C. (c) The representative CLSM images of the first system after incubating at room temperature (24 °C) for 48 hours. Scale bar, 10  $\mu$ m. (d) The 1D SAXS curve of the first system after incubating at room temperature (24 °C) for 48 hours.



Fig. S12 Universal validation of room-temperature synthesis approach. (a) The second system changed the shape of DNA origami frameworks and the sequence of sticky ends at the same time. The DNA origami used in this system is elongated-octahedral DNA origami framework. We anticipate that this system can be crystallized at room temperature (24 °C) with 3.6 M urea. (b) The original  $T_m$  of the second system measured by DLS, and the calculation of urea concentration required for crystallization at 24 °C. (c) The representative CLSM images of the second system after incubating at room temperature (24 °C) for 48 hours. Scale bar, 10 µm. (d) The 1D SAXS curve of the second system after incubating at room temperature (24 °C) for 48 hours.



**Fig. S13** Universal validation of room-temperature synthesis approach. (a) The third system changed the flexible region of sticky ends into a hairpin DNA conformation, and the complementary base number was increased to 10 bases. We anticipate that this system can be crystallized at room temperature (24 °C) with 4.0 M urea. (b) The original  $T_m$  of the third system measured by DLS, and the calculation of urea concentration required for crystallization at 24 °C. (c) The representative TEM images of the third system after incubating at room temperature (24 °C) for 48 hours. Scale bar, 100 nm. (d) The 1D SAXS curve of the third system after incubating at room temperature (24 °C) for 48 hours.



Fig. S14 Large-scaled CLSM images of the assemblies grown at outdoor temperature for 48 h without urea.



**Fig. S15** (a) Large-scaled CLSM images of the assemblies grown at outdoor temperature for 48 h with 3.0 M urea. (b) Crystal size distribution histogram of the crystals incubated at outdoor temperature for 48 h with 3.0 M urea.



**Fig. S16** Crystallization of DNA origami building blocks at different outdoor conditions. (a) (d) (g) Temperature variation curves of 48 h outdoor temperature. (b) (e) (h) 1D SAXS curves and fitting curve (red) of assemblies incubated with 1.6 M (b), 2.0 M (e), and 3.6 M (h) urea, respectively, at outdoor temperature. (c) (f) (i) Representative CLSM image of the crystals incubated with 1.6 M (c), 2.0 M (f), and 3.6 M (i) urea, respectively, at outdoor temperature.



Fig. S17 Large-scaled CLSM images of the assemblies grown at simulated outdoor temperature for48 h without urea.



Fig. S18 Large-scaled CLSM images of the assemblies grown at simulated outdoor temperature for48 h with 2.8 M urea.

Section 5. DNA Sequence (all sequences are in order 5' to 3') a. Staples of regular-octahedral DNA origami frames (PAGE purified) Staple-1: TCAAAGCGAACCAGACCGTTTTATATAGTC Staple-2: GCTTTGAGGACTAAAGAGCAACGGGGAGTT Staple-3: GTAAATCGTCGCTATTGAATAACTCAAGAA Staple-4: AAGCCTTAAATCAAGACTTGCGGAGCAAAT Staple-5: ATTTTAAGAACTGGCTTGAATTATCAGTGA Staple-6: GTTAAAATTCGCATTATAAACGTAAACTAG Staple-7: AGCACCATTACCATTACAGCAAATGACGGA Staple-8: ATTGCGTAGATTTTCAAAACAGATTGTTTG Staple-9: TAACCTGTTTAGCTATTTTCGCATTCATTC Staple-10: GTCAGAGGGTAATTGAGAACACCAAAATAG Staple-11: CTCCAGCCAGCTTTCCCCCTCAGGACGTTGG Staple-12: GTCCACTATTAAAGAACCAGTTTTGGTTCC Staple-13: TAAAGGTGGCAACATAGTAGAAAATAATAA Staple-14: GATAAGTCCTGAACAACTGTTTAAAGAGAA Staple-15: GGTAATAGTAAAATGTAAGTTTTACACTAT Staple-16: TCAGAACCGCCACCCTCTCAGAGTATTAGC Staple-17: AAGGGAACCGAACTGAGCAGACGGTATCAT Staple-18: GTAAAGATTCAAAAGGCCTGAGTTGACCCT Staple-19: AGGCGTTAAATAAGAAGACCGTGTCGCAAG Staple-20: CAGGTCGACTCTAGAGCAAGCTTCAAGGCG Staple-21: CAGAGCCACCACCCTCTCAGAACTCGAGAG Staple-22: TTCACGTTGAAAATCTTGCGAATGGGATTT Staple-23: AAGTTTTAACGGGGGTCGGAGTGTAGAATGG Staple-24: TTGCGTATTGGGCGCCCGCGGGGGTGCGCTC Staple-25: GTCACCAGAGCCATGGTGAATTATCACCAATCAGAAAAGCCT Staple-26: GGACAGAGTTACTTTGTCGAAATCCGCGTGTATCACCGTACG Staple-27: CAACATGATTTACGAGCATGGAATAAGTAAGACGACAATAAA Staple-28 AACCAGACGCTACGTTAATAAAACGAACATACCACATTCAGG

