Supporting information

PLL-poly(HPMA) Bottle Brush-Based Antifouling Coatings: Three Grafting Routes

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1 Experimental Section

1.1 Materials

Milli-Q water was purified by a Barnsted water purification system, with a resistivity of <18.3 MΩ·cm. Commercially available reagents were used without purification unless mentioned otherwise. Poly-L-lysine hydrobromide (PLL MW 15,000–30,000 by viscosity, average MW of 20.9 kDa, with a degree of polymerization of 100, Sigma Aldrich[†]); 4-(2-hydroxyethyl)piperazine-1-ethanesulfonic acid (HEPES, ≥99.5% by titration, Sigma Aldrich); hydrogen peroxide solution (H_2O_2, 50 wt.% in H_2O, Honeywell); Sulfuric acid (H_2SO_4, 95.0-97.0%); 4-cyano-4-(phenylcarbonothioylthio)pentanoic acid *N*-succinimidyl ester (RAFT-NHS, Sigma Aldrich); triethanolamine (TEOA, Sigma Aldrich); eosin Y (EY, Sigma Aldrich); triethylamine (TEA, Sigma Aldrich); tetrahydrofuran (THF, dry, 99.9%, Sigma Aldrich); phosphate-buffered saline (PBS, Sigma-Aldrich); ethanol (EtOH, absolute, Merck); acetone (semiconductor grade, Honeywell); Poly(N-(2-hydroxypropyl)methacrylamide (average M_n 30,000–50,000, PDI ≤1.3, Sigma Aldrich); *N*-(2-hydroxypropyl) methacrylamide (HPMA, Polysciences, Inc); (3-acryloylamino-propyl)-(2-carboxy-ethyl)-dimethyl-ammonium (CBMA, was synthetized according to a previously described procedure[S1]); Bovine serum albumin-Alexa488 (BSA-Alexa488, Fisher Thermo Scientific); Lysozyme, FITC labeled (LYS-FITC, NANOCS); Silicon single side polished (Si(111), N-type, phosphorus-doped, Siltronix); deuterium oxide (D_2O, 99.9 atom% D, Sigma Aldrich).

The surfaces were plasma-cleaned by a Diener Femto plasma system. Sonication steps were performed in an Elmasonic P 30 H ultrasonic unit. Cellulose membrane dialysis tubing (25 mm flat width, Digma-Aldrich) was used for ≥8 mL volume dialyses. Float-a-lyzer G2 8 mL dialysis

[†] P7890 Certificate of Analysis (accessed 2020/05/19) https://www.sigmaaldrich.com/Graphics/COfAInfo/SigmaSAPQM/COFA/P7/P7890/P7890-BULK_____SLCF0233__.pdf

membranes with a 3.5–5 kD MWCO (VWR) were used for the final purification step of the bottlebrush polymers. LEDs with a maximum intensity at 410 nm (Intelligent LED Solutions product number: ILH-XO01-S410-SC211-WIR200) were used for polymerization. The current was set at 700 mA, corresponding to a total radiometric power of 2.9 W, according to manufacturer specifications.

1.2 Methods

Nuclear magnetic resonance (NMR). ^{1}H NMR measurements were recorded on a Bruker Avance III NMR at 400 MHz. Chemical shifts are reported in parts per million (ppm).

Ellipsometry. The dry thickness of the brushes was measured using an Accurion Nanofilm_ep4 Imaging Ellipsometer. The ellipsometric data were acquired in air at room temperature using light in the wavelength range of λ = 400.6 – 761.3 nm at an angle of incidence of 50°. The data were fitted with EP4 software using a multilayer model.

X-ray Photoelectron Spectroscopy (XPS). XPS spectra were obtained using a JPS-9200 photoelectron spectrometer (JEOL, Japan) with monochromatic Al-Kα X-Ray radiation at 12 kV and 20 mA. The obtained spectra were analyzed using CASA XPS software (version 2.3.16 PR 1.6). In C1s and N1s narrow-range spectra, the positions are set to 285 eV and 400 eV for the C–C and N–C signals, respectively. For layers <15 nm, the thickness was calculated based on the attenuation of the Si signal in XPS, according to a published procedure.[S2]

Infrared (IR). IR analyses were performed on a Bruker Tensor 27 spectrometer with platinum ATR accessory.

Gel Permeation Chromatography (GPC). The polymer molecular weight and polydispersity index (PDI) were determined using gel permeation chromatography (Agilent G5654A quaternary pump, G7162A refractive index detector), where a PSS SUPREMA Combination medium (P/N 206-0002) 1000 Å single porosity column was employed (0.05% NaN_3 in Milli-Q water as eluent, 0.4 mL/min). Polymer solutions were freshly prepared in Milli-Q and sonicated for 5 min to assure full dissolution. An injection volume of 20 μL was used for each analysis. An Agilent PL2080-0101 PEG calibration kit was used for calibration purposes. Commercially available poly(HPMA) with a molecular weight of 30–50 kDa was used as a secondary calibration standard to account for the stronger hydrophilic character of the poly(HPMA) polymer. The PEG calibration curve was adjusted to account for those interactions on a factor equal to 3.3.

Dynamic Light Scattering (DLS). Analyses were performed with an ALV/CGS-3 Compact Goniometer analyzer at room temperature in Milli-Q water. Solutions of modified PLL in fresh Milli-Q water were freshly prepared and sonicated to assure full solvation.

Fluorescence microscopy. A Leica TCS SP8 confocal laser scanning microscope (CLMS) (Leica Microsystems, Mannheim, Germany) was used to measure protein fouling and specific interactions of the coated surfaces. A Leica HyDTM hybrid detector was used in photon counting mode to measure the intensity of the fluorescence signal. The fluorescence was measured in the wavelength range from 500 to 535 nm with laser excitation wavelength at 488 nm. A 10× objective was used, and the samples were set in focus by maximizing the reflected light intensity from the laser. Fluorescence images were obtained by accumulating ten consecutive images. Images were analyzed with the Leica LAS X Life Science software.

Atomic force microscopy. AFM surface topography images were acquired by an Asylum Research MFP-3D SA AFM (Oxford Instruments, United Kingdom). Gwyddion software was used to process and analyze the AFM topography images.[S3] The average root mean square roughness (*Rq)* was calculated from topography images from 2 independent surfaces.

Preparation of PLL coatings. Silicon wafers were cut into 1×1 cm pieces and cleaned by sonication in acetone for 5 min and drying in a argon stream, subsequently oxidized by air plasma for 5 min and cleaned in a piranha solution (3 : 1 mixture by volume of H_2SO_4 : H_2O_2) for 15 minutes, after which they were soaked in and extensively rinsed with Milli-Q water and finally dried by a stream of nitrogen. The freshly cleaned surfaces were immediately used for modification by being covered with a 0.1 mg/mL (sonicated, 5 min) solution of PLL or modified PLL in HEPES buffer (10 mM, pH 7.4) and stored in a humidity chamber overnight at room temperature. Afterwards, they were again extensively rinsed with Milli-Q water and dried by a stream of nitrogen.

