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Electronic Supplementary Information

Nanoscale patterning of polymers on DNA-origami

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CHEMICALS

Diethyl ether (Honeywell), petrolium ether (30-40 °C, Fisher Scientific), dichloromethane (Fisher Scientific), TEMED (Roth, >98.5%, p a), GeneRuler 1kb DNA ladder (Thermofisher), GeneRuler Ultra Low Range DNA Ladder (Thermofisher), tris-borate-EDTA buffer (Sigma Aldrich, 10x concentrate), nuclease-free water (QIAGEN), *N,N*-diisopropylethylamine (Roth, >99,5%), chloroform-d (Sigma Aldrich, 99.8 atom %), deuterium oxide (Sigma Aldrich, 99.9 atom %), 1,4-dioxane (Sigma Aldrich, anhydrous, 99.8%), dimethyl form amide (ACROS, extra dry, 99.8%), 4-cyano-4-(phenylcarbonothioylthio)pentanoic acid *N*-succinimidyl ester (Sigma Aldrich) and 2-(dodecylthiocarbonothioylthio)-2-methylpropionsöure-*N*-hydroxysuccinimidester (Sigma Aldrich) were used as received.

2,2'-Azobis(2-methylpropionitrile) (Fluka analytics, >98%) was recrystallized in methanol prior usage.

Poly(ethylene glycol) methyl ether methacrylate (PEGMA, Sigma Aldrich, average M_n = 300 g/mol, stabilized with 100 ppm MeHQ and 300 ppm BHT), 2-hydroxyethyl acrylate (HEA, Acros, 97%, stabilized), *N,N*-dimethylacrylamide (DMA, Sigma Aldrich, 99%, stabilized with 500 ppm MeHQ) were purified prior polymerization by removing the stabiliser with a small column filled with alumina.

N-Isopropyl acrylamide (NIPAM, TCI, >98%, stabilized with MeHQ) and diacetone acrylamide (DAAM, Alfa Aesar, 99%,) were purified prior polymerization by dissolving in dioxan and removing the stabiliser with a small column filled with alumina.

PROCEDURES AND METHODS

POLYMERIZATION

For the polymerization of homo polymers or the first block of the block copolymers, the monomer, CTA and AIBN were dissolved in the polymerization solvent, purged with argon for 45-90 min and heated up to the respective temperature. The ratio of initiator to CTA was 1:10. After the reaction time, the reaction mixture was cooled down with an ice bath and a precipitate was formed in the respective precipitation solvent (Table S3). The collected solid was again dissolved and precipitated twice. The obtained solid was dried under vacuum.

The second block was obtained by dissolving the first block (macro CTA) in the polymerization solvent, adding AIBN and monomer, purged with argon for 45-90 min and heated up to respective temperature. After the reaction time, the reaction solution was cooled with an ice bath and precipitated in the precipitation solvent. The obtained solid was collected and dried under vacuum.

Table S3 shows the respective solvents, reagents and reaction parameters for the respective polymerization.

To calculate the amounts of monomer, initiator and CTA formula 1 was used:

$$\bar{X}_n = \frac{p[M]_0}{(p'[RAFT]_0 + 2fp''[I]_0)} \quad (1)$$

Where $[RAFT]_0$, $[I]_0$ and $[M]_0$ are the starting concentrations of CTA, initiator and monomer. The fractional conversions of the monomer, CTA and initiator are p, p' and p'', here set to 1. f is the functionality of the initiator and $\bar{X}_n$ is the degree of polymerization. The scale for the polymerization was 0.5 g to 1 g of monomer.

CTA REMOVAL

The obtained polymer was dissolved in dioxane and an excess of AIBN was added. The reaction solution was heated up to 80 °C. After the reaction time, the reaction solution was cooled in an ice bath, and the polymer was participated in the respective precipitation solvent (Table S3). The obtained polymer was dried under vacuum and analysed with SEC (DMF, PMMA standard) and ^{1}H-NMR (300 MHz).

CONJUGATION REACTION

For a typical conjugation reaction, 5'amino-oligonucleotide (StA^c: NH_2-TTTTCTCTACCACCTACTA or StE^c: NH_2-CAGTCAGTCAGTCAGTCAGT) (10 nmol), polymer (50 equiv.) and DIPEA (200 equiv.) were mixed in the respective reaction solvent, to a total volume of 80 µL and shacken for 44-69 h at room temperature. After the reaction time, 1 µL of the reaction solution was diluted and analysed with PAGE.

The obtained reaction solution was purified via spin filtration (Amicon Ultra-6 mL Centrifugal Filters MWCO 10k or 30k) by adding 5 mL to the reaction solution and centrifuge for 1 h. This was repeated 10 times.

ORIGAMI SYNTHESIS

DNA origami nanostructures were prepared by mixing the respective staple strands (8 equiv.), folding strands (16 equiv.) and scaffold DNA (M13mp18) in origami buffer (1 mM Na_2EDTA, 5 mM NaCl, 5 mM TRIS, 12 mM $MgCl_2$ pH 8) and a temperature program run, starting at 70 °C and cooling down to 20 °C over 2 h (0.5 °C/min to 35 °C, 1 °C/min to 20 °C). The obtained DNA origami structures were purified by precipitation from PEG solution (15% PEG_{8000}, 5 mM TRIS buffer, 1 mM Na_2EDTA buffer, 0.505 M NaCl) with a 1:1 reaction solution to PEG solution ratio. The mixtures were centrifuged for 25 min at 12 xg at room temperature. The supernatant was removed, the DNA origami was redissolved in origami buffer and precipitated again from PEG solution. This procedure was repeated twice. The concentration was determined by measuring the absorption at 260 nm with Spark® 20M with Nanoquant plate™. The DNA origami were stored at 4 °C.

Table S 1: DNA-origami used in the experiments. For StA, the respective staple strand was elongated with the StA sequence. For StE, the respective staple strand was elongated with the StE sequence. To create DNA-origami tubes, the normal staple strand (number) was changed to the respective folding strand listed in Table S9.

Origami	Sequences different then staple strand or folding strand:
O_0	StA: 53-60; 63-74; 77-98; 158-179; 182-203 Folding strand (for tube): 1, 25, 27, 28, 51, 52, 75, 76, 99, 100, 111, 132, 133, 156, 157, 180, 181, 204, 205, 216
O_4	StA: 31-50, 81-98, 113, 115, 117, 118, 120, 122, 124, 125, 127, 129, 131, 134, 136, 138, 140, 142, 144, 146, 148, 150, 152, 154, 182-200 Folding strand (for tube): 1, 25, 27, 28, 51, 52, 75, 76, 99, 100, 111, 132, 133, 156, 157, 180, 181, 204, 205, 216
O_6	StA: 13-15, 17, 19, 37-44, 46, 59-70, 72, 81, 83, 85, 87, 89, 91, 93, 95, 119-122, 124, 140-147, 148, 162-173, 175, 184, 186-188, 190, 192, 194, 196, 198
O_8	StA: 161, 163, 165, 167, 169, 171, 173, 175, 177, 179, 182-200 StE: 56, 58, 60, 62, 64, 66, 68, 70, 72, 74, 77, 79, 81-98 Folding strand (for tube): 1, 25, 27, 28, 51, 52, 75, 76, 99, 100, 111, 132, 133, 156, 157, 180, 181, 204, 205, 216

ORIGAMI ANNEALING

To anneal the DNA-polymer conjugates to DNA origami, DNA origami (10 nM, final concentration) and DNA-polymer conjugate (50 equiv.) were mixed in origami buffer (1 mM Na_2EDTA, 5 mM NaCl, 5 mM TRIS, 12 mM $MgCl_2$ pH 8) to a total volume of 40 µL. The sample was heated up to 37 °C for 1 h and cooled to 30 °C and kept at 30 °C overnight. The obtained reaction solution was purified via spin filtration (Amicon Ultra-0.5 mL Centrifugal Filters MWCO 100K) by adding 400 µL origami buffer to the reaction solution and centrifuge 5 min at 5xg. The filtrate was removed and the purification was repeated two more times. The supernatant was collected, and the concentration was determined by measuring the absorption at 260 nm with Spark® 20M with Nanoquant plate™.

PAGE

The PAGE gel (15%) was manufactured by mixing 40% acrylamide/bis-acrylamide solution 37.5:1 (5.63 mL), 10x tris/borate/EDTA buffer (TBE buffer) (1.5 mL), water (7.9 mL), tetramethylethylenediamine (TEMED) (7.5 µL and 10% ammonium persulfate (APS) solution (75 µL) and casting the gel.

For monitoring the conjugation reaction via PAGE, 1 µL of diluted reaction solution (1:12.5, 10 fmol DNA) was hybridized with the complementary Rh6G-DNA sequence (100 µM, 20 fmol, 2 equiv.) in 10x origami buffer (0.5 µL) and nuclease-free water (1.5 µL) to a total volume of 5 µL. The obtained solution was heated to 35°C for 30 min (hybridization). The reaction solution was the cooled down to room temperature, mixed with loading dye (1.7 µL, 6x Thermo Fisher) and nuclease-free water

(3.3 µL) to a total volume of 10 µL and loaded onto the gel. The gel was run fist at 100 V for 10 min and then at 150 V for 50 min on a Cell SureLockTM mini-cell electrophoresis system from Thermo Fisher using 0.5 × TBE buffer as the running buffer (44.5 mM Tris-Borate, 1 mM EDTA). Generuler ultra low range DNA ladder (Thermo Fisher) was used as the DNA ladder. Controls contain 5'amino-oligonucleotide and polymer (50 equiv.) to show that the polymer does not entangle with DNA. The gels are stained with SYBR Gold (1x, 50 mL) for 45 min at room temperature. The images were taken with G:BOX Chemi Gel Doc System from Syngene.