Staple-29: TGACCTACTAGAAAAAGCCCCAGGCAAAGCAATTTCATCTTC Staple-30: TGCCGGAAGGGGACTCGTAACCGTGCATTATATTTTAGTTCT Staple-31: AGAACCCCAAATCACCATCTGCGGAATCGAATAAAAATTTTT Staple-32: GCTCCATTGTGTACCGTAACACTGAGTTAGTTAGCGTAACCT Staple-33: AGTACCGAATAGGAACCCAAACGGTGTAACCTCAGGAGGTTT Staple-34: CAGTTTGAATGTTTAGTATCATATGCGTAGAATCGCCATAGC Staple-35: AAGATTGTTTTTAACCAAGAAACCATCGACCCAAAAACAGG Staple-36: TCAGAGCGCCACCACATAATCAAAATCAGAACGAGTAGTATG Staple-37: GATGGTTGGGAAGAAAAATCCACCAGAAATAATTGGGCTTGA Staple-38: CTCCTTAACGTAGAAACCAATCAATAATTCATCGAGAACAGA Staple-39: AGACACCTTACGCAGAACTGGCATGATTTTCTGTCCAGACAA Staple-40: GCCAGCTAGGCGATAGCTTAGATTAAGACCTTTTTAACCTGT Staple-41: CCGACTTATTAGGAACGCCATCAAAAATGAGTAACAACCCCA Staple-42: GTCCAATAGCGAGAACCAGACGACGATATTCAACGCAAGGGA Staple-43: CCAAAATACAATATGATATTCAACCGTTAGGCTATCAGGTAA Staple-44: AACAGTACTTGAAAACATATGAGACGGGTCTTTTTTAATGGA Staple-45: TTTCACCGCATTAAAGTCGGGAAACCTGATTTGAATTACCCA Staple-46: GAGAATAGAGCCTTACCGTCTATCAAATGGAGCGGAATTAGA Staple-48: GCACCCAGCGTTTTTTATCCGGTATTCTAGGCGAATTATTCA Staple-49: GGAAGCGCCCACAAACAGTTAATGCCCCGACTCCTCAAGATA Staple-50: GTTTGCCTATTCACAGGCAGGTCAGACGCCACCACCACCACCC Staple-51: CGCGAGCTTAGTTTTTCCCAATTCTGCGCAAGTGTAAAGCCT Staple-52: AGAAGCAACCAAGCCAAAAGAATACACTAATGCCAAAACTCC Staple-53: ATTAAGTATAAAGCGGCAAGGCAAAGAAACTAATAGGGTACC Staple-54: CAGTGCCTACATGGGAATTTACCGTTCCACAAGTAAGCAGAT Staple-55: ATAAGGCGCCAAAAGTTGAGATTTAGGATAACGGACCAGTCA Staple-56: TGCTAAACAGATGAAGAAACCACCAGAATTTAAAAAAAGGCT Staple-57: CAGCCTTGGTTTTGTATTAAGAGGCTGACTGCCTATATCAGA Staple-58: CGGAATAATTCAACCCAGCGCCAAAGACTTATTTTAACGCAA

Staple-59: CGCCTGAATTACCCTAATCTTGACAAGACAGACCATGAAAGA Staple-60: ACGCGAGGCTACAACAGTACCTTTTACAAATCGCGCAGAGAA Staple-61: CAGCGAACATTAAAAGAGAGAGTACCTTTACTGAATATAATGAA Staple-62: GGACGTTTAATTTCGACGAGAAACACCACCACTAATGCAGAT Staple-63: AAAGCGCCAAAGTTTATCTTACCGAAGCCCAATAATGAGTAA Staple-64: GAGCTCGTTGTAAACGCCAGGGTTTTCCAAAGCAATAAAGCC Staple-65: AATTATTGTTTTCATGCCTTTAGCGTCAGATAGCACGGAAAC Staple-66: AAGTTTCAGACAGCCGGGATCGTCACCCTTCTGTAGCTCAAC Staple-67: ACAAAGAAATTTAGGTAGGGCTTAATTGTATACAACGGAATC Staple-68: AACAAAAATAACTAGGTCTGAGAGACTACGCTGAGTTTCCCT Staple-69: CATAACCTAAATCAACAGTTCAGAAAACGTCATAAGGATAGC Staple-70: CACGACGAATTCGTGTGGGCATCAATTCTTTAGCAAAATTACG Staple-71: CCTACCAACAGTAATTTTATCCTGAATCAAACAGCCATATGA Staple-72: GATTATAAAGAAACGCCAGTTACAAAATTTACCAACGTCAGA Staple-73: AGTAGATTGAAAAGAATCATGGTCATAGCCGGAAGCATAAGT Staple-74: TAGAATCCATAAATCATTTAACAATTTCTCCCGGCTTAGGTT Staple-75: AAAGGCCAAATATGTTAGAGCTTAATTGATTGCTCCATGAGG Staple-76: CCAAAAGGAAAGGACAACAGTTTCAGCGAATCATCATATTCC Staple-77: GAAATCGATAACCGGATACCGATAGTTGTATCAGCTCCAACG Staple-78: TGAATATTATCAAAATAATGGAAGGGTTAATATTTATCCCAA Staple-79: GAGGAAGCAGGATTCGGGTAAAATACGTAAAACACCCCCCAG Staple-80: GGTTGATTTTCCAGCAGACAGCCCTCATTCGTCACGGGATAG Staple-81: CAAGCCCCCACCCTTAGCCCGGAATAGGACGATCTAAAGTTT Staple-82: TGTAGATATTACGCGGCGATCGGTGCGGGCGCCATCTTCTGG Staple-83: CATCCTATTCAGCTAAAAGGTAAAGTAAAAGCAAGCCGTTT Staple-84: CAGCTCATATAAGCGTACCCCGGTTGATGTGTCGGATTCTCC Staple-85: CATGTCACAAACGGCATTAAATGTGAGCAATTCGCGTTAAAT Staple-86: AGCGTCACGTATAAGAATTGAGTTAAGCCCTTTTTAAGAAAG Staple-87: TATAAAGCATCGTAACCAAGTACCGCACCGGCTGTAATATCC Staple-88: ATAGCCCGCGAAAATAATTGTATCGGTTCGCCGACAATGAGT

Staple-89: AGACAGTTCATATAGGAGAAGCCTTTATAACATTGCCTGAGA Staple-90: AACAGGTCCCGAAATTGCATCAAAAAGATCTTTGATCATCAG Staple-91: ACTGCCCTTGCCCCGTTGCAGCAAGCGGCAACAGCTTTTCT Staple-92: TCAAAGGGAGATAGCCCTTATAAATCAAGACAACAACCATCG Staple-93: GTAATACGCAAACATGAGAGATCTACAACTAGCTGAGGCCGG Staple-94: GAGATAACATTAGAAGAATAACATAAAAAGGAAGGATTAGGA Staple-95: CAGATATTACCTGAATACCAAGTTACAATCGGGAGCTATTTT Staple-96: CATATAACTAATGAACACAACATACGAGCTGTTTCTTTGGGG Staple-97:

ATGTTTTGCTTTTGATCGGAACGAGGGTACTTTTTCTTTTGATAAGAGGTCATT Staple-98:

CTTCGCTGGGCGCAGACGACAGTATCGGGGCACCGTCGCCATTCAGGCTGCGCA Staple-99:

Staple-100:

TGTCGTCATAAGTACAGAACCGCCACCCATTTTCACAGTACAAACTACAACGCC Staple-101:

CGATTATAAGCGGAGACTTCAAATATCGCGGAAGCCTACGAAGGCACCAACCTA Staple-102:

AACATGTACGCGAGTGGTTTGAAATACCTAAACACATTCTTACCAGTATAAAGC Staple-103:

Staple-104:

GCCTTGAATCTTTTCCGGAACCGCCTCCCAGAGCCCAGAGCCGCCGCCAGCATT Staple-105:

CGCTGGTGCTTTCCTGAATCGGCCAACGAGGGTGGTGATTGCCCTTCACCGCCT Staple-106:

ACATAACTTGCCCTAACTTTAATCATTGCATTATAACAACATTATTACAGGTAG Staple-107:

TTATTTTACCGACAATGCAGAACGCGCGAAAAATCTTTCCTTATCATTCCAAG

Staple-108:

TTTCAATAGAAGGCAGCGAACCTCCCGATTAGTTGAAACAATAACGGATTCGCC Staple-109:

ATGACCACTCGTTTGGCTTTTGCAAAAGTTAGACTATATTCATTGAATCCCCCT Staple-111:

TCCAAATCTTCTGAATTATTTGCACGTAGGTTTAACGCTAACGAGCGTCTTTCC Staple-112:

GGGTTATTTAATTACAATATGTGAGTAATTAATAAGAGTCAATAGTGAATTT

b. Sticky ends of regular-octahedral DNA origami frames (ULTRAPAGE purified)

SE-A-1:

SE-A-2:

SE-A-3:

SE-A-4:

SE-A-5:

SE-A-6:

SE-A-7:

SE-A-8:

TTTATCCGTTA

SE-A-9:

SE-A-10:

TTTATCCGTTA

SE-A-11:

TTTTATCCGTTA

SE-A-12:

TTTTTATCCGTTA

SE-A-13:

TTTTATCCGTTA

SE-A-14:

SE-A-15:

SE-A-16:

SE-A-17:

SE-A-18:

TTTTATCCGTTA

SE-A-19:

SE-A-20:

TTTTATCCGTTA

SE-A-21:

TTTTATCCGTTA

SE-A-22:

TTTATCCGTTA

SE-A-23:

TTTTTATCCGTTA

SE-A-24:

TTTTATCCGTTA

SE-B-1:

SE-B-2:

SE-B-3:

SE-B-4:

TTTTTTAACGGAT

SE-B-5:

TTTTTAACGGAT

SE-B-6:

TTTTAACGGAT

SE-B-7:

TTTTTTAACGGAT

SE-B-8:

TTTTAACGGAT

SE-B-9:

TTTTTTAACGGAT

SE-B-10:

TTTTAACGGAT

SE-B-11:

TTTTTAACGGAT

SE-B-12:

SE-B-13:

SE-B-14:

SE-B-15:

TTTTAACGGAT

SE-B-16:

TTTTAACGGAT

SE-B-17:

TTTTTTAACGGAT

SE-B-18:

TTTTTAACGGAT

SE-B-19:

TTTTTTAACGGAT

SE-B-20:

TTTTTAACGGAT

SE-B-21:

SE-B-22:

## SE-B-23:

SE-B-24:

c. Inner strands of regular-octahedral DNA origami frames (ULTRAPAGE purified) Inner-1:

TTAGAGCGCAAGGCGCACCGTAATCAGTAGCGA

Inner-2:

ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTGTTCCCACGCGCAAAATGGTTGA GTGTTGTTCGTGGACTTGCTTTCGAGGTGAATTT

Inner-3:

ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTGTTGGCCGAAAGTCTCTCTTTTGA

TGATACAAGTGCCTTAAGAGCAAGAAACAATGA

Inner-4:

AATTAACTGCGCTAATTTCGGAACCTATTATTCT

Inner-5:

ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTGGGGGTGCCAGTTGAGACCAT

TAGATACAATTTTCACTGTGTGAAATTGTTATCC

Inner-6:

ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTGTTTCAGAGCTGGGTAAACGACG

GCCAGTGCGATCCCCGTAGTAGCATTAACATCCA

Inner-7:

ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTGTTGTGGGAAATCATATAAATATTT

AAATTGAATTTTTGTCTGGCCTTCCTGTAGCC

Inner-8:

# ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTGTTTGATTATCAACTTTACAACTA AAGGAATCCAAAAAGTTTGAGTAACATTATCAT

# d. Thiolated DNA for AuNPs function (HPLC purified)

Thiolated-DNA-1: GAAGTGATGGATGAT

# e. Sticky ends of regular-octahedral DNA origami frames-universality verification-1st system

# (ULTRAPAGE purified)

1<sup>st</sup> system-SE-A-1:

TTTTTATGTGGTA

1<sup>st</sup> system-SE-A-2:

TTTTATGTGGTA

1<sup>st</sup> system-SE-A-3:

TTTTATGTGGTA

1<sup>st</sup> system-SE-A-4:

TTTTTATGTGGTA

1<sup>st</sup> system-SE-A-5:

TTTTATGTGGTA

1<sup>st</sup> system-SE-A-6:

TTTATGTGGTA

1<sup>st</sup> system-SE-A-7:

TTTTTATGTGGTA

1<sup>st</sup> system-SE-A-8:

TTTATGTGGTA

1<sup>st</sup> system-SE-A-9:

TTTTTATGTGGTA

1<sup>st</sup> system-SE-A-10:

TTTATGTGGTA

1<sup>st</sup> system-SE-A-11:

TTTTATGTGGTA

1<sup>st</sup> system-SE-A-12:

TTTTTATGTGGTA

1<sup>st</sup> system-SE-A-13:

TTTTATGTGGTA

1<sup>st</sup> system-SE-A-14:

TTTTTATGTGGTA

1<sup>st</sup> system-SE-A-15:

TTTATGTGGTA

1<sup>st</sup> system-SE-A-16:

TTTATGTGGTA

1<sup>st</sup> system-SE-A-17:

TTTTTATGTGGTA

1<sup>st</sup> system-SE-A-18:

TTTTATGTGGTA

1<sup>st</sup> system-SE-A-19:

TTTTTATGTGGTA

1<sup>st</sup> system-SE-A-20:

TTTTATGTGGTA

1<sup>st</sup> system-SE-A-21:

TTTTATGTGGTA

1<sup>st</sup> system-SE-A-22:

TTTATGTGGTA

1<sup>st</sup> system-SE-A-23:

TTTTTATGTGGTA

1<sup>st</sup> system-SE-A-24:

TTTTATGTGGTA

1st system-SE-B-1:

TTTTTTACCACAT

1<sup>st</sup> system-SE-B-2:

TTTTTACCACAT

1<sup>st</sup> system-SE-B-3:

TTTTTACCACAT

1<sup>st</sup> system-SE-B-4:

TTTTTTACCACAT

1<sup>st</sup> system-SE-B-5:

TTTTTACCACAT

1<sup>st</sup> system-SE-B-6:

TTTTACCACAT

1<sup>st</sup> system-SE-B-7:

TTTTTTACCACAT

1<sup>st</sup> system-SE-B-8:

TTTTACCACAT

1<sup>st</sup> system-SE-B-9:

TTTTTTACCACAT

1<sup>st</sup> system-SE-B-10:

TTTTACCACAT

1<sup>st</sup> system-SE-B-11:

TTTTTACCACAT

1<sup>st</sup> system-SE-B-12:

TTTTTTACCACAT

1<sup>st</sup> system-SE-B-13:

TTTTTACCACAT

1<sup>st</sup> system-SE-B-14:

TTTTTTACCACAT

1<sup>st</sup> system-SE-B-15:

TTTTACCACAT

1<sup>st</sup> system-SE-B-16:

TTTTACCACAT

1<sup>st</sup> system-SE-B-17:

TTTTTTACCACAT

1<sup>st</sup> system-SE-B-18:

TTTTTACCACAT

1<sup>st</sup> system-SE-B-19:

TTTTTTACCACAT

1<sup>st</sup> system-SE-B-20:

TTTTTACCACAT

1<sup>st</sup> system-SE-B-21:

TTTTTACCACAT

1<sup>st</sup> system-SE-B-22:

TTTTACCACAT

1<sup>st</sup> system-SE-B-23:

TTTTTTACCACAT

1<sup>st</sup> system-SE-B-24:

# f. Staples of elongated-octahedral DNA origami frames-universality verification-2<sup>nd</sup> system (PAGE purified)

EL-octa-Staple-1: CTCGTTTACCAGACGACAACACTAAAGATT EL-octa-Staple-2: AAAAGGGACATTCTGGTCACACGTTGCAAC EL-octa-Staple-3: GCCACTACGAAGGCACGGGTAAAGCGAAAG EL-octa-Staple-4: TTGGGGCGCGAGCTGATTAGCTATTCCATA EL-octa-Staple-5: TTCAAATATATTTTAGAACGCGACCTCCGG EL-octa-Staple-6: CAATATAATCCTGATTGATGATGATGATTTAA EL-octa-Staple-7: CAGACTGTAGCGCGTTAGTTTGCCCAGTAG EL-octa-Staple-8: GTCCACTATTAAAGAACCAGTTTTGGTTCC EL-octa-Staple-9: GAATAATAATTTTTTCCAACTAATAACGAT EL-octa-Staple-10: GGCCGATTAAAGGGATCGGGAGCCCGCCGC EL-octa-Staple-11: GCCTCTTCGCTATTACAGGGCGAGCACCGC EL-octa-Staple-12: AAGCCAGAATGGAAAGAAATAAACAGAGCC EL-octa-Staple-13: CCAGACGACGACAATAGGTAAAGCTCAACA EL-octa-Staple-14: TACCCAAATCAACGTAAGAACCGACGGTCA EL-octa-Staple-15: TTGCGCTCACTGCCCGACTCACACATGGTC EL-octa-Staple-16: AATTACATTTAACAATTCAAGAAATTGCTT EL-octa-Staple-17: GCCATCAAAAATAATTTTTAACCTAATCAG EL-octa-Staple-18: AGTCAAATCACCATCAGAGAAAGTTTCAAC EL-octa-Staple-19: AACAAAGTCAGAGGGTTTAACTGTTATCCC EL-octa-Staple-20: TTATTTGTCACAATCACACCACGCAGT EL-octa-Staple-21: ATCTGGTCAGTTGGCACAAACCCAGTATTA EL-octa-Staple-22: TAAGTATAGCCCGGAAGTCGAGAAAACATG EL-octa-Staple-23: AGCGAACCAGACCGGATTAATTCGTCAGAA EL-octa-Staple-24: GACTTGCGGGAGGTTTTTTTAGCTTACCGC EL-octa-Staple-25: ACAGCATGCTCCATAGATTTGTATCATCCCCAGCGAAACGAA EL-octa-Staple-26: GAAATCGGCCCCCTACGGGGTCAGTGCCCTTTTGATCCAACG EL-octa-Staple-27: AACGGGTCCTGAACAAGAAAAATAATATCTTATCATTCCAAG EL-octa-Staple-28: AAAAGCCCTCAGGACGTTGGTGTAGATGGGGAACAGGCCTTC EL-octa-Staple-29: ATTAAATCAGCTTTCATCAACATTAAATTTGTTAAAATTCGC EL-octa-Staple-30: TACATTTAATAGTACATCCAATAAATCAAAGCTAACCAAAAA EL-octa-Staple-31: GCCCAATTTTGCCATAACGAGCGTCTTTGCACCCATTAAATC EL-octa-Staple-33: GTTGGGATGAAAGAGGACAGATGAACGGAGTAGATCATTAGA EL-octa-Staple-34: CTTTTTCAAAGAATACTCATCTTTGACCGCCTGATGAAATCC EL-octa-Staple-35: AAGCCTGCGTGCCAGCTGCATTAATGAAAAGCATAAAGTGTA EL-octa-Staple-36: TCAGTGATCATCAAGAACTGACCAACTTAGAAAAATCTACGT EL-octa-Staple-37: TAACAGTACCCTGTAGCCTCAGAGCATATACAGGCGCATCAA EL-octa-Staple-38: CTAATGCGAATATAAGAATCGCCATATTTACCGCACTCATCG EL-octa-Staple-39: ATAGCTGTTGCCCCCGGGCAACAGCTGAATTGGGCGTCGGGA EL-octa-Staple-40: GCCGCCATGTAGCGGGAAGGGAAGAAGAGAGAGCTTTCTGAAT EL-octa-Staple-41: GGAATTAAATGGAACTACCATATCAAAACGTCAGAGTAACAG EL-octa-Staple-42: TCTGAATTCATCATTTATCATTTTGCGGTAATACATGAATGG EL-octa-Staple-43: AGAGGCAATGAGGAAGGGTAGCAACGGCAGGTGTCAAATTCC EL-octa-Staple-44: CGTTCTATAGGTAATTTTAGAACCCTCAAGGATGAACGGTAA EL-octa-Staple-45: TTCTACTCGCAAATCAATTCTGCGAACGTGTTGTAATCGGTA EL-octa-Staple-47: GAGCCGGTCGTAAGAAAGCGGCCAACGCTGATCGTGCTCAAG EL-octa-Staple-48: AAACAGGAGATAACCCACAAGAATTGAGAGAGAATAACATAA EL-octa-Staple-49: GTGCATCACAACCCGTCGGATTCTCCGTGGCGCATCGTAACC EL-octa-Staple-50: CTAAAGTAGGCCGCACAATGACAACAACTGAATTTAAATCTC EL-octa-Staple-51: AATCCAACAAAAGAAAGTAAGCAGATAGAATAGCACGCTAAT EL-octa-Staple-52: TAACGTGAGAATCCGTGAGTGAATAACCACATAGCGATAGCT EL-octa-Staple-53: GGATTATTGACCTGAATACGTGGCACAGAACATCGTACCGAA EL-octa-Staple-54: GTACGCCCTTTCCTTACAGGGCGCGTACAGAGTCAATAGTGA EL-octa-Staple-55: ATCATTTCGAAAGGAGCGGGAATAGCCCGCGAAAAAGCGTCA