Immobilization of NHS-RAFT on PLL-modified surfaces. The synthesis was performed according to a previously published procedure.[S4] Freshly prepared PLL-modified surfaces were submerged in a solution of RAFT-NHS (20 mg, 53 μmol) and TEA (7.3 mg, 10 μL, 72 μmol) in 1 mL of dry THF at RT for 16 h. The substrates were subsequently rinsed with THF, acetone, EtOH, and Milli-Q water and dried by a stream of nitrogen. The substrates were immediately used for polymerization or stored under argon protection before use.

SI-PET-RAFT. The synthesis was performed according to a previously published procedure.[S4] A stock solution with photocatalyst was prepared to contain: EY (25 mg, 39 μmol), TEOA (160 mg, 1.6 mmol) in 10 mL of Milli-Q water. The monomer HPMA (200 mg, 1.4 mmol) was

dissolved in Milli-Q water (1 mL), and subsequently, 10 μL of the stock solution was added. The mixture was vortexed and added to the vials containing surfaces with an immobilized RAFT agent. Immediately after this, the polymerization was conducted by irradiating the vials with visible light from an LED light source for different periods of time. The thickness of the polymerization solution on top of the surfaces was 2 mm. In these experiments, the light source was placed 3–4 cm from the substrates. The polymerization was stopped by turning off the light. The samples were removed from the solution and subsequently rinsed with Milli-Q water and ethanol and blown dry under a stream of argon.

Protein fouling studies. Fouling of the coated surfaces was investigated by incubating surfaces in the single-protein solution of fluorescein isothiocyanate labeled lysozyme (LYS-FITC) (0.1 $mg \cdot mL^{-1}$) or Alexa488 labeled bovine serum albumin (BSA-Alexa488) (0.1 $mg \cdot mL^{-1}$) 15 min at room temperature according to a published procedure.[S4] The surfaces were then washed with PBS buffer (pH 7.4) and Milli-Q water and subsequently dried in a stream of argon. Subsequently, the samples were mounted on microscope slides using double sides tape and fluorescence intensity of the adsorbed proteins was measured. Each sample was produced and measured *in duplo*.

1.3 Synthesis

Synthesis of macroinitiator PLL-RAFT agent. The PLL-RA macroinitiator synthesis was based on a previously published method.[S5] Firstly, the following solutions were prepared in falcon tubes:

1) 50 mg PLL (2.4 μmol polymer, 0.24 mmol lysine monomer) in 2.0 mL HEPES buffer (10 mM) and sonicate for 5 min to assure full solvation;
2) 17.4 mg sulfo-NHS (80 μmol) in 2.0 mL HEPES buffer (10 mM);
3) 153.5 mg EDC·HCl (0.80 mmol) in 2.0 mL HEPES buffer (10 mM);
4) 30.1 mg NHS-RAFT (80 μmol) in 2.0 mL dry DMSO.

Solutions 1–3 were combined and mixed in a falcon tube. Solution 4 was freshly prepared and added, after which the mixture was shaken in an end-over-end shaker overnight (16 hour) at room temperature. The day after, the pink, slightly opaque solution was dialyzed against Milli-Q water for 3 days with 3 medium exchanges using dialysis membranes with a 14 kDa MWCO. After evaporation of the solvent and lyophilization, 52.4 mg (MW 24 kDa, 2.2 μmol) of a fluffy slightly ping/orange powder was obtained with a yield of 92%.

PLL-RA characterization:

^{1}H-NMR (400 MHz, D_2O, 298K) δ 1.35 (H_f; t, 2H), δ 1.6 (H_e; t, 2H), δ 1.81 (H_d; s, 3H), δ 2.90 (H_c; t, 2H), δ 4.20 (H_b; t, 1H), δ 7.41-7.58 (H_a; m, 5H) see Figure S1.

^{13}C-NMR (100 MHz, D_2O, 298K) due to poor solubility it was not possible to get a ^{13}C NMR spectrum, instead a **^{1}H-^{13}C-HSQC** (100 MHz, D_2O, 298K) was recorded (see Figure S2).

IR 3279 cm^{-1} (N–H stretch); 3064 cm^{-1} (aromatic C–H stretch); 2933 cm^{-1} (C–H stretch); 1641 cm^{-1} (carbonyl C=O stretching; Amide I band); 1533 cm^{-1} (carbonyl N–H bending; Amide II band); 1038 cm^{-1} (thiocarbonyl C=S stretch). See also Figure S7.

XPS Surface immobilized PLL-RA (Figure 6 and Figure S9).

PET-RAFT synthesis of polymer brushes. A stock solution with photocatalyst was prepared to contain: EY (25 mg, 39 μmol), TEOA (160 mg, 1.6 mmol) in 10 mL of Milli-Q water. The PLL-RAFT macroinitiator (2 mg, $8.4 \cdot 10^{-2}$ μmol) was dissolved in Milli-Q water 1.0 mL and sonicated for 1 hour. The monomer HPMA (61 mg, 0.4 mmol) or mixture of monomers HPMA (58 mg, 0.38 mmol) and CBMA (5 mg, 20 μmol) and subsequently 10 μL of the stock solution were added. Then the polymerization was conducted by irradiating the vials with visible light from an LED light source for 80 minutes. In these experiments, the light source was placed 3–4 cm from the substrates. The polymerization was stopped by turning off the light. The samples were dialyzed against demi water for 3 days with 3 medium exchanges using dialysis membranes with a 3.5–5 kDa MWCO. After evaporation of the solvent and lyophilization, fluffy, slightly orange powders were obtained.

PLL-HPMA characterization:

^{1}H-NMR (400 MHz, D_2O, 298K) δ 0.90 (H_j; d-overlapping, 3H), δ 1.10 (H_i; s,), δ 1.35 (H_f; t, 2H), δ 1.62 (H_k and H_e overlapping; m), δ 2.92 (H_c; t, 2H), δ 3.08 (H_h; d-overlapping, 2H), δ 3.84 (H_g; t, 1H), δ 4.15 (H_b; t, 1H), δ 7.44 (H_a; m, 5H) see Figure S3 and Figure S1.

^{13}C-NMR (100 MHz, D_2O, 298K) due to poor solubility it was not possible to get a ^{13}C NMR spectrum, instead a **^{1}H-^{13}C-HSQC** (100 MHz, D_2O, 298K) was recorded (see Figure S4).

IR 3288 cm^{-1} (N–H stretch); 3064 cm^{-1} (aromatic C–H stretch); 2933 cm^{-1} (C–H stretch); 1641 cm^{-1} (carbonyl C=O stretching; Amide I band); 1533 cm^{-1} (carbonyl N–H bending; Amide II band); 1038 cm^{-1} (thiocarbonyl C=S stretch). See also Figure S7.

GPC M_n = 32000, M_w = 43000, PDI = 1.4 (see Figure S10).

DLS R_{avg} = 111 nm (see Figure S11).

XPS Surface immobilized PLL-HPMA (Figure S9).