AGAROSE

Agarose gels (1% TBE ethidium bromide (EtBr), BioRad, ReadyAgarose Precast Gel) were used as received.

For analysing DNA origami via agarose gel electrophoresis, origami solution (10 fmol), loading dye (6x Thermo Fisher) and 1x origami buffer (1 mM Na_2EDTA, 5 mM NaCl, 5 mM TRIS, 12 mM $MgCl_2$ pH 8) with a total volume of 10 µL were loaded on the gel. The electrophoresis was conducted at 90 V for 60 min at 4 °C. The gels were stained with SYBR Gold for 1 h. The images were taken with G:BOX Chemi Gel Doc System from Syngene.

AFM

For imaging the DNA origami architectures, Bruker Dimension FastScan Bio^{TM} atomic force microscope was used in the liquid state, which was operated in PeakForce mode. FastScan-D tips from Burker with a nominal spring constant of 0.25 Nm^{-1} were used.

For sample preparation, origami solution (40 µL, 0.5-2 nM in origami buffer) was added to a circular mica substrate (20 mm) and incubated for 10-15 min. The excess liquid was removed and 300 µL origami buffer was added to the mica to measure in liquid. Images were analysed with NanoScope Analysis 1.9.

GPC

GPC experiments were performed on a PSS SECurity instrument comprising an auto sampler, a column oven with three GRAM columns (10^3, 10^3 and 10^2 Å, 300 × 8 mm, 10 µm particle size) and a RI as well as an UV detector (Agilent Technologies 1260 Infinity). DMF containing 1 g/L lithium bromide was used as the eluent at a flowrate of 1 mL/min. Poly(methyl methacrylate) (1600 kDa–800 Da) served as the calibration standard for molecular weight measurements. The samples were filtered (0.4 µm) prior to injection. The data were fitted with OriginPro 2021.

NMR

NMR spectra were recorded on a Burker Avance (300 MHz) NMR spectrometer, using solvent signals of deuterated chloroform (δ=7.26) or deuterated water (δ=4.80) as reference. The data were processed with MestReNova 14.2.1.

RESULTS AND DISCUSSION

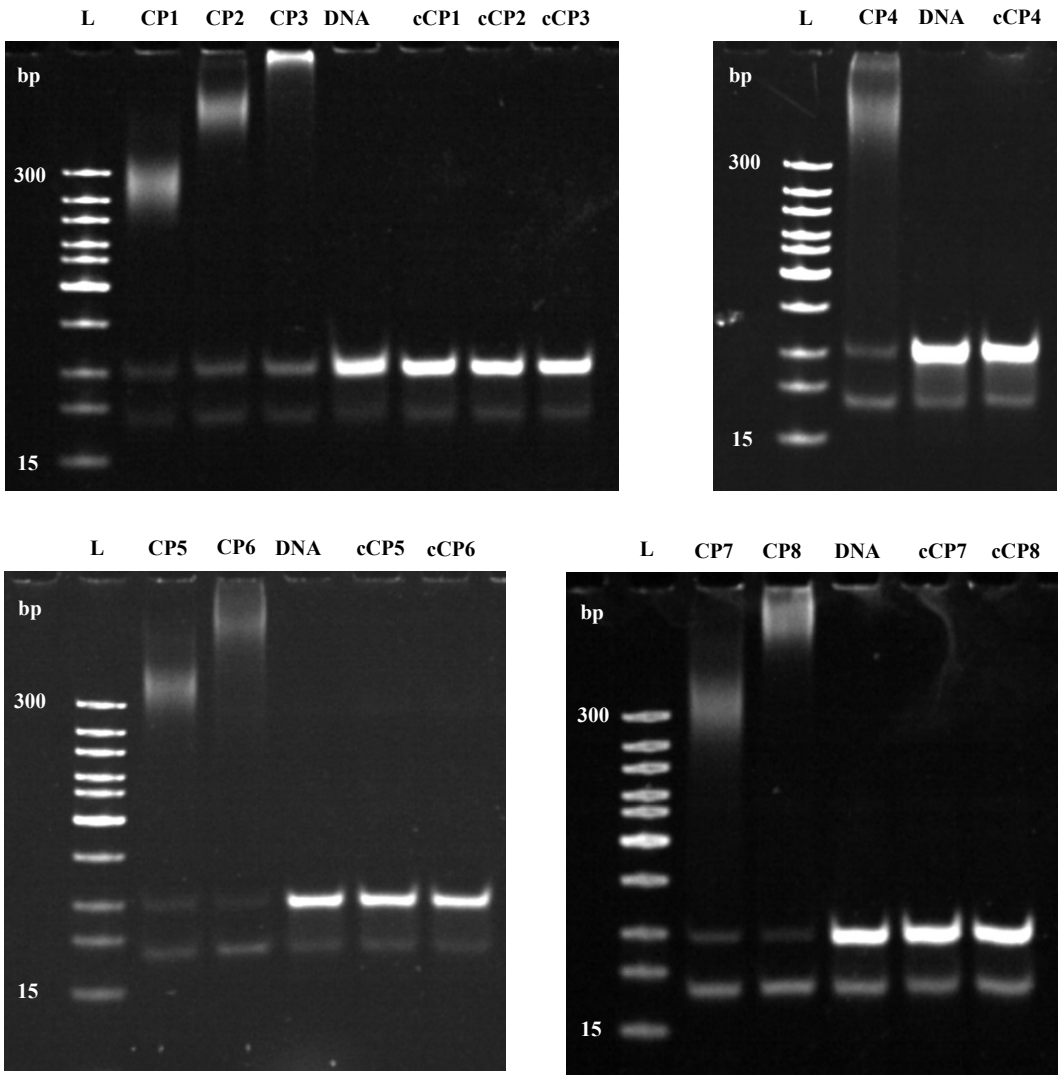

Figure S 1: PAGE gel (15%) of conjugation reaction of P1-P8 with 5'amino oligonucleotide and controls, stained with SYBR Gold. The controls contain the same amount of the respective polymer and 5'amino oligonucleotide without DIPEA and without reaction time to show that the oligonucleotide is not entangling with the respective polymer. L: DNA ladder; CP: respective conjugation reaction solution; cCP: control of respective conjugation reaction.

Table S 2: Conversion determination using the software ImageJ to analyse intensity and calculate conversion of the respective conjugation reaction. Used PAGE gels are shown in Figure 2 and S1.

	Intensity I	Intensity II	Conversion I	Conversion II
DNA	71.85	74.80		
CP1	13.55	15.43	81%	93%
CP2	18.05	20.04	75%	88%
CP3	22.51	24.55	69%	80%

	Intensity I	Intensity II	Conversion I	Conversion II
DNA	99.80	87.78		
CP4	28.57	22.91	71%	94%

	Intensity I	Intensity II	Conversion I	Conversion II
DNA	56.53	52.26		
CP5	16.71	13.62	71%	93%
CP6	15.46	12.58	73%	97%

	Intensity I	Intensity II	Conversion I	Conversion II
DNA	54.07	57.79		
CP7	5.83	6.95	89%	94%
CP8	4.77	5.79	91%	96%

Table S 3: Overview of the synthesized polymers, polymerization parameters and purification solvents.

Polymer	M_w (SEC)	Đ (SEC)	Amount I. Block (SEC)	Used CTA	Polym. solvent	Precipitation solvent	Reaction Temp.	Reaction time
P(DMA) (P1, P2, P3)	9649	1.08	/	NHS-DDMAT	dioxane	diethyl ether	70 °C	18 h
	22125	1.08					70 °C	18 h
	48637	1.27					65 °C	4 h
P(PEGMA) (P4)	21090	1.19	/	NHS-CPADB	dioxane	diethyl ether (-20 °C, phase separation)	70 °C	20.5 h
P(NIPAM) (P5)	17078	1.11	/	NHS-DDMAT	dioxane	diethyl ether	75 °C	19 h
P(NIPAM-*b*-DMA) (P6)	30445	1.19	43%	NHS-DDMAT	dioxane	diethyl ether	I. Block: 70 °C II. Block: 65 °C	15 h 17 h
P(HEA) (P7)	23264	1.27	/	NHS-DDMAT	DMF	diethyl ether	70 °C	16 h
P(DAAM-*b*-DMA) (P8)	26013	1.20	29%	NHS-DDMAT	dioxane	petrol ether	I. Block: 70 °C II. Block: 55 °C	17 h 21 h