EL-octa-Staple-56: TTAATTGATATAATGCTGTGGAAGCCCGATTAGAGAAGGCGA EL-octa-Staple-57: ATCATAAACGAACTATGCGATTTTAAGAATGGTTTTGCTCAT EL-octa-Staple-58: AAGCATCGAGGAAGATATCTTTAGGAGCGAAGTATAAACAAT EL-octa-Staple-59: AAAGTATTCAAAAAGTCATAAATATTCAAAATGTTATCACCG EL-octa-Staple-60: GCAAGGAACTAGCAGAGAGTCTGGAGCATTTTTGAATTCAAC EL-octa-Staple-61: ATCAGAGGAAGCGCACGATTTTTTGTTTACGCAATAATAACG EL-octa-Staple-62: CACCATTACCACCCGCCTCCCTCAGAGCTAATCAAGCATTTT EL-octa-Staple-63: TTTGCTAAAAGCGTTTATTTTGTATCGGATACCATATGAAAT EL-octa-Staple-64: TAATGTGGCTGATAAATTATGCTATTTTCCGCAATGCCTGAG EL-octa-Staple-65: TAGATTAAATATATTGAGAAGTGTTTTTTGGACGAGCACGTA EL-octa-Staple-66: CGAGGAAAACGTCAAAAATGAAAATAGCTACAGAGCTAAAGA EL-octa-Staple-67: ATGTTAGTTATACACCGGAATCATAATTGACCGTGAATTCAT EL-octa-Staple-68: CAAAAGGGAGGCTTGCCACCCTCAGAACAACCCATAACTACA EL-octa-Staple-69: AAGATTAGTATTCTAAATCAGATATAGATATATTTTAAATAG EL-octa-Staple-70: AACCGATTTTATCAGCTTGCTTTCGAGGCATCGCCCACGCAT EL-octa-Staple-71: CAAAAAACGGAGTGTCTTTCCAGACGTTCTGAGGCTTGCAGG EL-octa-Staple-72: TCGGTCGAGTAAATGAATTTTCTGTATGGTCACCACGATAGC EL-octa-Staple-73: CAGACCAAAATTAAGTAGCCACCAGAACGGTTGACTTAGTAC EL-octa-Staple-74: CAGAGGCAAAGAACGGGTTTAGATAAGTATACCAGAAACCTA EL-octa-Staple-75: GTTTCAGAAGGCTCCAAAAGGAGCCTTTAACAACTTTCAACA EL-octa-Staple-76: AACCTGTGGGTGCCTGTGAAATTGTTATCAGCAAGCGGTCCA EL-octa-Staple-77: CAGAAGGAATAAGAGCAAGAAACAATGACCGAACAAAGTTAC EL-octa-Staple-78: TTTACAGTTAAAACACACTAAGCCCAATAAGAGGAGCTTTAC EL-octa-Staple-79: GTAGGGCGCAAGCCATCGGCTGTCTTTCCCCATCCTGTTCAG EL-octa-Staple-80: AAGTTTGTACATCGATTTTCAGGTTTAATTATTTGTTATACT EL-octa-Staple-81: ACTTGCCATAATCAACAGTACATAAATCAGATTTCTATTCAC EL-octa-Staple-82: TAATATTGTCTAAAGTTATGAGCGAGTATGATGAAAGCAACC EL-octa-Staple-83: ATGGTTTACATATAAGAAAATACATACAAACTGTTTAGTATC EL-octa-Staple-84: CGGTCATTAATCAGGCAAGGCCGGAAACGGAACCGCTCAGAT EL-octa-Staple-85: ACCCTTCTTACATTTGGAAATACCTACAATAAAAACCATTAC