PLL-HPMA/CBMA characterization:

^{1}H-NMR (400 MHz, D_2O, 298K) δ 0.98 (H_j; d-overlapping, 3H), δ 1.11 (H_i; s,), δ 1.35 (H_f; t, 2H), δ 1.68 (H_k and H_e overlapping; m), δ 1.85 (H_d; m, 3H), δ 1.89 (H_p; t, 2H), δ 2.62 (H_o; t, 2H), δ 3.01 (H_c; t, 2H), δ 3.09 (H_l and H_h overlapping, m), δ 3.24 (H_n; d, 2H), δ 3.39 (H_m; d, 2H), δ 3.88 (H_g; t, 1H), δ 4.17 (H_b; t, 1H), δ 7.43 (H_a; m, 5H) see Figure S5.

^{13}C-NMR (100 MHz, D_2O, 298K) due to poor solubility it was not possible to get a ^{13}C NMR spectrum, instead a **^{1}H-^{13}C-HSQC** (100 MHz, D_2O, 298K) was recorded (see Figure S6).

IR 3327 cm^{-1} (N–H stretching); 3060 cm^{-1} (aromatic C–H stretch); 2929 cm^{-1} (C–H stretching); 1631 cm^{-1} (carbonyl C=O stretching; Amide I band); 1527 cm^{-1} (carbonyl N–H bending; Amide II band); 1037 cm^{-1} (thiocarbonyl C=S stretching). See also Figure S7.

GPC M_n = 34000, M_w = 53000, PDI = 1.6 (see Figure S10).

DLS R_{avg} = 111 nm (see Figure S11).

XPS Surface immobilized PLL-HPMA/CBMA (Figure S9).

2 Supporting data

2.1 ^{1}H-NMR spectra

2.1.1 PLL-RA macroinitiator

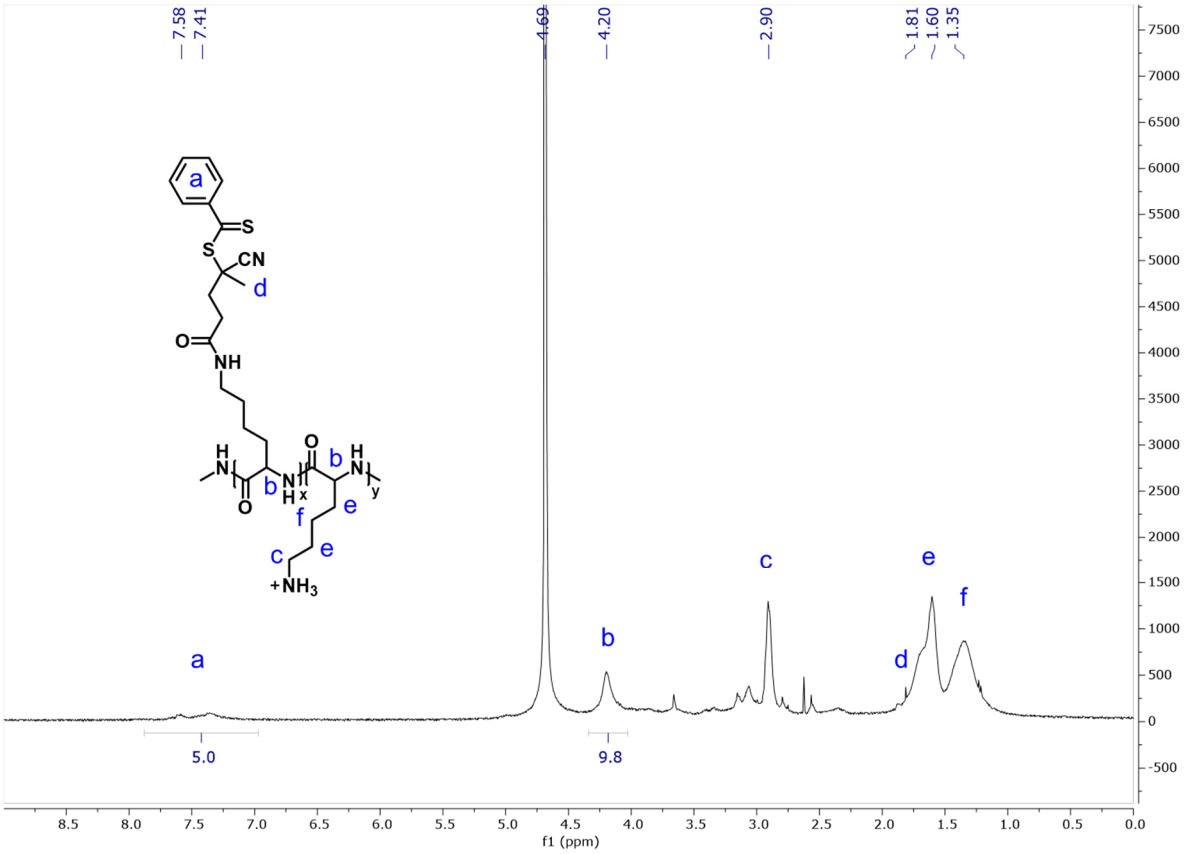


Figure S1. ^{1}H-NMR spectrum of PLL-RA measured in D_2O (400 MHz, 298 K).

The protons on the benzyl group of the RA gave a broad signal around at δ = 7.5 ppm. When comparing the integral to the ^{1}H PLL backbone signal at δ = 4.2 ppm (which is not changed upon binding to the RA), we could determine the ratio of RAFT agent : lysine to be 1 : 9.8. That is, approximately 10% of pendant amine groups of PLL were functionalized with the RAFT agent. Based on the NMR-based conversion and the average MW of the PLL starting material is 20.9 kDa with a degree of polymerization of 100, the MW of the synthesized PLL-RA was estimated to be 24 kDa.