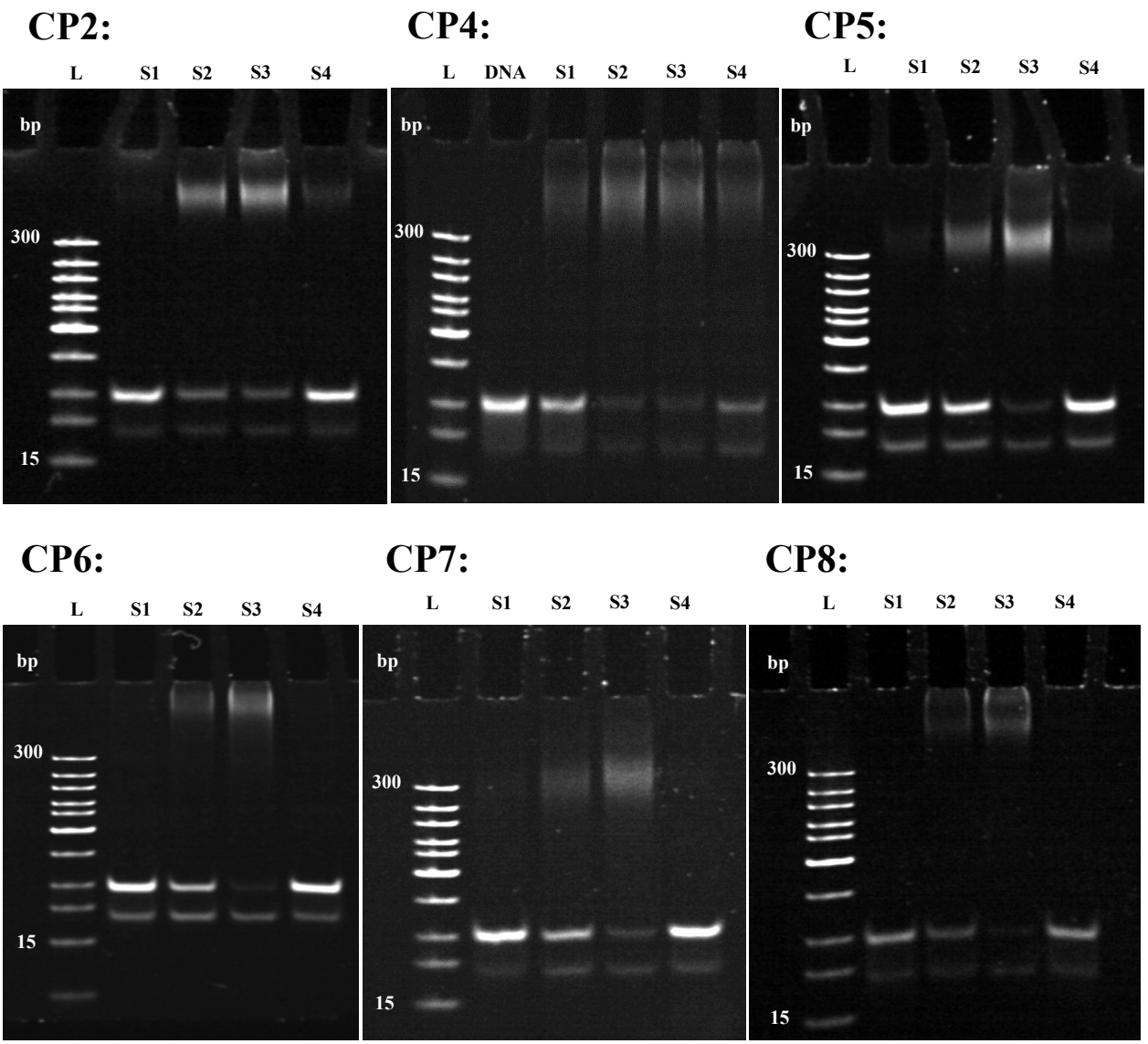

Figure S 2: PAGE gels (15%) of the respective polymer conjugation reactions accomplished in different solvent mixtures; S1 water, S2 DMF/water (1:1), S3 DMF/water (3:1) and S4 DMF. Stained with SYBR Gold.

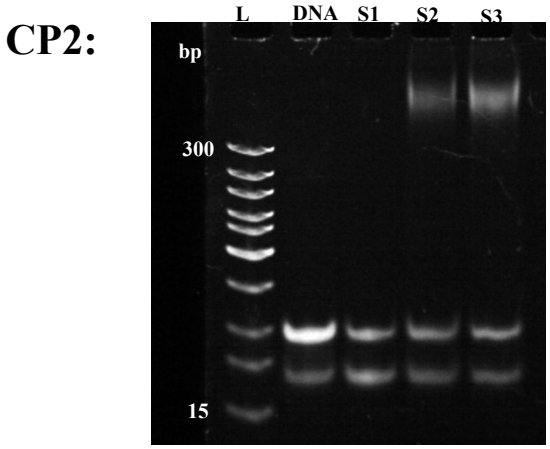

Figure S 3: PAGE gels (15%) of the respective polymer conjugation reaction of P2 accomplished in different solvent mixtures of ACN; S1 ACN, S2 ACN/water (1:1), S3 ACN/water (3:1). Stained with SYBR Gold.

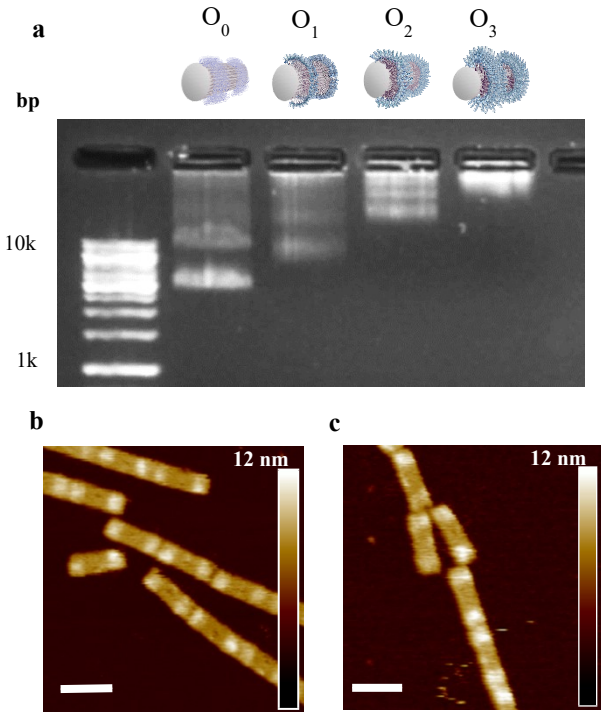

Figure S 4: c) Monitoring of the coated and uncoated DNA-origami (O_0-O_3) containing StA with CP1-CP3 by 1% agarose gel, stained with SYBR Gold. L: DNA ladder; O_0: uncoated DNA-origami; $O_{1\text{-}3}$: Coated DNA-origami (from left to right). b) Monitored DNA-origami (O_1) coated with CP1 via AFM. Scale bar = 80 nm c) Monitored DNA-origami (O_2) coated with CP2 via AFM. Scale bar = 80 nm.

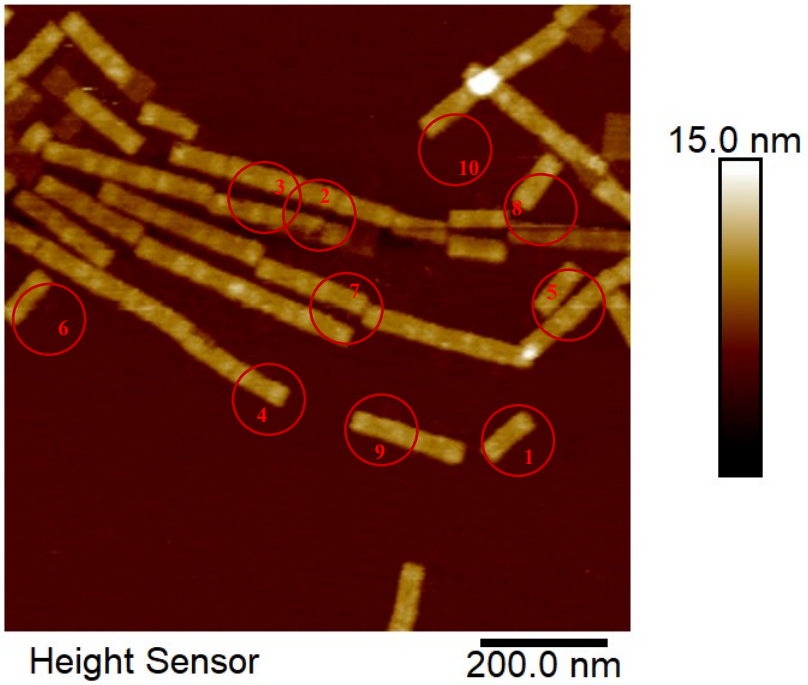

Figure S 5: AFM images of DNA origami (O_0) and marked DNA tubes which were used for height measurement. Results of height measurement are shown in Table S 4.

Table S 4: Heights of the uncoated DNA-origami (O_0)

Origami	Max height base	Max height coated area
1	4.82	5.69
2	4.88	5.54
3	4.65	5.61
4	5.11	6.00
5	4.63	5.57
6	4.45	5.68
7	4.84	5.28
8	4.95	5.82
9	5.04	6.02
10	5.09	5.90

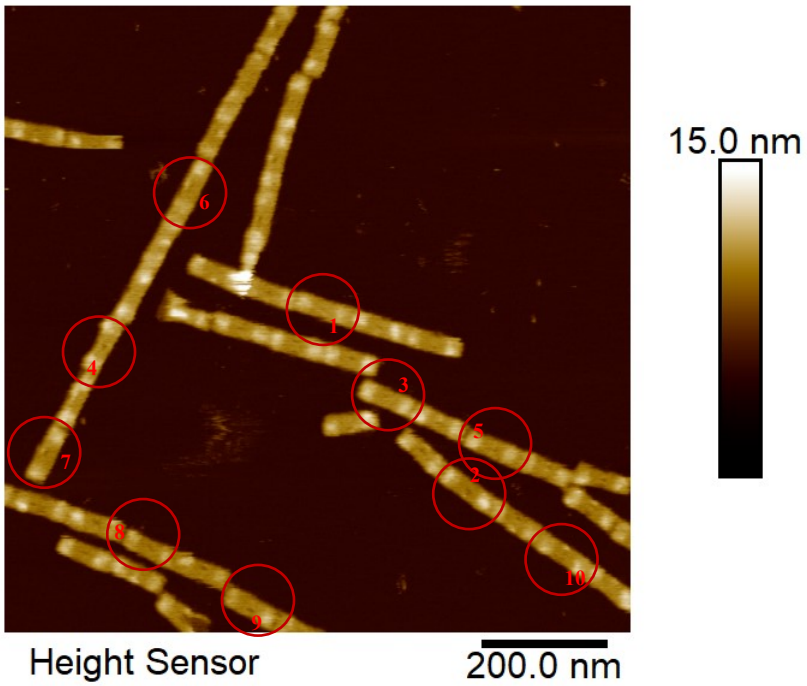

Figure S 6: AFM images of the coated DNA origami (O_1) and marked DNA tubes which were used for height measurement. Results of height measurement is shown in Table S 5.