EL-octa-Staple-86: CAGTTCAGAGAAGGATTAGTTTCGTCACTCAACTAATAACGC EL-octa-Staple-87: GGTCAGGAAAGACTATCAAAAAGATTAACACCTGCAGGTCGA EL-octa-Staple-88: TGAGCAAAATGGAAGTGAGGCCACCGAGTTAGTAACTATCGG EL-octa-Staple-89: AGAGTTGCCGCTCACAATTCCACAACTTTTGACCTGAAAT EL-octa-Staple-90: AGCACCGAGCCCCCTTGCCATCTTTTCACGCCACCCCACCCT EL-octa-Staple-91: CTTTTTTAAGAAGACAAAATCGCGCAGAACTCAAATAACATC EL-octa-Staple-92: AGGAATTACCTTGCAGTGCCACGCTGAGACTTTACTAGACGT EL-octa-Staple-93: CCAGTCAGAGTAGTAAATTGGGGCTTGAGACTGGCTCATTATA EL-octa-Staple-94: GCAAAGCGGATCCCACGACGGCCAGTGCGGGTAACTCCAACA EL-octa-Staple-95: AACACTGCAGAACCTTGCAAAAGAAGTTTAGATACATGCAAA EL-octa-Staple-96: CCACCCTAGGATTAGCGGGGTTTTGCTCGAGGTTTAGGGGGGT EL-octa-Staple-97: GCTGAGATATGGTTGCTTTAGTAGAAGAGGCGAATAATTACC EL-octa-Staple-98: AGTTTGAAAGCAAATATTTAAATTGTAAAGCCAGCAAATCTA EL-octa-Staple-99: GCTTAATAAAATCATAGAATCCTTGAAATTGCTTCAGGAACG EL-octa-Staple-100: TCAAAGGGAGATAGCCCTTATAAATCAAAGGCCCGTATAAAC EL-octa-Staple-101: TTCTGGTATGCAACAGCTTAATTGCTGACTCCTTTGGCGAAA AATAAGAAGAACGCGCCTGTTTATCAACATTTTCGAGCCAGT EL-octa-Staple-102: CTTAGGTGAGCCATGACGGAAATTATTCGCGACATCATCTTC EL-octa-Staple-103: AAATACCACTAGAAAAAGCTGCTGATGCAATTTAACCAAAGA EL-octa-Staple-104: TGAATACGGTAATACAATACTTCTTTGATAAAAGAGAAATTAC EL-octa-Staple-105: TGACCTAAAATCCATATAACTATATGTATATTATCACCGTCA EL-octa-Staple-106: EL-octa-Staple-107: CTGTAGCTTTTGTTCAGGAAGATTGTATGGGGACGACGACAG GGGGGATCAGGCTGACCAGGCAAAGCGCGAAGCTCAACATGT EL-octa-Staple-108: EL-octa-Staple-109: ACACCGCCTCGTATCATTTGAGGATTTAACTAACAAGTTGAA EL-octa-Staple-110: CTGGCCCGCGGGGAGAGGCGGTTTGCGTTTGCCCTTCACCGC TACTCAGAGTACCACTGAGACTCCTCAAGAAAACGAGAATGA EL-octa-Staple-111: EL-octa-Staple-112: AATAGTATTGAATCCCCCTCAAATGCTTTTGCCAGAGTACCG EL-octa-Staple-113: CAAAAGGATTAAAGGTGAAAAGGTGGCAACCAGCGTGGTTTG TTACCGTTTGGCCTCAGGAGGTTGAGGCAAGCGCTAGGGCGC EL-octa-Staple-114: TACATGGTTGAGTAACAGTGTCAGACGATCCAGTAACCGTCT EL-octa-Staple-115:

EL-octa-Staple-116:TTAAGTTCAAGCTTGCATGTTCGCCATTGTGCTGCAGTACCTEL-octa-Staple-117:TTATCCGGTATGCCGGAGAGGGGTAGCTAAACAAGAGAATCGCEL-octa-Staple-118:CTGACCTATAAGGCTTGCCCTGACGAGAGGCGCATAGGCTGGEL-octa-Staple-119:AACCAAGTAACAACGCCAACATGTAATTAACAAAGAAGGAGCEL-octa-Staple-120:CATCAGTTAGCATTGCAAGCCCAATAGGCGCCACCAACAAAEL-octa-Staple-121:

GAATACCATAAGAAATTAGACGGGAGAAAATTGAGATAGCTATCTTACCGAAGC EL-octa-Staple-122:

GCGACCTCGGAACGAGTTTCCATTAAACCAACCTAATTATACCAAGCGCGAAAC EL-octa-Staple-123:

TATCGGCCCAAAAAAAATCAGCTCATTTCGCGTCTAACGGCGGATTGACCGTAA EL-octa-Staple-124:

CATTATGTGATTCCGGTCAATAACCTGTAAAGGTGAAGGCAAAGAATTAGCAAA EL-octa-Staple-125:

AGAACAATTAATTGAAGTACCGACAAAAAACAACATAATTTACGAGCATGTAGA EL-octa-Staple-126:

ACGCCTGTGAGATTAGGCATAGTAAGAGCGATAAACTCAGAGCCACCACCCTCA EL-octa-Staple-127:

CGAACCAACGCTCAGGCAGATTCACCAGCCAACAGTTTTGAATGGCTATTAGTC EL-octa-Staple-129:

TAATAAAGGGAACCGAGTAATCTTGACAACAAAGCAATTTCAACTTTAATCATT EL-octa-Staple-130:

GAGTTAATTTGTCGAGAATAGAAAGGAAACGTTGACTTAAACAGCTTGATACCG EL-octa-Staple-132:

TCGACAACTGCAACTGAACCTCAAATATAATCAACACTAATAGATTAGAGCCGT EL-octa-Staple-133: CAGAGCCACCATTATAGCGACAGAATCATTCATCGAATCACCGGAACCAGAGCC EL-octa-Staple-134:

TACCTTTAGTAACAATTCCTGATTATCAGTTTGGACACGTAAAACAGAAATAAA EL-octa-Staple-135:

ATTTATCGCGCCGCCGTTAGAATCAGAGTTTAGACTGTAAATCGTCGCTATTAA EL-octa-Staple-136:

CCATAAATAAGAGGGGGGGGGATAAGTGCCTAGGTGTTAGACTGGATAGCGTCCAA

g. Inner strands of elongated-octahedral DNA origami frames-universality verification-2<sup>nd</sup> system (ULTRAPAGE purified)

EL-octa-inner-1:

GCGAGGCGTGAAGCCGCTACAATTTTATCCTGAA

EL-octa-inner-2:

AAAGGGTATATGATGAGATCTACAAAGGCTATC

EL-octa-inner-3:

ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTGTTGTTATATGCGCAAACGTAAAGAA

ACGCAAAGAATAGAATGATAAATAAGGCGTTAAA

EL-octa-inner-4:

ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTGTTCCGACTTTGGGTTAATCGCAA

GACAAAGTTAATTTTCAACCGATTGAGGGAGGG

EL-octa-inner-5:

TGTTGTTCGTGGACTGATACAGGAGTGTACTGG

EL-octa-inner-6:

ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTGTTGGCAAGGCATTGATGATATT

CACAAACCGCAGTCGACGGGGAAAGCCGGCGAA

EL-octa-inner-7:

ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTGTTCTCTAGAGGATTGCTCAAATA

# TCGCGTTAGCAAACGCCAGGGTTTTCCCAGTCA

EL-octa-inner-8:

ATCCATCACTTCATACTCTACGTTGTTGTTGTTGTTGTTGTTTTTAAATGCCGGAACGCAACT GTTGGGAGCCAGCTTGATAAGAGGTCATTTTTG

h. Sticky ends of elongated-octahedral DNA origami frames-universality verification-2<sup>nd</sup> system (ULTRAPAGE purified)

EL-octa-SE-A-1:

TTTTATCCGTTA

EL-octa-SE-A-2:

TTTTATCCGTTA

EL-octa-SE-A-3:

TTTTATCCGTTA

EL-octa-SE-A-4:

TTTTATCCGTTA

EL-octa-SE-A-5:

TTTTCATCAAGT

EL-octa-SE-A-6:

TTTTTCATCAAGT

EL-octa-SE-A-7:

TTTCATCAAGT

EL-octa-SE-A-8:

TTTTCATCAAGT

EL-octa-SE-A-9:

TTTTCATCAAGT

EL-octa-SE-A-10:

TTTTCATCAAGT

EL-octa-SE-A-11:

TTTTCATCAAGT

EL-octa-SE-A-12:

TTTCATCAAGT

EL-octa-SE-A-13:

TTTTCATCAAGT

EL-octa-SE-A-14:

TTTTTCATCAAGT

EL-octa-SE-A-15:

TTTTCATCAAGT

EL-octa-SE-A-16:

TTTTCATCAAGT

EL-octa-SE-A-17:

TTTCATCAAGT

EL-octa-SE-A-18:

TTTTCATCAAGT

EL-octa-SE-A-19:

TTTCATCAAGT

EL-octa-SE-A-20:

TTTCATCAAGT

EL-octa-SE-A-21:

TTTATCCGTTA

EL-octa-SE-A-22:

TTTTTATCCGTTA

EL-octa-SE-A-23:

TTTTATCCGTTA

EL-octa-SE-A-24:

TTTTATCCGTTA

EL-octa-SE-B-1:

TTTTTAACGGAT

EL-octa-SE-B-2:

TTTTTAACGGAT

EL-octa-SE-B-3:

TTTTTAACGGAT

EL-octa-SE-B-4:

TTTTTAACGGAT

EL-octa-SE-B-5:

TTTTACTTGATG

EL-octa-SE-B-6:

TTTTTACTTGATG

EL-octa-SE-B-7:

TTTACTTGATG

EL-octa-SE-B-8:

TTTTACTTGATG

EL-octa-SE-B-9:

TTTTACTTGATG

EL-octa-SE-B-10:

TTTTACTTGATG

EL-octa-SE-B-11:

TTTTACTTGATG

EL-octa-SE-B-12:

TTTACTTGATG

EL-octa-SE-B-13:

TTTTACTTGATG

EL-octa-SE-B-14:

TTTTTACTTGATG

EL-octa-SE-B-15:

TTTTACTTGATG

EL-octa-SE-B-16:

TTTTACTTGATG

EL-octa-SE-B-17:

TTTACTTGATG

EL-octa-SE-B-18:

TTTTACTTGATG

EL-octa-SE-B-19:

TTTACTTGATG

EL-octa-SE-B-20:

TTTACTTGATG

EL-octa-SE-B-21:

TTTTAACGGAT

EL-octa-SE-B-22:

TTTTTTAACGGAT

EL-octa-SE-B-23:

TTTTTAACGGAT

EL-octa-SE-B-24:

# TTTTTAACGGAT

i. Sticky ends with hairpin DNA conformation of regular-octahedral DNA origami framesuniversality verification-3<sup>rd</sup> system (ULTRAPAGE purified)

R-Octa-SE-hairpin-XY-A-1:

GGTAGCTATTTTAGAGAATCGATGAAAAACATTAAATGTGTAGTTTTACTCTTTTTTAGA

GTTTTTATCCGTTA

R-Octa-SE-hairpin-XY-A-2:

ATAAATCATACATAAATCGGTTGTACTGTGCTGGCATGCCTGTTTTACTCTTTTTTAGA

GTTTTTTATCCGTTA

R-Octa-SE-hairpin-XY-A-3:

ACTGTTGGGAAGCAGCTGGCGAAAGGATAGGTCAAGATCGCATTTTACTCTTTTTTA

GAGTTTTTTATCCGTTA

R-Octa-SE-hairpin-XY-A-4:

GTTTTTATCCGTTA

R-Octa-SE-hairpin-XY-A-5:

GACAGGAGGTTGAAACAAATAAATCCGCCCCCCCCGCCACCCTTTTACTCTTTTTAG

AGTTTTTTATCCGTTA

R-Octa-SE-hairpin-XY-A-6:

CAGAATCAAGTTTCGGCATTTTCGGTTAAATATATCACCAGTTTTTACTCTTTTTTAGA

GTTTTTTATCCGTTA

R-Octa-SE-hairpin-XY-A-7:

GTTTTTATCCGTTA

R-Octa-SE-hairpin-XY-A-8:

AATAGCAATAGCACCAGAAGGAAACCTAAAGCCACTGGTAATTTTTACTCTTTTTTAG

AGTTTTTTATCCGTTA

R-Octa-SE-hairpin-XY-A-9:

TGTAGCATTCCAACGTTAGTAAATGAAGTGCCGCGCCACCCTTTTTACTCTTTTTAG

AGTTTTTTATCCGTTA

R-Octa-SE-hairpin-XY-A-10:

GAAACATGAAAGCTCAGTACCAGGCGAAAAATGCTGAACAAATTTTACTCTTTTTTA

GAGTTTTTTATCCGTTA

R-Octa-SE-hairpin-XY-A-11:

AGAGCCTAATTTGATTTTTGTTTAAATCCTGAAATAAAGAATTTTACTCTTTTTTAGA

GTTTTTTATCCGTTA

R-Octa-SE-hairpin-XY-A-12:

GTTTTTTATCCGTTA

R-Octa-SE-hairpin-XY-A-13:

TTTGCGGATGGCCAACTAAAGTACGGGCTTGCAGCTACAGAGTTTTACTCTTTTTTAG

AGTTTTTTATCCGTTA

R-Octa-SE-hairpin-XY-A-14:

CTTAAACAGCTTATATATTCGGTCGCTTGATGGGGAACAAGATTTTACTCTTTTTTAGA

GTTTTTATCCGTTA

R-Octa-SE-hairpin-XY-A-15:

GGCCCTGAGAGAAGCAGGCGAAAATCATTGCGTAGAGGCGGTTTTTACTCTTTTTTA

GAGTTTTTTATCCGTTA

R-Octa-SE-hairpin-XY-A-16:

GCTCACAATTCCGTGAGCTAACTCACTGGAAGTAATGGTCAATTTTACTCTTTTTTAG

AGTTTTTTATCCGTTA

R-Octa-SE-hairpin-Z-A-1:

 ${\sf CAAATGCTTTAAAAAAATCAGGTCTTTAAGAGCAGCCAGAGGGTTCCGAGTCAATTAAG}$ 

ACTCCCTTGTATTGGTCA

R-Octa-SE-hairpin-Z-A-2:

AAAGATTCATCAGGAATTACGAGGCATGCTCATCCTTATGCGTTCCGAGTCAATTAAGA

CTCCCTTGTATTGGTCA

R-Octa-SE-hairpin-Z-A-3:

CTTCATCAAGAGAAATCAACGTAACAGAGATTTGTCAATCATTTCCGAGTCAATTAAG

ACTCCCTTGTATTGGTCA

R-Octa-SE-hairpin-Z-A-4:

AAACGAAAGAGGGCGAAACAAAGTACTGACTATATTCGAGCTTTCCGAGTCAATTAA

GACTCCCTTGTATTGGTCA

R-Octa-SE-hairpin-Z-A-5:

CAACGCTCAACAGCAGAGGCATTTTCAATCCAATGATAAATATTCCGAGTCAATTAAG

ACTCCCTTGTATTGGTCA

R-Octa-SE-hairpin-Z-A-6:

ATCAAAATCATATATGTAAATGCTGAACAAACACTTGCTTCTTTCCGAGTCAATTAAGA

CTCCCTTGTATTGGTCA

R-Octa-SE-hairpin-Z-A-7:

TGATTGCTTTGAGCAAAAGAAGATGAAATAGCAGAGGTTTTGTTCCGAGTCAATTAAG

ACTCCCTTGTATTGGTCA

R-Octa-SE-hairpin-Z-A-8:

AACGGGTATTAAGGAATCATTACCGCCAGTAATTCAACAATATTCCGAGTCAATTAAGA

CTCCCTTGTATTGGTCA

R-Octa-SE-hairpin-XY-B-1:

GGTAGCTATTTTAGAGAATCGATGAAAACATTAAATGTGTAGTTTTAGCTATTTTTTAG

CTTTTTTAACGGAT

R-Octa-SE-hairpin-XY-B-2:

ATAAATCATACATAAATCGGTTGTACTGTGCTGGCATGCCTGTTTTAGCTATTTTTTAG

CTTTTTTAACGGAT

R-Octa-SE-hairpin-XY-B-3:

ACTGTTGGGAAGCAGCTGGCGAAAGGATAGGTCAAGATCGCATTTTAGCTATTTTTTA

GCTTTTTTAACGGAT

R-Octa-SE-hairpin-XY-B-4:

AGCTTTCATCAACGGATTGACCGTAAAATCGTATAATATTTTTTTAGCTATTTTTTAGC

TTTTTTAACGGAT

R-Octa-SE-hairpin-XY-B-5:

GCTTTTTTAACGGAT

R-Octa-SE-hairpin-XY-B-6:

 ${\sf CAGAATCAAGTTTCGGCATTTTCGGTTAAATATATCACCAGTTTTTAGCTATTTTTTAG}$ 

CTTTTTTAACGGAT

R-Octa-SE-hairpin-XY-B-7:

CTTTTTTAACGGAT

R-Octa-SE-hairpin-XY-B-8:

AATAGCAATAGCACCAGAAGGAAACCTAAAGCCACTGGTAATTTTTAGCTATTTTTTA

GCTTTTTTTAACGGAT

R-Octa-SE-hairpin-XY-B-9:

TGTAGCATTCCAACGTTAGTAAATGAAGTGCCGCGCCACCCTTTTTAGCTATTTTTTA

GCTTTTTTTAACGGAT

R-Octa-SE-hairpin-XY-B-10:

GAAACATGAAAGCTCAGTACCAGGCGAAAAATGCTGAACAAATTTTAGCTATTTTTT

AGCTTTTTTTAACGGAT

R-Octa-SE-hairpin-XY-B-11:

AGAGCCTAATTTGATTTTTGTTTAAATCCTGAAATAAAGAATTTTAGCTATTTTTTAG

CTTTTTTAACGGAT

R-Octa-SE-hairpin-XY-B-12:

TTTTTTAACGGAT

R-Octa-SE-hairpin-XY-B-13:

TTTGCGGATGGCCAACTAAAGTACGGGCTTGCAGCTACAGAGTTTTAGCTATTTTTTA

GCTTTTTTTAACGGAT

R-Octa-SE-hairpin-XY-B-14:

CTTTTTTAACGGAT

R-Octa-SE-hairpin-XY-B-15:

GGCCCTGAGAGAAGCAGGCGAAAATCATTGCGTAGAGGCGGTTTTTAGCTATTTTTT

AGCTTTTTTAACGGAT

R-Octa-SE-hairpin-XY-B-16:

GCTTTTTTTAACGGAT

R-Octa-SE-hairpin-Z-B-1:

CAAATGCTTTAAAAAATCAGGTCTTTAAGAGCAGCCAGAGGGTTTTGGATGTTTTTTC

ATCCCCCCTGACCAATAC

R-Octa-SE-hairpin-Z-B-2:

AAAGATTCATCAGGAATTACGAGGCATGCTCATCCTTATGCGTTTTGGATGTTTTTCAT

CCCCCCTGACCAATAC

R-Octa-SE-hairpin-Z-B-3:

CTTCATCAAGAGAAATCAACGTAACAGAGATTTGTCAATCATTTTTGGATGTTTTTTCA

TCCCCCTGACCAATAC

R-Octa-SE-hairpin-Z-B-4:

AAACGAAAGAGGGCGAAACAAAGTACTGACTATATTCGAGCTTTTTGGATGTTTTTTC

ATCCCCCCTGACCAATAC

R-Octa-SE-hairpin-Z-B-5:

CAACGCTCAACAGCAGAGGCATTTTCAATCCAATGATAAATATTTTGGATGTTTTTTCA

TCCCCCTGACCAATAC

R-Octa-SE-hairpin-Z-B-6:

ATCAAAATCATATATGTAAATGCTGAACAAACACTTGCTTCTTTTGGATGTTTTTCAT

CCCCCCTGACCAATAC

R-Octa-SE-hairpin-Z-B-7:

TGATTGCTTTGAGCAAAAGAAGAAGAAGAAGAAGAAGAAGAGGGTTTTGTTTTGGATGTTTTTCA

TCCCCCCTGACCAATAC

R-Octa-SE-hairpin-Z-B-8:

AACGGGTATTAAGGAATCATTACCGCCAGTAATTCAACAATATTTTGGATGTTTTTTCAT CCCCCCTGACCAATAC

# Section 6. Supplementary References

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