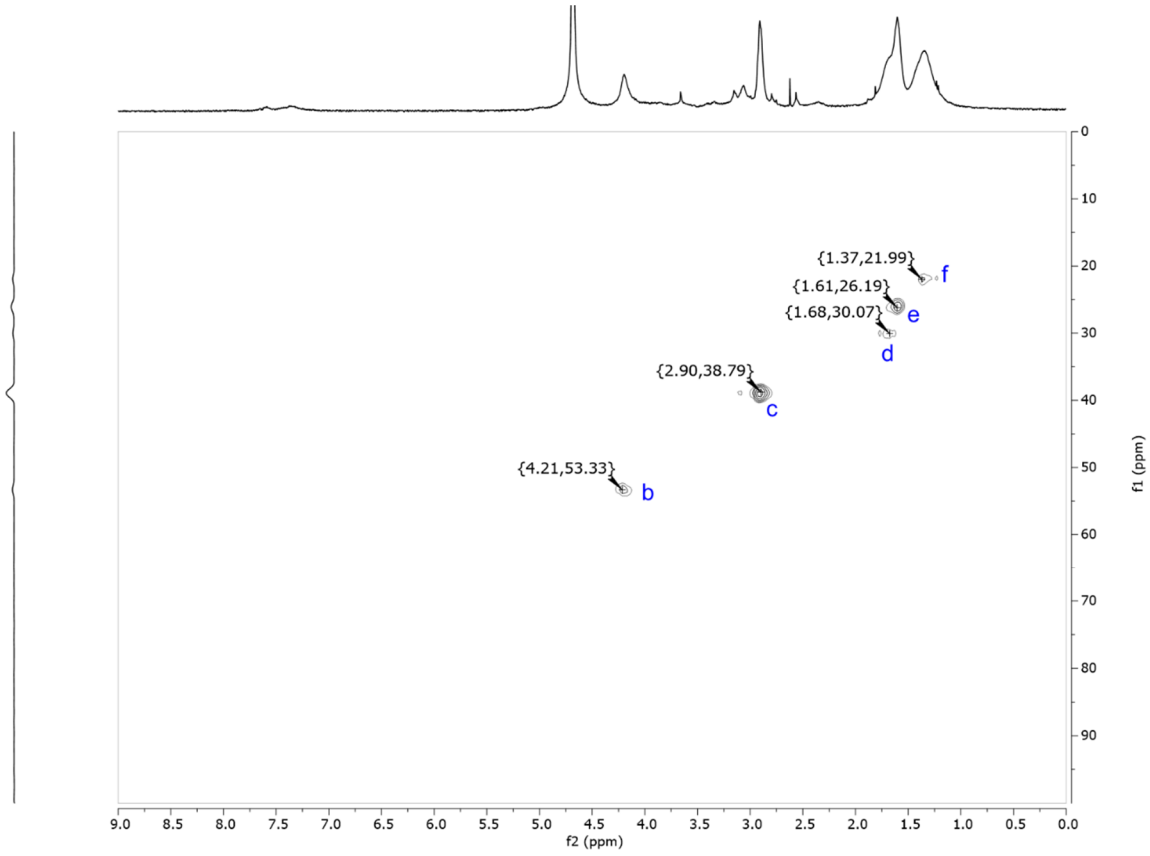


Figure S2. ^{1}H-^{13}C-HSQC NMR spectrum of macroinitiator PLL-RA measured in D_2O (100 MHz, 298 K). Peaks were assigned following the labelling of the structure as shown in Figure S1.

2.1.2 PLL-HPMA bottlebrush

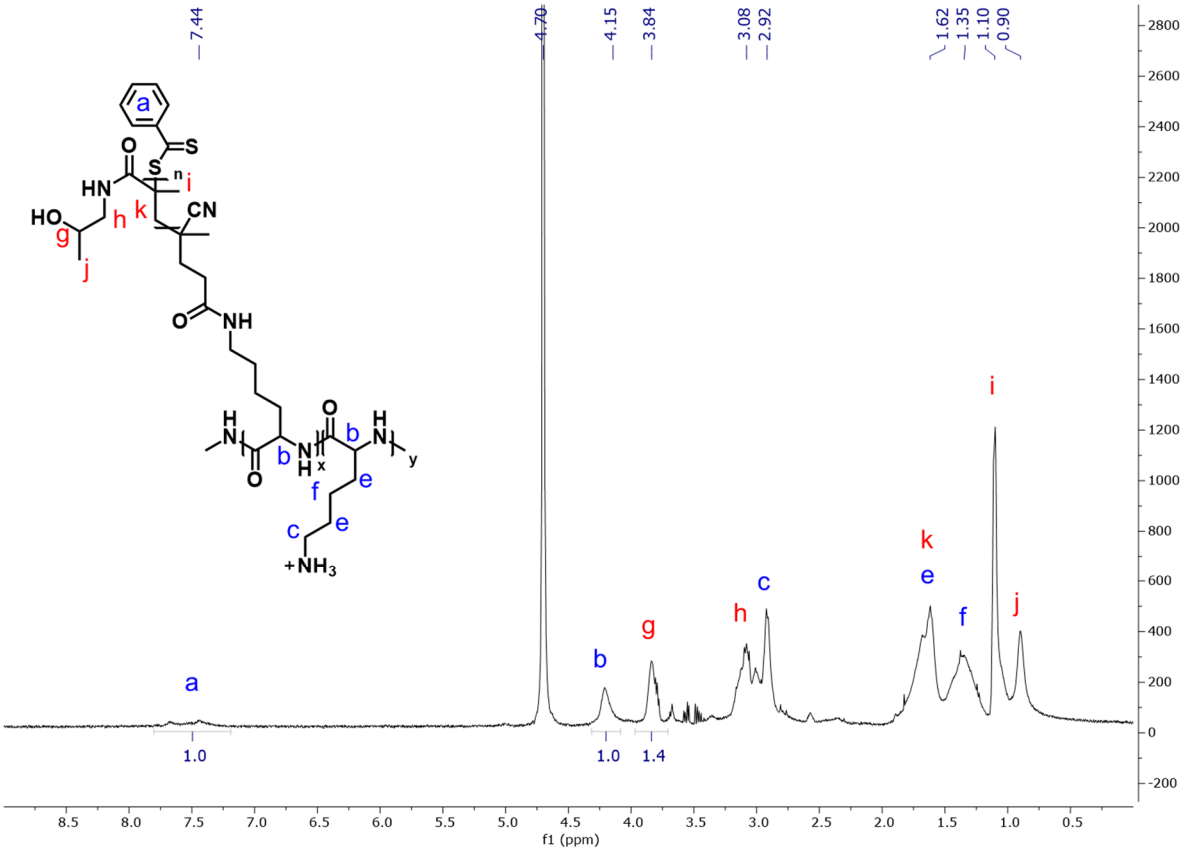


Figure S3. ^{1}H-NMR spectrum of PLL-HPMA (D_2O, 400 MHz, 298 K). (Figure is also placed in main text and copied here to support calculations).

From the integrals of the ^{1}H signal at δ = 4.15 ppm and δ = 3.84 ppm, the ratio between HPMA monomer : lysine monomer was determined to be 1.4 : 1. Combined with the 1 : 9.8. ratio of RA : lysine of the PLL-RA macroinitiator, the average chain length of the each HPMA side chain was calculated to be 14 repeating monomers (MW 143 g·mol^{-1}), corresponding to approximately 2 kDa. Based on this information and assuming the average MW of the PLL starting material is 20.9 kDa with a degree of polymerization of 100, the molar ratio between lysine : HPMA in the bottlebrush was calculated to be approximately 1 : 1 with a total weight of 40.9 kDa.