Table S 5: Heights of the coated DNA-origami (O_1)

Origami	Max height base	Max height coated area
1	5.66	7.05
2	5.00	6.49
3	5.38	7.38
4	5.41	7.67
5	5.31	7.22
6	5.21	7.16
7	5.49	7.37
8	4.69	6.86
9	5.01	6.04
10	4.96	6.78

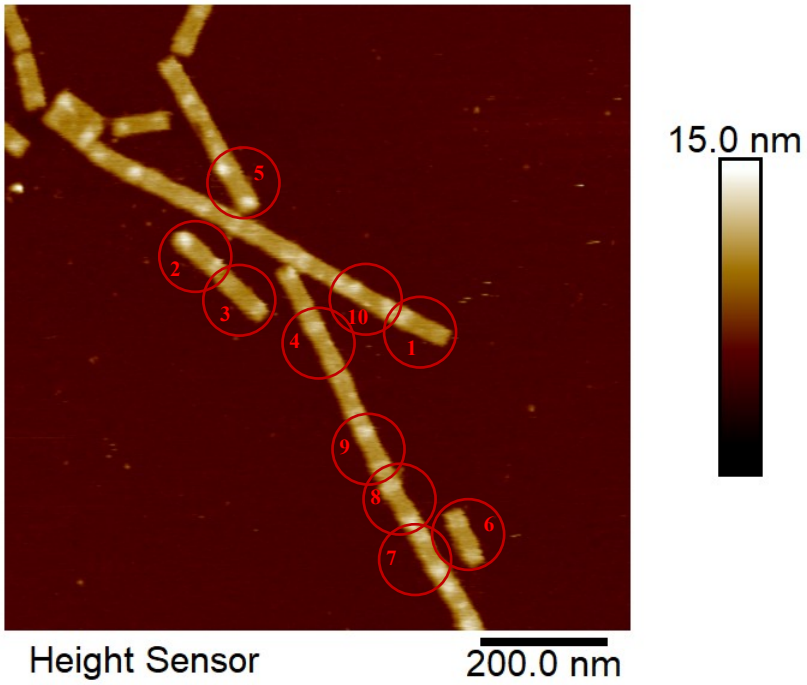

Figure S 7: AFM image of coated DNA origami (O_2) and marked DNA tubes which were used for height measurement. Results of height measurement is shown in Table S 6.

Table S 6: Heights of the coated DNA-origami (O_2)

Origami	Max height base	Max height coated area
1	4.99	6.10
2	5.76	8.40
3	5.28	6.88
4	5.47	7.43
5	5.86	8.72
6	5.65	6.96
7	5.80	7.62
8	5.80	7.87
9	5.45	7.79
10	5.74	7.84

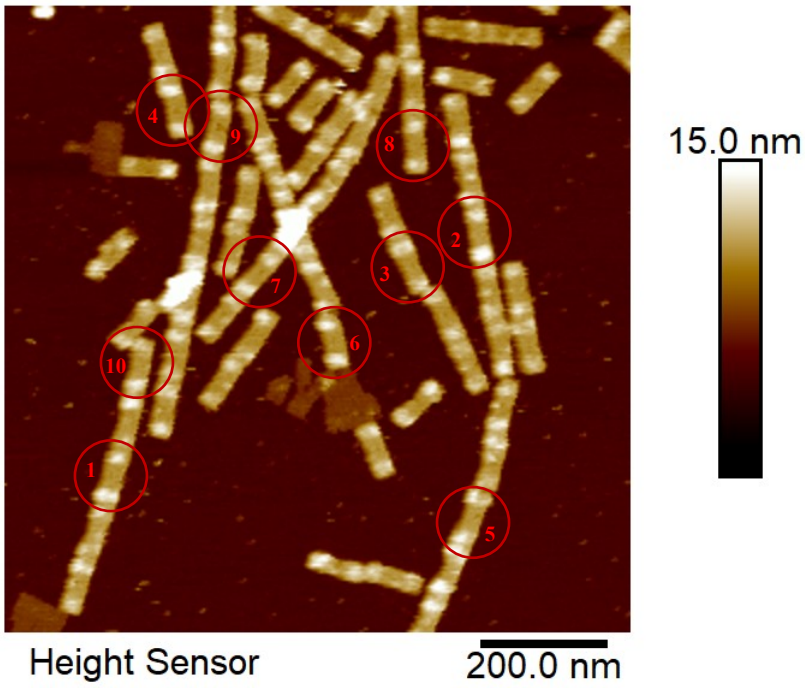

Figure S 8: AFM images of coated DNA origami (O_3) and marked DNA tubes which were used for height measurement. Results of height measurement are shown in Table S 7.

Table S 7: Heights of the coated DNA-origami (O_3)

Origami	Max height base	Max height coated area
1	5.72	8.98
2	6.22	9.34
3	5.94	8.59
4	5.88	9.41
5	5.94	9.11
6	5.72	9.01
7	5.54	8.51
8	5.47	8.37
9	5.41	8.83
10	5.92	9.03

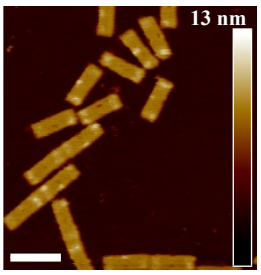

Figure S 9: AFM images of uncoated DNA origami (O_8) containing StA and StE sequences.

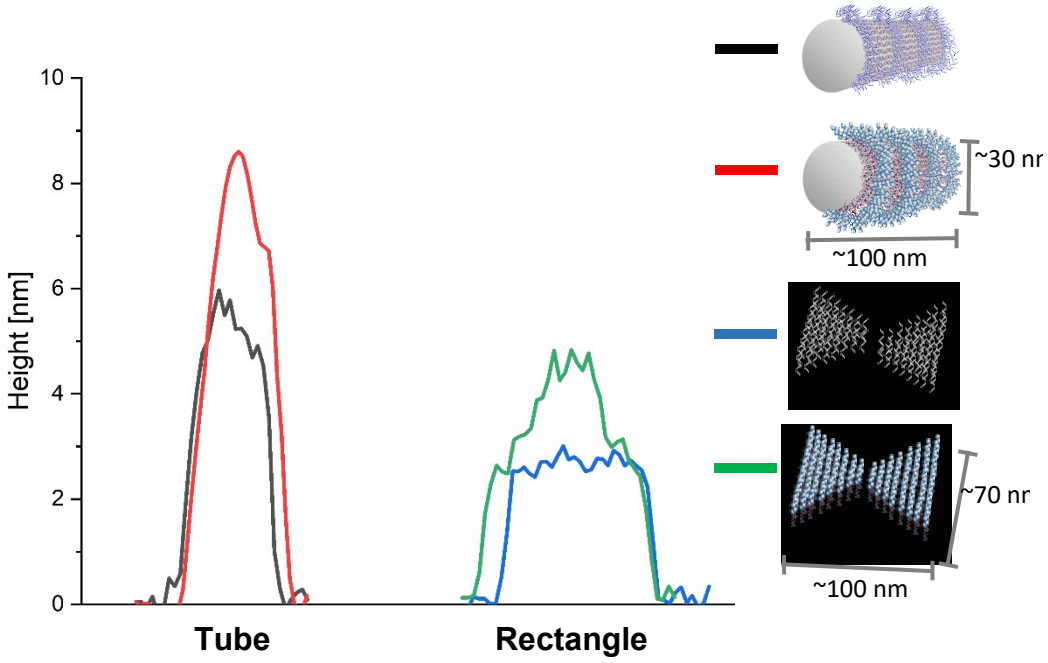

Figure S 10: AFM topographic images of O_4, O_5, O_6 and O_7 (Figure 3 f,g,h,i) reveal a significant increase in height of the respective coating area.