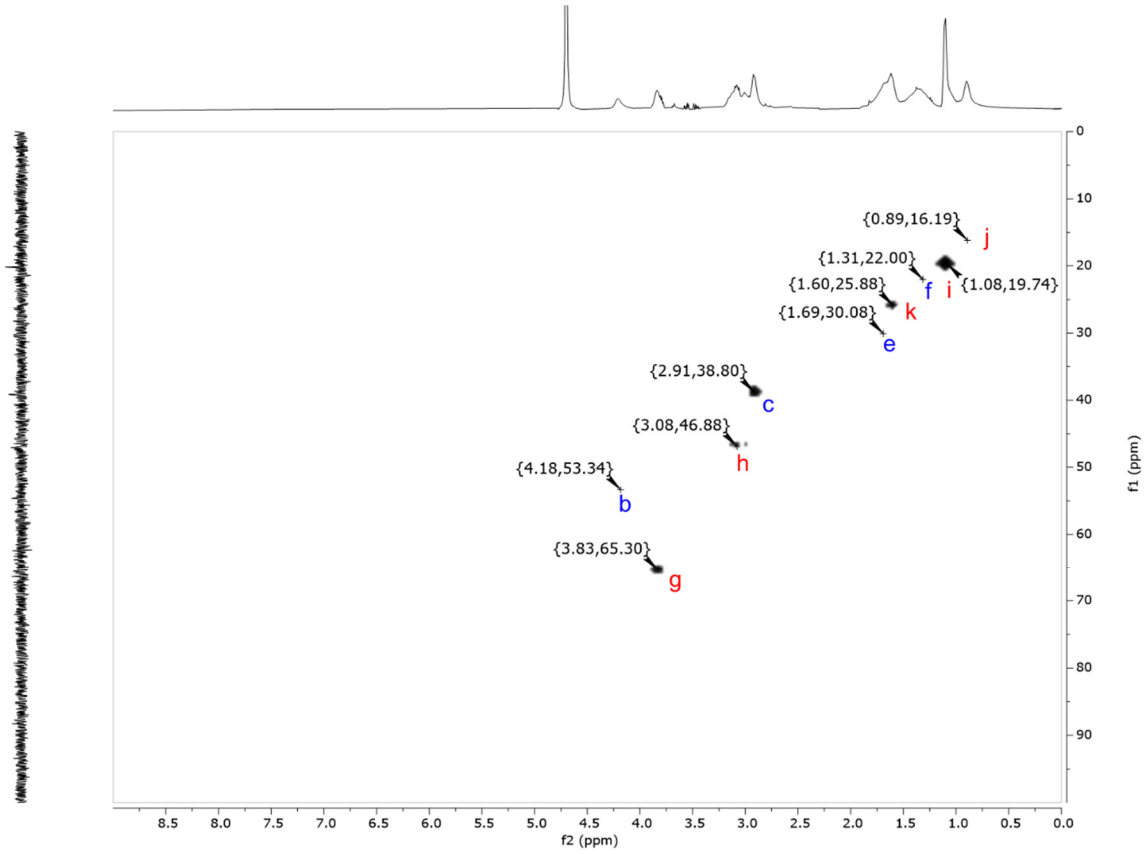


Figure S4. ^{1}H-^{13}C-HSQC NMR spectrum of PLL-HPMA measured in D_2O (100 MHz, 298 K). Peaks were assigned following the labelling of the structure as shown in Figure S3.

2.1.3 PLL-HPMA/CBMA bottlebrush

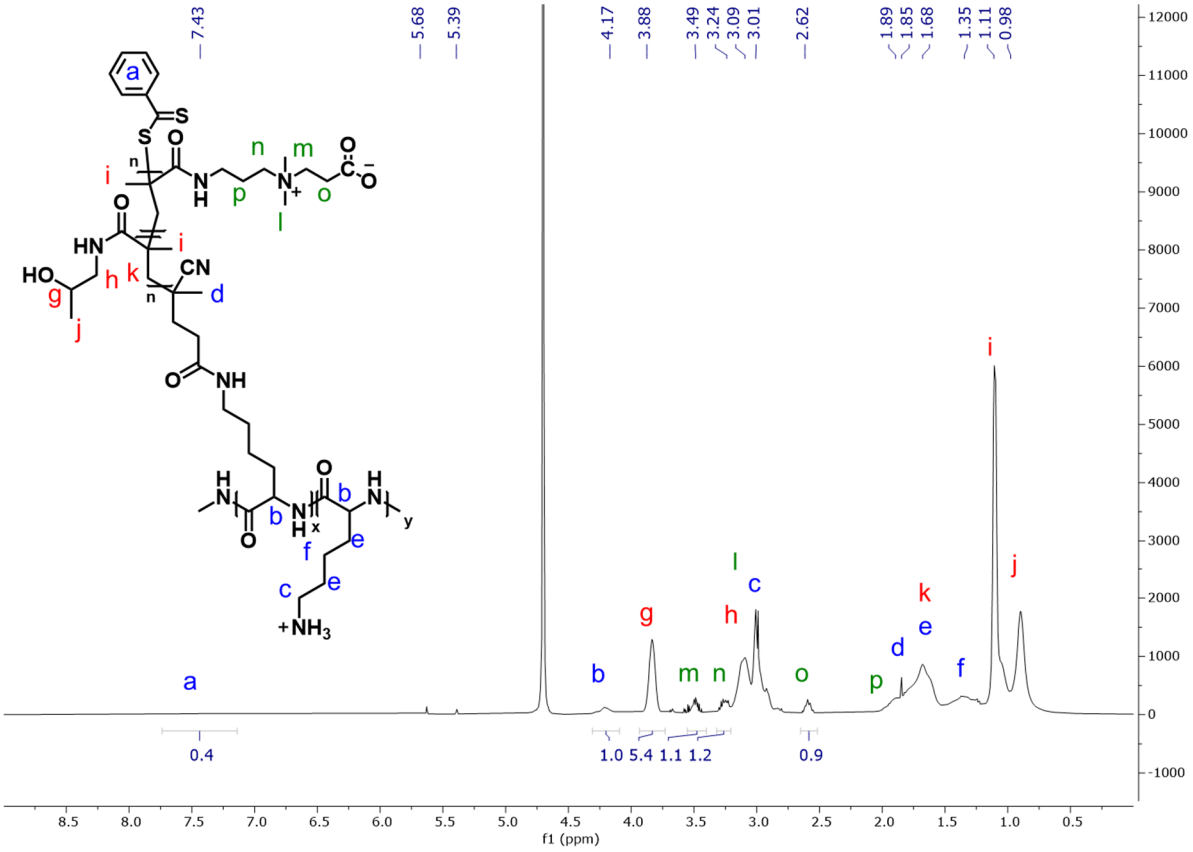


Figure S5. ^{1}H-NMR spectrum of PLL-HPMA/CBMA (D_2O, 400 MHz, 298 K).

The isolated ^{1}H peak at δ = 2.62 ppm originates from the CBMA monomers and was used to determine the ratio between HPMA and CBMA in the bottlebrush side chains. By comparing this CBMA signal at δ = 2.62 ppm to the HPMA signal at δ = 3.88 ppm, we found the content of CBMA to be 7.7%. The ^{1}H-NMR spectrum was used to estimate the average HPMA/CBMA side chain length by comparing their combined integrals to the isolated the PLL backbone signal at δ = 4.17 ppm. Since it is known from the NMR data of PLL-RA macroinitiator, that the ratio between RAFT : lysine is roughly 1 : 9.8, the average chain length of the HPMA/CBAA side chains could be calculated to be roughly 100 repeating monomers. The average weight of the HPMA/CBMA monomer was estimated 150 g·mol^{-1} (7.7% CBMA 242 g·mol^{-1} and 92.3% HPMA 143 g·mol^{-1}) which brings the total MW of the side chains to approximately 15 kDa.

The difference in side chain weight can partly be explained due to the higher molecular weight of the CBMA monomer (242 g·mol^{-1}) compared to HPMA (143 g·mol^{-1}) and the difference in reactivity as described in the main text.

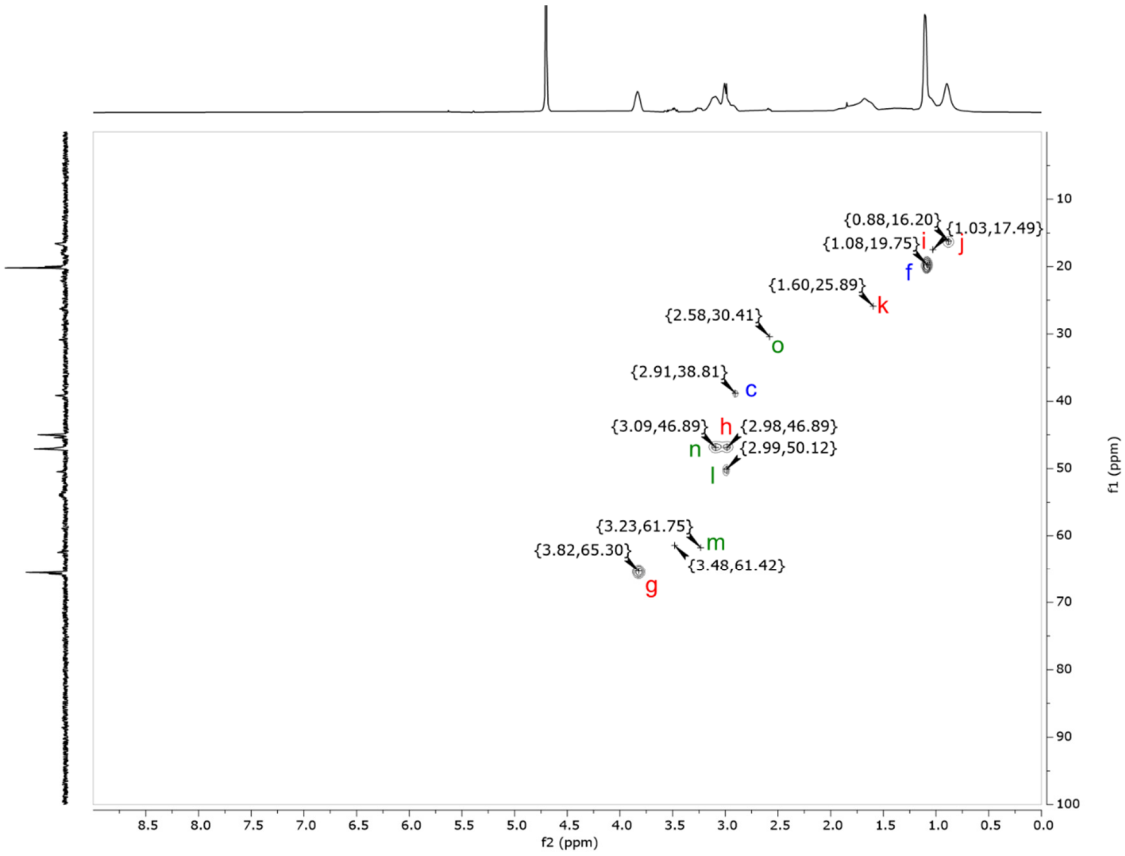


Figure S6. ^{1}H-^{13}C-HSQC NMR spectrum of PLL-HPMA/CBMA measured in D_2O (100 MHz, 298 K). Peaks were assigned following the labelling of the structure as shown in Figure S5.

2.2 Infrared spectra

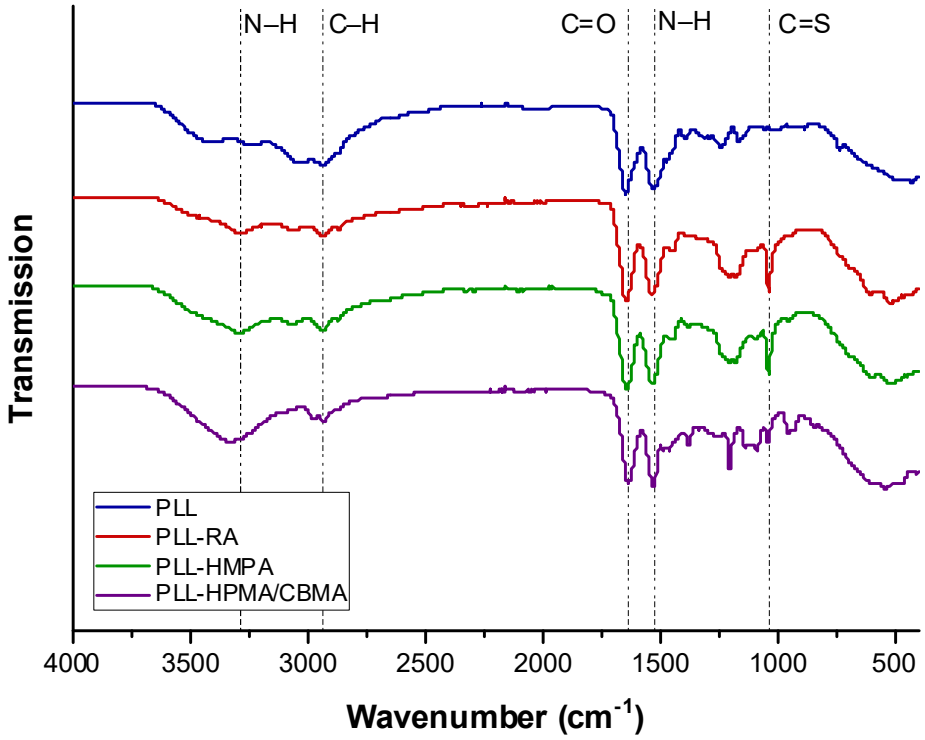


Figure S7. IR spectra of PLL, PLL-RA, PLL-HPMA and PLL-HPMA/CBMA. On the *y*-axis the spectra are shown with an offset with respect to each other.