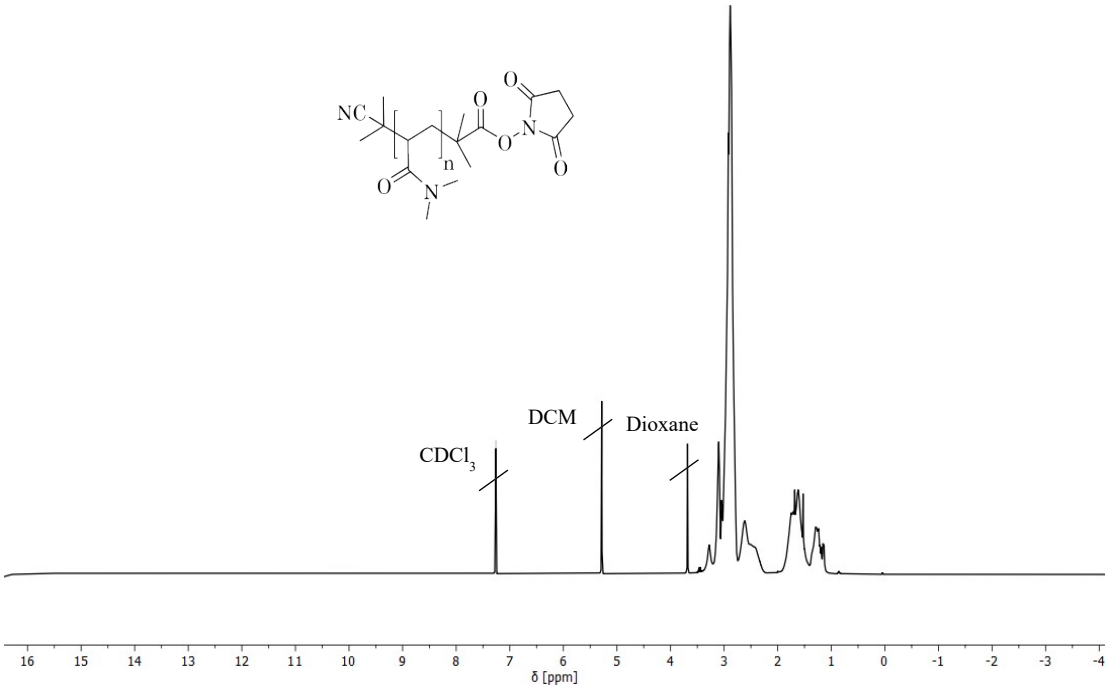

Figure S 11:^{1}H-NMR of P1 synthesized by RAFT polymerization. CTA group was removed with an excess of AIBN.

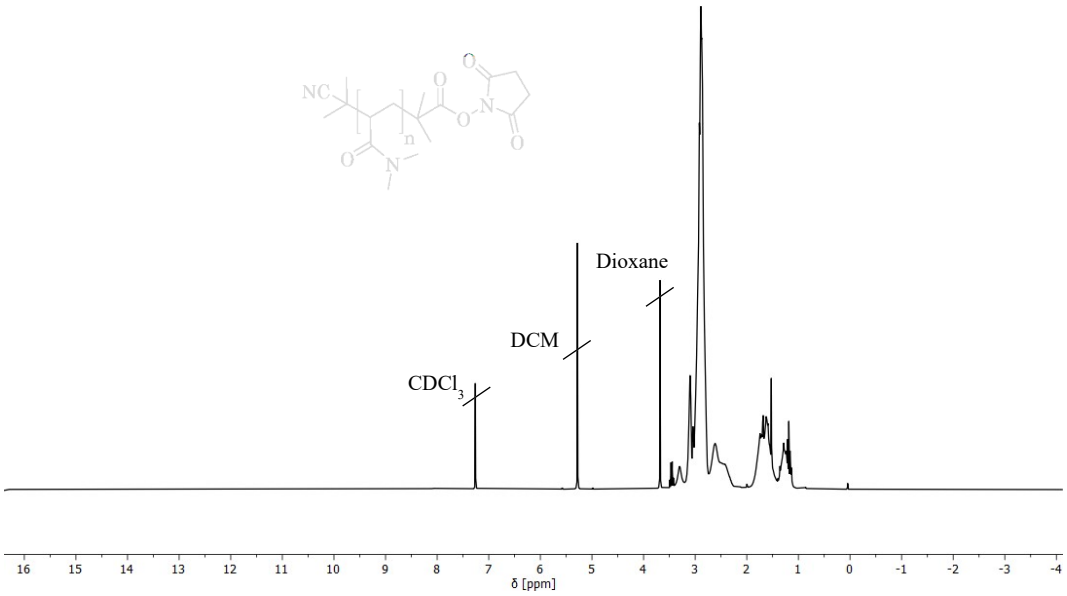

Figure S 12: ^{1}H-NMR of P2 synthesized by RAFT polymerization. CTA group was removed with an excess of AIBN.

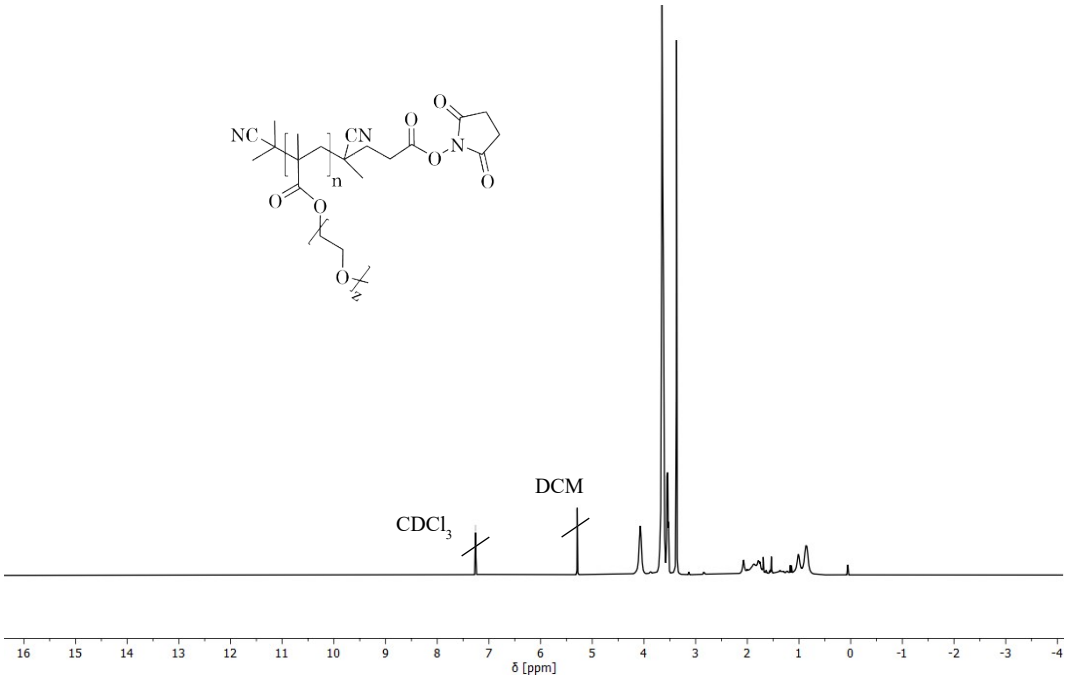

Figure S13: ^{1}H-NMR of P4 synthesized by RAFT polymerization. CTA group was removed with an excess of AIBN.

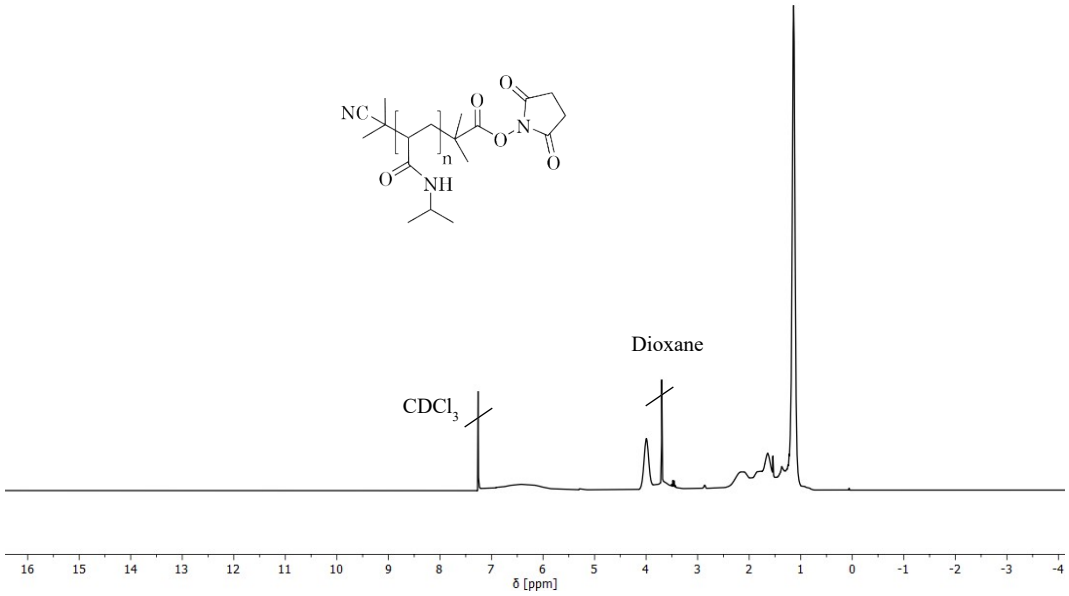

Figure S 14: ^{1}H-NMR of P5 synthesized by RAFT polymerization. CTA group was removed with an excess of AIBN.