2.3 XPS data and calculations PLL-RA conversion (route A, step A2)

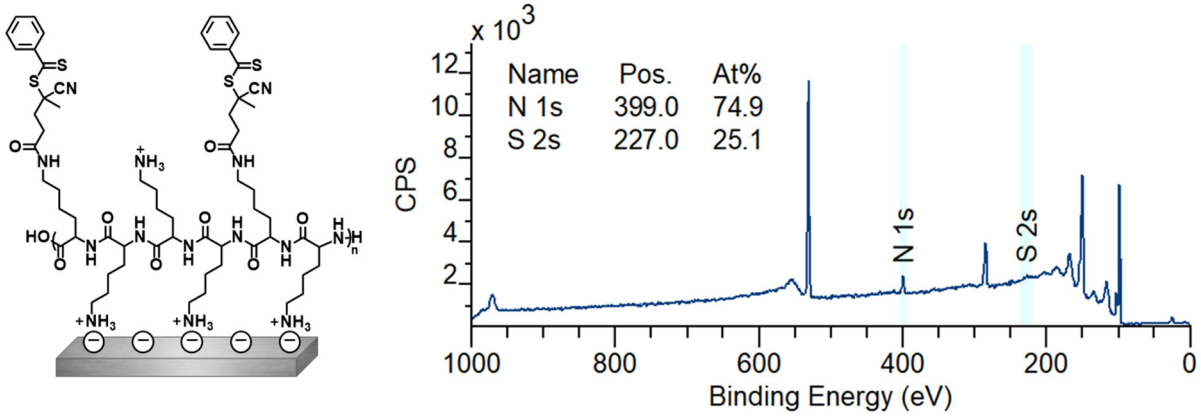
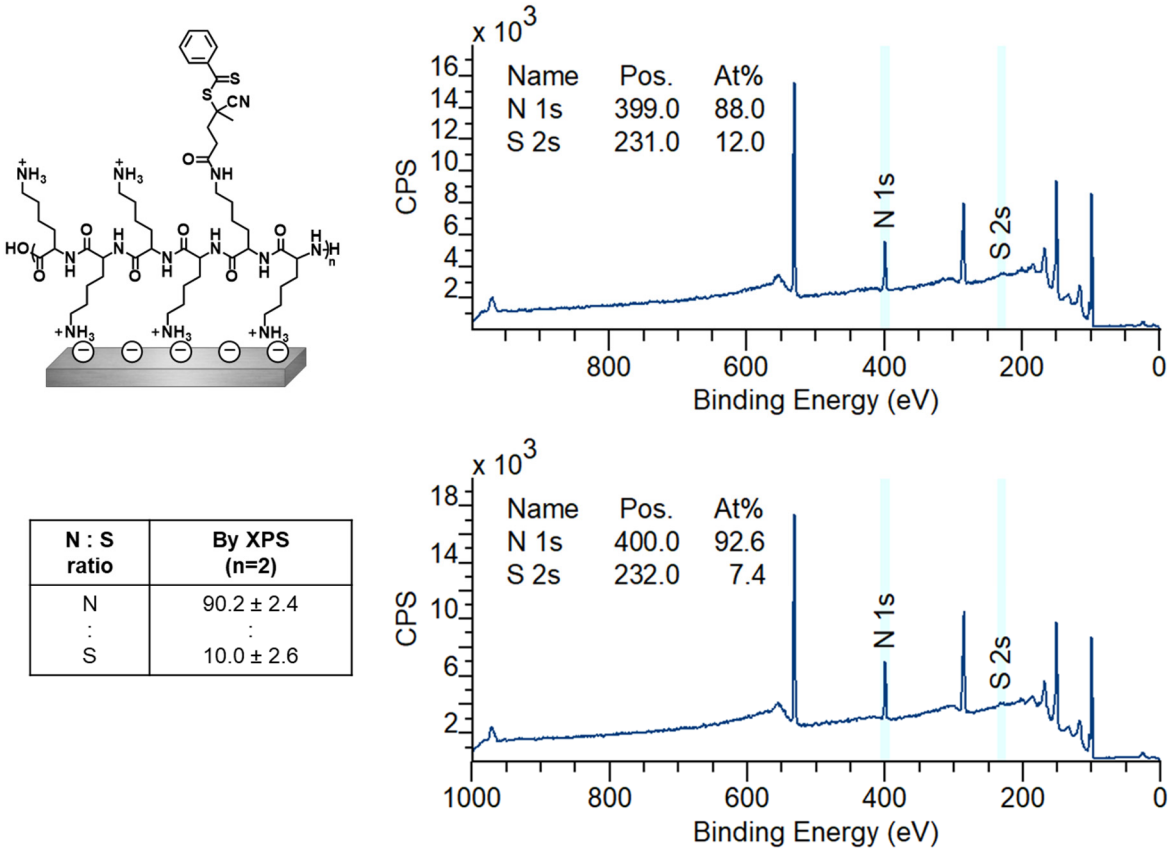


Figure S8. Structure and XPS wide scan of surface-immobilized PLL, modified by NHS-RAFT (according to route A, step 2).

From the above XPS spectrum, the conversion of the reaction in which the terminal amine groups of the PLL were equipped with the RAFT agent can directly be calculated by considering the N/S atomic ratio. This N_{1s} : S_{2p} ratio was found to be 74.9 : 25.1, which equals 6N : 2S. Since the RAFT initiator contains 1N, this leaves 5N originating from PLL, corresponding to 2.5 lysine monomers. This gives a RAFT : lysine ratio of 1 : 2.5, which translates to an on-surface conversion of roughly 40%.

2.4 XPS data and calculations PLL-RAFT conversion (route B, step B1)



N : S ratio	By XPS (n=2)
N : S	90.2 ± 2.4 : 10.0 ± 2.6

Figure S9. Structure and XPS wide scan spectra data of B1) surface immobilized PLL-RA, *in duplo*. The table contains the average atomic ratios as determined by XPS.

The found N_{1s} : S_{2p} ratio was found to be approximately 90 : 10, which equals 18N : 2S. Since the RAFT agent contains 1N, this leaves 17N originating from PLL, corresponding to 8.5 lysine monomers. This gives a RAFT : lysine ratio of 1 : 8.5.

2.5 GPC data

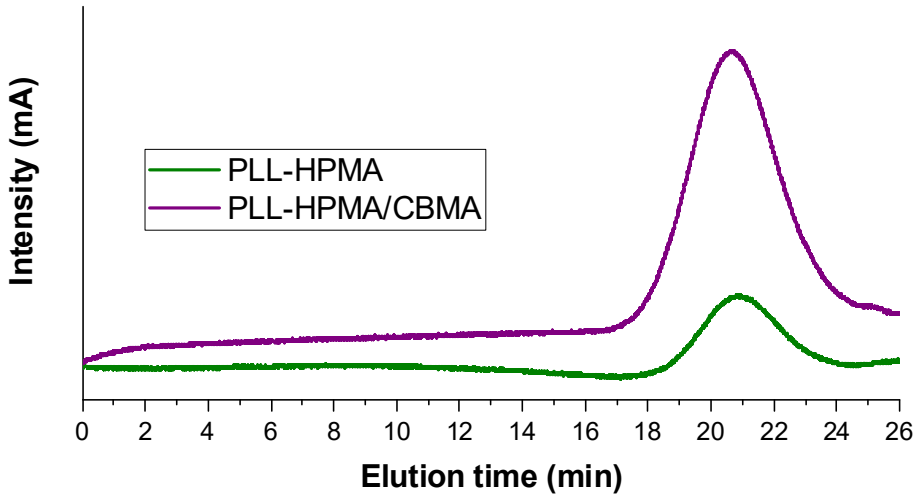


Figure S10. GPC traces of both bottlebrush polymers PLL-HPMA and PLL-HPMA/CBMA.

Table S1. Number-averaged and weight-averaged molecular weight and polydispersity index (PDI) of the synthesized bottlebrush polymers by GPC.