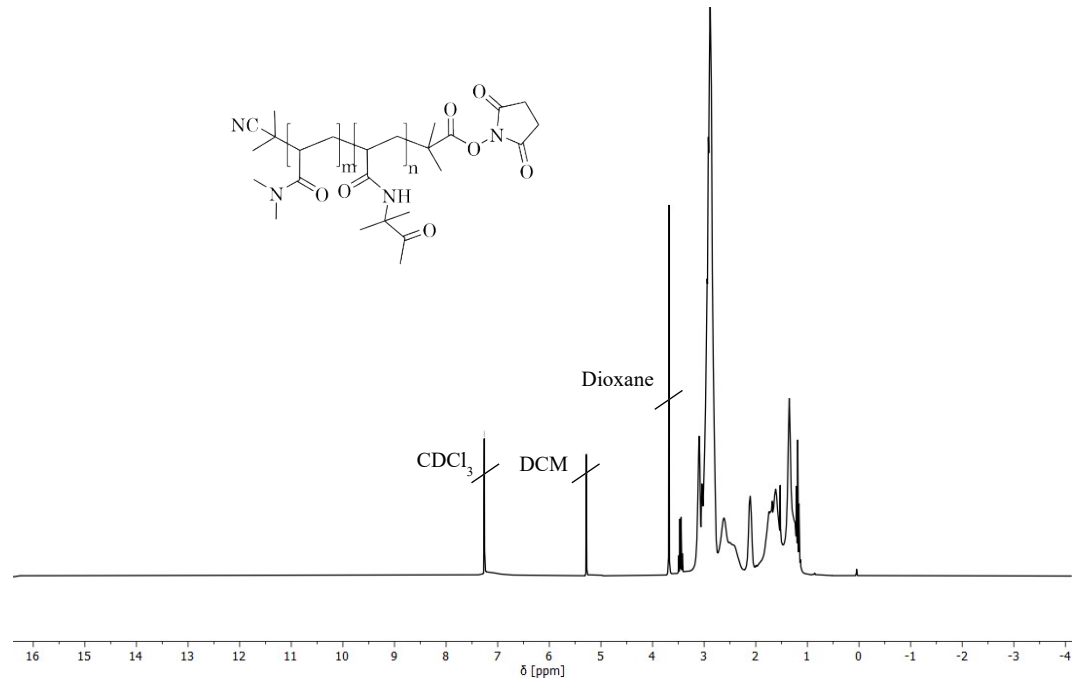

Figure S 15: ^{1}H-NMR of P8 synthesized by RAFT polymerization. CTA group was removed with an excess of AIBN.

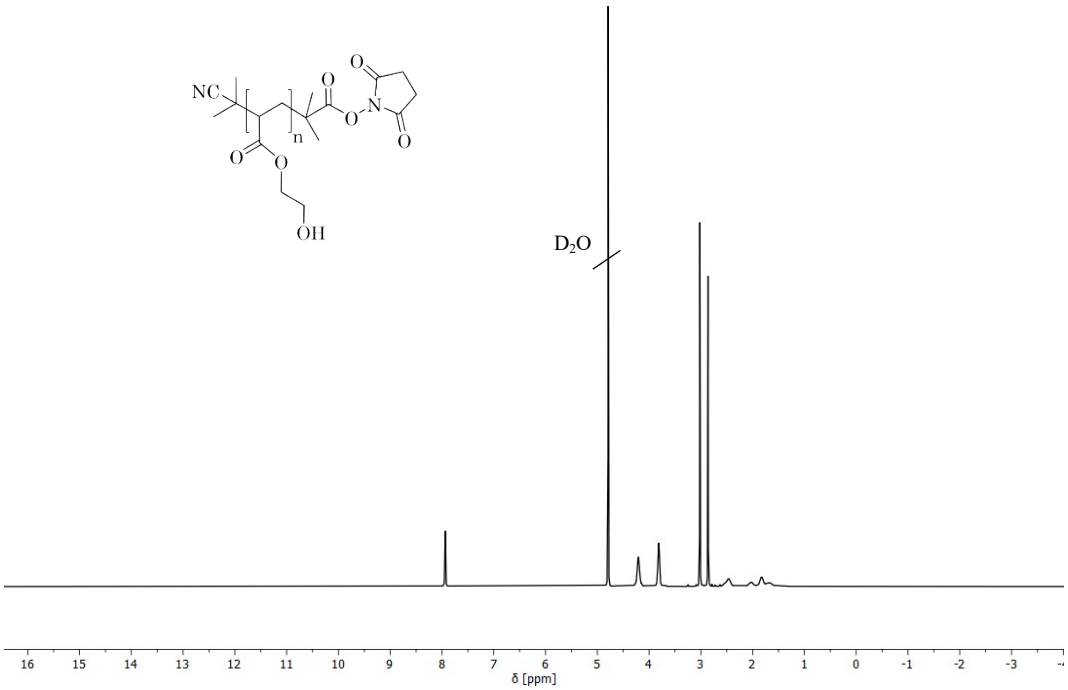

Figure S 16: ^{1}H-NMR of P7 synthesized by RAFT polymerization. CTA group was removed with an excess of AIBN.

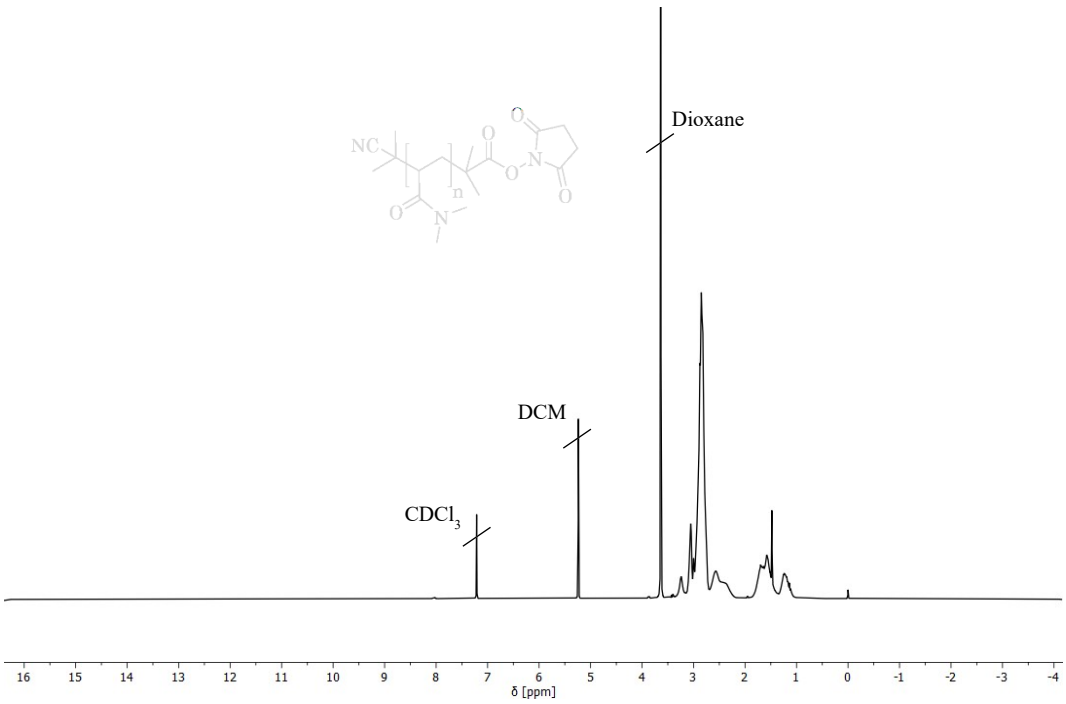

Figure S 17: ^{1}H-NMR of P3 synthesized by RAFT polymerization. CTA group was removed with an excess of AIBN.

Table S 8: Staple sequences for DNA-origami synthesis.

Name	Sequence
1	CAAGCCCAATAGGAACCCATGTACAAACAGTT
2	AATGCCCCGTAACAGTGCCCGTATCTCCCTCA
3	TGCCTTGACTGCCTATTTCGGAACAGGGATAG
4	GAGCCGCCCCACCACCGGAACCGCGACGGAAA
5	AACCAGAGACCCTCAGAACCGCCAGGGGTCAG
6	TTATTCATAGGGAAGGTAAATATTCATTCAGT
7	CATAACCCGAGGCATAGTAAGAGCTTTTTAAG
8	ATTGAGGGTAAAGGTGAATTATCAATCACCGG
9	AAAAGTAATATCTTACCGAAGCCCTTCCAGAG
10	GCAATAGCGCAGATAGCCGAACAATTCAACCG
11	CCTAATTTACGCTAACGAGCGTCTAATCAATA
12	TCTTACCAGCCAGTTACAAAATAAATGAAATA
13	ATCGGCTGCGAGCATGTAGAAACCTATCATAT
14	CTAATTTATCTTTCCTTATCATTCATCCTGAA
15	GCGTTATAGAAAAAGCCTGTTTAGAAGGCCGG
16	GCTCATTTTCGCATTAAATTTTTGAGCTTAGA
17	AATTACTACAAATTCTTACCAGTAATCCCATC
18	TTAAGACGTTGAAAACATAGCGATAACAGTAC
19	TAGAATCCCTGAGAAGAGTCAATAGGAATCAT
20	CTTTTACACAGATGAATATACAGTAAACAATT
21	TTTAACGTTCGGGAGAAACAATAATTTTCCCT
22	CGACAACTAAGTATTAGACTTTACAATACCGA
23	GGATTTAGCGTATTAAATCCTTTGTTTTCAGG
24	ACGAACCAAAACATCGCCATTAAATGGTGGTT
25	GAACGTGGCGAGAAAGGAAGGGAACAAACTAT
26	TAGCCCTACCAGCAGAAGATAAAAACATTTGA
27	CGGCCTTGCTGGTAATATCCAGAACGAACTGA
28	CTCAGAGCCACCACCCTCATTTTCCTATTATT
29	CTGAAACAGGTAATAAGTTTTAACCCCTCAGA
30	AGTGTACTTGAAAGTATTAAGAGGCCGCCACC
31	GCCACCACTCTTTTCATAATCAAACCGTCACC
32	GTTTGCCACCTCAGAGCCGCCACCGATACAGG
33	GACTTGAGAGACAAAAGGGCGACAAGTTACCA
34	AGCGCCAACCATTTGGGAATTAGATTATTAGC
35	GAAGGAAAATAAGAGCAAGAAACAACAGCCAT

36	GCCCAATACCGAGGAAACGCAATAGGTTTACC
37	ATTATTTAACCCAGCTACAATTTTCAAGAACG
38	TATTTTGCTCCCAATCCAAATAAGTGAGTTAA
39	GGTATTAAGAACAAGAAAAATAATTAAAGCCA
40	TAAGTCCTACCAAGTACCGCACTCTTAGTTGC
41	ACGCTCAAAATAAGAATAAACACCGTGAATTT
42	AGGCGTTACAGTAGGGCTTAATTGACAATAGA
43	ATCAAAATCGTCGCTATTAATTAACGGATTCG
44	CTGTAAATCATAGGTCTGAGAGACGATAAATA
45	CCTGATTGAAAGAAATTGCGTAGACCCGAACG
46	ACAGAAATCTTTGAATACCAAGTTCCTTGCTT
47	TTATTAATGCCGTCAATAGATAATCAGAGGTG
48	AGATTAGATTTAAAAGTTTGAGTACACGTAAA
49	AGGCGGTCATTAGTCTTTAATGCGCAATATTA
50	GAATGGCTAGTATTAACACCGCCTCAACTAAT
51	CCGCCAGCCATTGCAACAGGAAAAATATTTTT
52	CCCTCAGAACCGCCACCCTCAGAACTGAGACT
53	CCTCAAGAATACATGGCTTTTGATAGAACCAC
54	TAAGCGTCGAAGGATTAGGATTAGTACCGCCA
55	CACCAGAGTTCGGTCATAGCCCCCGCCAGCAA
56	TCGGCATTCCGCCGCCAGCATTGACGTTCCAG
57	AATCACCAAATAGAAAATTCATATATAACGGA
58	TCACAATCGTAGCACCATTACCATCGTTTTCA
59	ATACCCAAGATAACCCACAAGAATAAACGATT
60	ATCAGAGAAAGAACTGGCATGATTTTATTTTG
61	TTTTGTTTAAGCCTTAAATCAAGAATCGAGAA
62	AGGTTTTGAACGTCAAAAATGAAAGCGCTAAT
63	CAAGCAAGACGCGCCTGTTTATCAAGAATCGC
64	AATGCAGACCGTTTTTATTTTCATCTTGCGGG
65	CATATTTAGAAATACCGACCGTGTTACCTTTT
66	AATGGTTTACAACGCCAACATGTAGTTCAGCT
67	TAACCTCCATATGTGAGTGAATAAACAAAATC
68	AAATCAATGGCTTAGGTTGGGTTACTAAATTT
69	GCGCAGAGATATCAAAATTATTTGACATTATC
70	AACCTACCGCGAATTATTCATTTCCAGTACAT
71	ATTTTGCGTCTTTAGGAGCACTAAGCAACAGT
72	CTAAAATAGAACAAAGAAACCACCAGGGTTAG
73	GCCACGCTATACGTGGCACAGACAACGCTCAT

74	GCGTAAGAGAGAGCCAGCAGCAAAAAGGTTAT
75	GGAAATACCTACATTTTGACGCTCACCTGAAA
76	TATCACCGTACTCAGGAGGTTTAGCGGGGTTT
77	TGCTCAGTCAGTCTCTGAATTTACCAGGAGGT
78	GGAAAGCGACCAGGCGGATAAGTGAATAGGTG
79	TGAGGCAGGCGTCAGACTGTAGCGTAGCAAGG
80	TGCCTTTAGTCAGACGATTGGCCTGCCAGAAT
81	CCGGAAACACACCACGGAATAAGTAAGACTCC
82	ACGCAAAGGTCACCAATGAAACCAATCAAGTT
83	TTATTACGGTCAGAGGGTAATTGAATAGCAGC
84	TGAACAAACAGTATGTTAGCAAACTAAAAGAA
85	CTTTACAGTTAGCGAACCTCCCGACGTAGGAA
86	GAGGCGTTAGAGAATAACATAAAAGAACACCC
87	TCATTACCCGACAATAAACAACATATTTAGGC
88	CCAGACGAGCGCCCAATAGCAAGCAAGAACGC
89	AGAGGCATAATTTCATCTTCTGACTATAACTA
90	TTTTAGTTTTTCGAGCCAGTAATAAATTCTGT
91	TATGTAAACCTTTTTTAATGGAAAAATTACCT
92	TTGAATTATGCTGATGCAAATCCACAAATATA
93	GAGCAAAAACTTCTGAATAATGGAAGAAGGAG
94	TGGATTATGAAGATGATGAAACAAAATTTCAT
95	CGGAATTATTGAAAGGAATTGAGGTGAAAAAT
96	ATCAACAGTCATCATATTCCTGATTGATTGTT
97	CTAAAGCAAGATAGAACCCTTCTGAATCGTCT
98	GCCAACAGTCACCTTGCTGAACCTGTTGGCAA
99	GAAATGGATTATTTACATTGGCAGACATTCTG
100	TTTTTATAAGTATAGCCCGGCCGTCGAG
101	AGGGTTGATTTTATAAATCCTCATTAAATGATATTC
102	ACAAACAATTTTAATCAGTAGCGACAGATCGATAGC
103	AGCACCGTTTTTTAAAGGTGGCAACATAGTAGAAAA
104	TACATACATTTTGACGGGAGAATTAACTACAGGGAA
105	GCGCATTATTTTGCTTATCCGGTATTCTAAATCAGA
106	TATAGAAGTTTTCGACAAAAGGTAAAGTAGAGAATA
107	TAAAGTACTTTTCGCGAGAAAACTTTTTATCGCAAG
108	ACAAAGAATTTTATTAATTACATTTAACACATCAAG
109	AAAACAAATTTTTTCATCAATATAATCCTATCAGAT
110	GATGGCAATTTTAATCAATATCTGGTCACAAATATC
111	AAACCCTCTTTTACCAGTAATAAAAGGGATTCACCAGTCACACGTTTT

112	CCGAAATCCGAAAATCCTGTTTGAAGCCGGAA
113	CCAGCAGGGGCAAAATCCCTTATAAAGCCGGC
114	GCATAAAGTTCCACACAACATACGAAGCGCCA
115	GCTCACAATGTAAAGCCTGGGGTGGGTTTGCC
116	TTCGCCATTGCCGGAAACCAGGCATTAAATCA
117	GCTTCTGGTCAGGCTGCGCAACTGTGTTATCC
118	GTTAAAATTTTAACCAATAGGAACCCGGCACC
119	AGACAGTCATTCAAAAGGGTGAGAAGCTATAT
120	AGGTAAAGAAATCACCATCAATATAATATTTT
121	TTTCATTTGGTCAATAACCTGTTTATATCGCG
122	TCGCAAATGGGGCGCGAGCTGAAATAATGTGT
123	TTTTAATTGCCCGAAAGACTTCAAAACACTAT
124	AAGAGGAACGAGCTTCAAAGCGAAGATACATT
125	GGAATTACTCGTTTACCAGACGACAAAAGATT
126	GAATAAGGACGTAACAAAGCTGCTCTAAAACA
127	CCAAATCACTTGCCCTGACGAGAACGCCAAAA
128	CTCATCTTGAGGCAAAAGAATACAGTGAATTT
129	AAACGAAATGACCCCCAGCGATTATTCATTAC
130	CTTAAACATCAGCTTGCTTTCGAGCGTAACAC
131	TCGGTTTAGCTTGATACCGATAGTCCAACCTA
132	TGAGTTTCGTCACCAGTACAAACTTAATTGTA
133	CCCCGATTTAGAGCTTGACGGGGAAATCAAAA
134	GAATAGCCGCAAGCGGTCCACGCTCCTAATGA
135	GAGTTGCACGAGATAGGGTTGAGTAAGGGAGC
136	GTGAGCTAGTTTCCTGTGTGAAATTTGGGAAG
137	TCATAGCTACTCACATTAATTGCGCCCTGAGA
138	GGCGATCGCACTCCAGCCAGCTTTGCCATCAA
139	GAAGATCGGTGCGGGCCTCTTCGCAATCATGG
140	AAATAATTTTAAATTGTAAACGTTGATATTCA
141	GCAAATATCGCGTCTGGCCTTCCTGGCCTCAG
142	ACCGTTCTAAATGCAATGCCTGAGAGGTGGCA
143	TATATTTTAGCTGATAAATTAATGTTGTATAA
144	TCAATTCTTTTAGTTTGACCATTACCAGACCG
145	CGAGTAGAACTAATAGTAGTAGCAAACCCTCA
146	GAAGCAAAAAAGCGGATTGCATCAGATAAAAA
147	TCAGAAGCCTCCAACAGGTCAGGATCTGCGAA
148	CCAAAATATAATGCAGATACATAAACACCAGA
149	CATTCAACGCGAGAGGCTTTTGCATATTATAG