Bottlebrush polymer	M_n (g/mol)	M_w (g/mol)	PDI
PLL-HPMA	32000	43000	1.4
PLL-HPMA/CB	34000	53000	1.6

2.6 DLS data

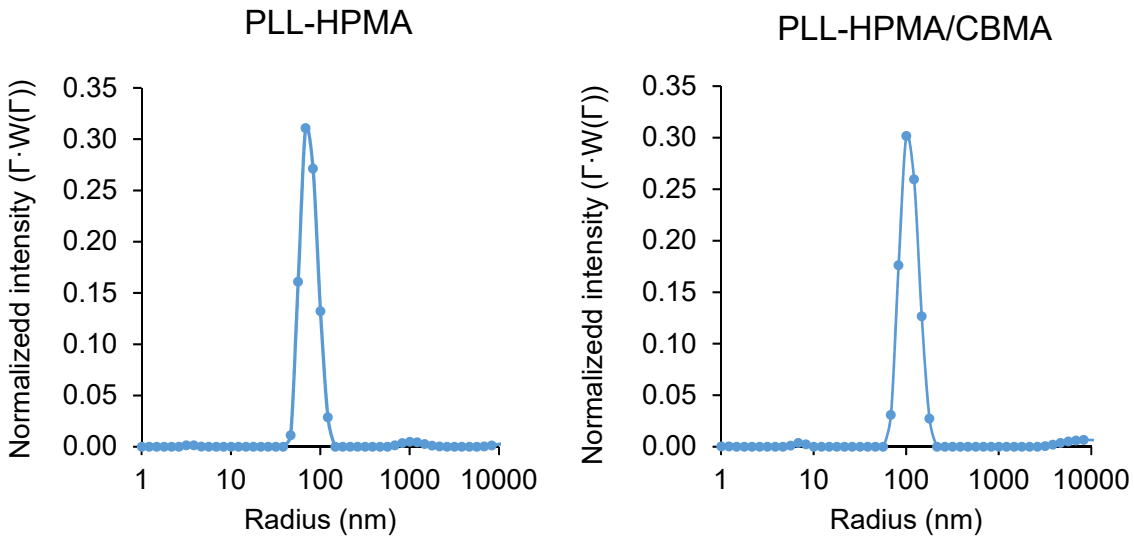


Figure S11. DLS data of PLL-HPMA and PLL-HPMA/CBAA bottlebrushes in Milli-Q water (prepared via route C). The contribution to the scattering is plotted against the hydrodynamic radius as intensity-based equal area representation as previously published.[S6]

The average radius (R_{avg}) was calculated by applying the following equation:

$$\boldsymbol{R_{avg}} = \frac{\sum_i \boldsymbol{n_i \cdot R_i}}{\sum_i \boldsymbol{n_i}} \qquad \text{(eq. S1)}$$

with:

R_{avg} = average radius

n_i = normalized intensity at given radius

R_i = radius at given intensity

Applying this equation, radiuses of 77 nm and 111 nm were found for PLL-HPMA and PLL-HPMA/CBMA, respectively.

2.7 AFM data

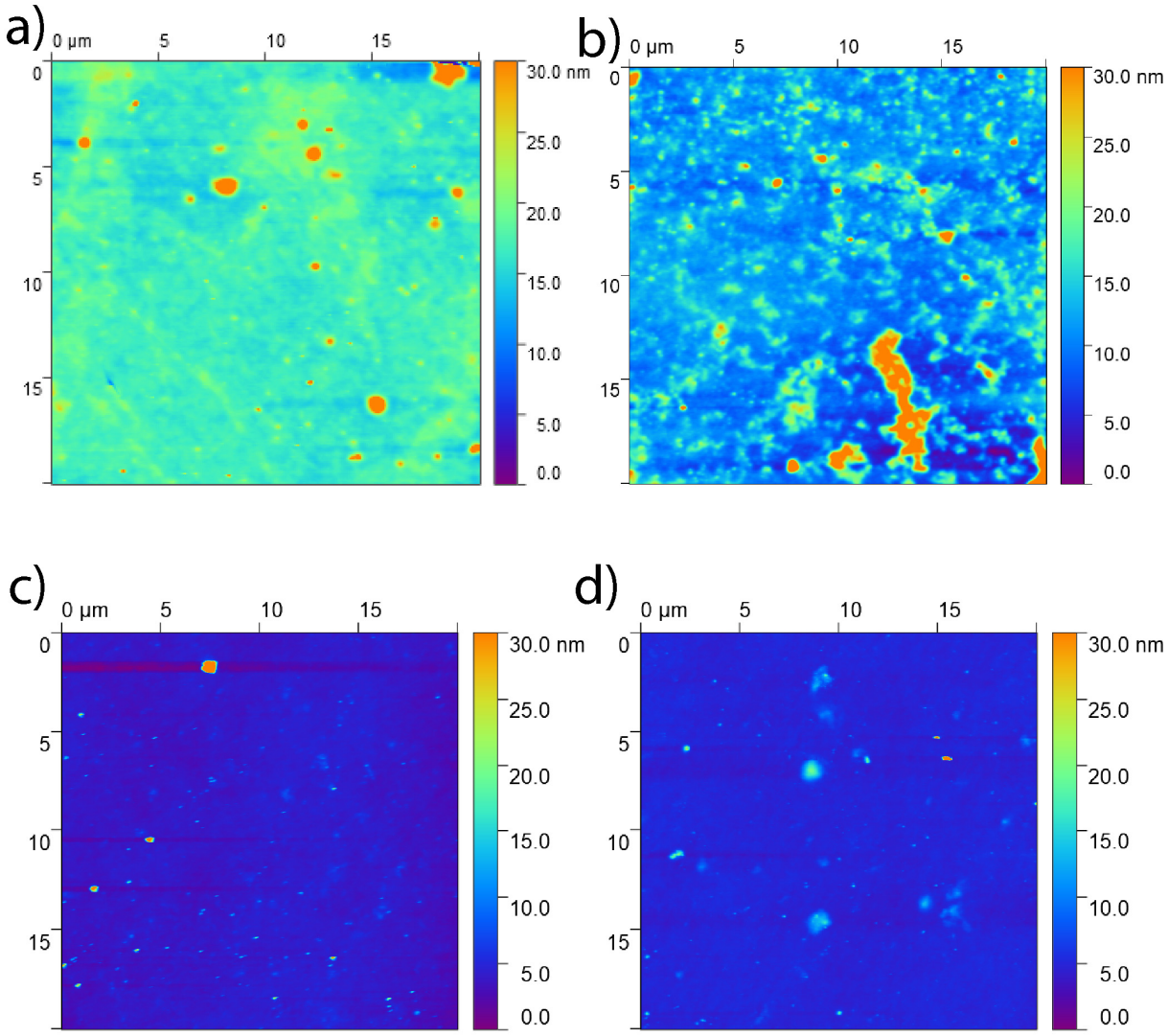


Figure S12. AFM surface topology; **a)** coating A3, **b)** coating B2, **c)** route C1 PLL-HPMA, **d)** route C1 PLL-HPMA/CBMA. The polymerization time for all three routes was 80 min.

3 References

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