150	ACGAGTAGTGACAAGAACCGGATATACCAAGC
151	AGTAATCTTAAATTGGGCTTGAGAGAATACCA
152	GCGAAACATGCCACTACGAAGGCATGCGCCGA
153	ATACGTAAAAGTACAACGGAGATTTCATCAAG
154	CAATGACACTCCAAAAGGAGCCTTACAACGCC
155	AAAAAAGGACAACCATCGCCCACGCGGGTAAA
156	TGTAGCATTCCACAGACAGCCCTCATCTCCAA
157	GTAAAGCACTAAATCGGAACCCTAGTTGTTCC
158	AGTTTGGAGCCCTTCACCGCCTGGTTGCGCTC
159	AGCTGATTACAAGAGTCCACTATTGAGGTGCC
160	ACTGCCCGCCGAGCTCGAATTCGTTATTACGC
161	CCCGGGTACTTTCCAGTCGGGAAACGGGCAAC
162	CAGCTGGCGGACGACGACAGTATCGTAGCCAG
163	GTTTGAGGGAAAGGGGGATGTGCTAGAGGATC
164	CTTTCATCCCCAAAAACAGGAAGACCGGAGAG
165	AGAAAAGCAACATTAAATGTGAGCATCTGCCA
166	GGTAGCTAGGATAAAAATTTTTAGTTAACATC
167	CAACGCAATTTTTGAGAGATCTACTGATAATC
168	CAATAAATACAGTTGATTCCCAATTTAGAGAG
169	TCCATATACATACAGGCAAGGCAACTTTATTT
170	TACCTTTAAGGTCTTTACCCTGACAAAGAAGT
171	CAAAAATCATTGCTCCTTTTGATAAGTTTCAT
172	TTTGCCAGATCAGTTGAGATTTAGTGGTTTAA
173	AAAGATTCAGGGGGTAATAGTAAACCATAAAT
174	TTTCAACTATAGGCTGGCTGACCTTGTATCAT
175	CCAGGCGCTTAATCATTGTGAATTACAGGTAG
176	CGCCTGATGGAAGTTTCCATTAAACATAACCG
177	TTTCATGAAAATTGTGTCGAAATCTGTACAGA
178	ATATATTCTTTTTTCACGTTGAAAATAGTTAG
179	AATAATAAGGTCGCTGAGGCTTGCAAAGACTT
180	CGTAACGATCTAAAGTTTTGTCGTGAATTGCG
181	ACCCAAATCAAGTTTTTTGGGGTCAAAGAACG
182	TGGACTCCCTTTTCACCAGTGAGACCTGTCGT
183	TGGTTTTTAACGTCAAAGGGCGAAGAACCATC
184	GCCAGCTGCCTGCAGGTCGACTCTGCAAGGCG
185	CTTGCATGCATTAATGAATCGGCCCGCCAGGG
186	ATTAAGTTCGCATCGTAACCGTGCGAGTAACA
187	TAGATGGGGGGTAACGCCAGGGTTGTGCCAAG

188	ACCCGTCGTCATATGTACCCCGGTAAAGGCTA
189	CATGTCAAGATTCTCCGTGGGAACCGTTGGTG
190	TCAGGTCACTTTTGCGGGAGAAGCAGAATTAG
191	CTGTAATATTGCCTGAGAGTCTGGAAAACTAG
192	CAAAATTAAAGTACGGTGTCTGGAAGAGGTCA
193	TGCAACTAAGCAATAAAGCCTCAGTTATGACC
194	TTTTTGCGCAGAAAACGAGAATGAATGTTTAG
195	AAACAGTTGATGGCTTAGAGCTTATTTAAATA
196	ACTGGATAACGGAACAACATTATTACCTTATG
197	ACGAACTAGCGTCCAATACTGCGGAATGCTTT
198	CGATTTTAGAGGACAGATGAACGGCGCGACCT
199	CTTTGAAAAGAACTGGCTCATTATTTAATAAA
200	GCTCCATGAGAGGCTTTGAGGACTAGGGAGTT
201	ACGGCTACTTACTTAGCCGGAACGCTGACCAA
202	AAAGGCCGAAAGGAACAACTAAAGCTTTCCAG
203	GAGAATAGCTTTTGCGGGATCGTCGGGTAGCA
204	ACGTTAGTAAATGAATTTTCTGTAAGCGGAGT
205	TTTTCGATGGCCCACTACGTAAACCGTC
206	TATCAGGGTTTTCGGTTTGCGTATTGGGAACGCGCG
207	GGGAGAGGTTTTTGTAAAACGACGGCCATTCCCAGT
208	CACGACGTTTTTGTAATGGGATAGGTCAAAACGGCG
209	GATTGACCTTTTGATGAACGGTAATCGTAGCAAACA
210	AGAGAATCTTTTGGTTGTACCAAAAACAAGCATAAA
211	GCTAAATCTTTTCTGTAGCTCAACATGTATTGCTGA
212	ATATAATGTTTTCATTGAATCCCCCTCAAATCGTCA
213	TAAATATTTTTTGGAAGAAAAATCTACGACCAGTCA
214	GGACGTTGTTTTTCATAAGGGAACCGAAAGGCGCAG
215	ACGGTCAATTTTGACAGCATCGGAACGAACCCTCAG
216	CAGCGAAAATTTTACTTTCAACAGTTTCTGGGATTTTGCTAAACTTTT
Loop1	AACATCACTTGCCTGAGTAGAAGAACT
Loop2	TGTAGCAATACTTCTTTGATTAGTAAT
Loop3	AGTCTGTCCATCACGCAAATTAACCGT
Loop4	ATAATCAGTGAGGCCACCGAGTAAAAG
Loop5	ACGCCAGAATCCTGAGAAGTGTTTTT
Loop6	TTAAAGGGATTTTAGACAGGAACGGT
Loop7	AGAGCGGGAGCTAAACAGGAGGCCGA
Loop8	TATAACGTGCTTTCCTCGTTAGAATC
Loop9	GTACTATGGTTGCTTTGACGAGCACG

Loop10	GCGCTTAATGCGCCGCTACAGGGCGC

Table S 9: Folding strands (tube) for DNA-origami synthesis.

Name	Sequence
F1	CGGCCTTGATAGGAACCCATGTACAAACAGTT
F25	TGAGTTTCCGAGAAAGGAAGGGAACAAACTAT
F27	CAAGCCCACTGGTAATATCCAGAACGAACTGA
F28	CCGCCAGCCACCACCCTCATTTTCCTATTATT
F51	CTCAGAGCCATTGCAACAGGAAAAATATTTTT
F52	GGAAATACACCGCCACCCTCAGAACTGAGACT
F75	CCCTCAGACTACATTTTGACGCTCACCTGAAA
F76	GAAATGGATACTCAGGAGGTTTAGCGGGGTTT
F99	TATCACCGTTATTTACATTGGCAGACATTCTG
F132	GAACGTGGGTCACCAGTACAAACTTAATTGTA
F133	TGTAGCATTAGAGCTTGACGGGGAAATCAAAA
F156	CCCCGATTTCCACAGACAGCCCTCATCTCCAA
F157	CGTAACGACTAAATCGGAACCCTAGTTGTTCC
F180	GTAAAGCATCTAAAGTTTTGTCGTGAATTGCG
F181	ACGTTAGTCAAGTTTTTTGGGGTCAAAGAACG
F204	ACCCAAATAAATGAATTTTCTGTAAGCGGAGT
F100	GTCACACGTTTTTATAAGTATAGCCCGGCCGTCGAG
F205	TGCTAAACTTTTCGATGGCCCACTACGTAAACCGTC
N-111	AAACCCTCTTTTACCAGTAATAAAAGGGATTCACCA
N-216	CAGCGAAATTTTAACTTTCAACAGTTTCTGGGATTT

Table S 10: Used StA and StE elongations for DNA-origami synthesis and their respective complementary sequences used for the coupling reaction.

Name	Sequence
StA	TTTTTTTAGTAGGTGGTAGAG
StA^c	NH_2-TTTTCTCTACCACCTACTA
StE	TTTTTTTACTGACTGACTGACTGACTG
StE^c	NH_2-CAGTCAGTCAGTCAGTCAGT

Eco Scale Calculations (Aken et al., *Beilstein J. Org. Chem.* **2006**, *2*, 3)

Eco Scale of the current *grafting to* strategy:

Yield:		(100-70)/2 to (100-90)/2
Price:	over 50$ for 10 mmol	5
Safety:	DMF (flammable)	5
	DIPEA (toxic)	5
Setup:	common procedure (eppi tube)	0
Temperature:	Room temp (>24 h)	1
	Heating (>1 h) (37 °C)	2

Workup: simple spin filtration		0

Total: 23 - 33 (depending on yield)
Eco Scale: 100-(23 to 33) = 67 to 77 (70% to 90% yield)

Grafting from method of "Bottom-Up Fabrication of Nanopatterned Polymers on DNA Origami by In Situ Atom-Transfer Radical Polymerization" (Tokura et. al, *Angew. Chem. Int. Ed.* **2016**, *128*, 5786–5791.)

Here we calculated the Eco Scale for the introduction of the DNA-initiator to the origami surface and the *in situ* polymerization. Therefore, it was needed to make some assumptions for example the yield of this technique.

Yield:		(100-80)/2= 10
Price:	over 50$ for 10 mmol	5
Safety:	DMF (flammable)	5
	$CuBr_2$ (ecological damage)	5
Setup:	Instruments for controlled addition of chemicals	1
	Pressure equipment (freeze pump)	3
	Special glassware (Schlenk)	1
	Inert gas (Argon for polym.)	1
Temperature/time:	Heating, > 1 h	3
	Cooling, < 0 °C	5
Workup:	simple spin filtration, precipitation	0

Total: 39
Eco Scale: 100-(39) = 61