Supplemental Materials for

Winning the fight against biofilms: First six-month study showing no biofilm formation on zwitterionic polyurethanes

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This file includes:

Synthesis of poly(carboxybetaine) degradable urethane (PCBDU)

1. **Synthesis of PCBGU-100**

The PU-prepolymer (DEAEA with HDI) was synthesized in a three-necked round bottom flask equipped with a mechanical stirrer, temperature controller, and a nitrogen inlet. Before synthesis, DEAEA (6.1 g, 0.03 mole) was placed into a vacuum oven at 110℃ for 2 hours to remove moisture. Once the DEAEA was placed in the flask, HDI (5 g, 0.03 mole) was added dropwise. Anhydrous DMF was added once the viscosity increased, approximately a half-hour later. This solution was stirred at 400 rpm for 2 hours at 80℃. Subsequently, the resulting solution was poured into PTEF dishes and stored in the oven at 100℃ for 12 hours without vacuum. The resulting polyurethane film was dried at 100℃ in the oven under vacuum for another 12 hours to remove residual solvent.

2. **Synthesis of PCBGU-75**

The PU-prepolymer (DEAEA with HDI) was synthesized in a three-necked round bottom flask equipped with a mechanical stirrer, temperature controller, and a nitrogen inlet. Before synthesis, DEAEA (9.11 g, 0.04 mole) was placed into a vacuum oven at 110℃ for 2 hours to remove moisture. Once the DEAEA was placed in the flask, HDI (10 g, 0.059 mole) was added dropwise. Anhydrous DMF was added once the viscosity increased, approximately a half-hour later. This solution was stirred at 400 rpm for 2 hours at 80℃ before glycerol (0.91 g, 9.87 mmole) was added dropwise. After another 30 minutes of stirring at the same rate and temperature, the mixed solution was poured into PTEF dishes and stored in the oven at 100℃ for 12 hours without vacuum. The resulting polyurethane film was dried at 100℃ in the oven under vacuum for another 12 hours to remove residual solvent.

3. Synthesis of PCBGU-50

The PU-prepolymer (DEAEA with HDI) was synthesized in a three-necked round bottom flask equipped with a mechanical stirrer, temperature controller, and a nitrogen inlet. Before synthesis, DEAEA (6.08 g, 0.03 mole) was placed into a vacuum oven at 110℃ for 2 hours to remove moisture. Once the DEAEA was placed in the flask, HDI (10 g, 0.059 mole) was added dropwise. Anhydrous DMF was added once the viscosity increased, approximately a half-hour later. This solution was stirred at 400 rpm for 2 hours at 80℃ before glycerol (1.82 g, 0.02 mole) was added dropwise. After another 30 minutes of stirring at the same rate and temperature, the mixed solution was poured into PTEF dishes and stored in the oven at 100℃ for 12 hours without vacuum. The resulting polyurethane film was dried at 100℃ in the oven under vacuum for another 12 hours to remove residual solvent.

4. Synthesis of PCBGU-25

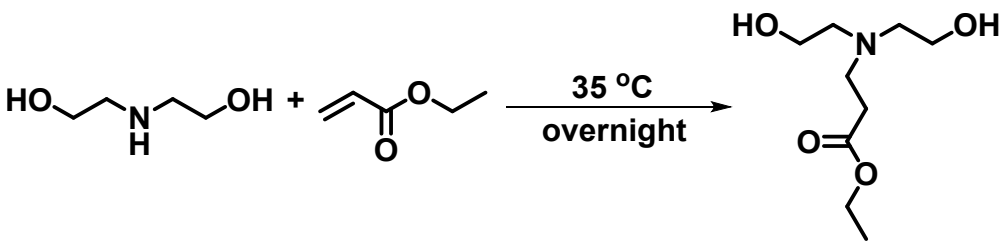
The PU-prepolymer (DEAEA with HDI) was synthesized in a three-necked round bottom flask equipped with a mechanical stirrer, temperature controller, and a nitrogen inlet. Before synthesis, DEAEA (3.04 g, 0.015 mole) was placed into a vacuum oven at 110℃ for 2 hours to remove moisture. Once the DEAEA was placed in the flask, HDI (10 g, 0.059 mole) was added dropwise. Anhydrous DMF was added once the viscosity increased, approximately a half-hour later. This solution was stirred at 400 rpm for 2 hours at 80℃ before glycerol (2.73 g, 0.03 mole) was added dropwise. After another 30 minutes of stirring at the same rate and temperature, the mixed solution was poured into PTEF dishes and stored in the oven at 100℃ for 12 hours without vacuum. The resulting polyurethane film was dried at 100℃ in the oven under vacuum for another 12 hours to remove residual solvent.

5. Synthesis of PGU

PGU was synthesized in a three-necked round bottom flask equipped with a mechanical stirrer, temperature controller, and a nitrogen inlet. Before synthesis, glycerol (3.63 g, 0.039 mole) was placed into a vacuum oven at 110℃ for 2 hours to remove moisture. Once the glycerol was placed in the flask, HDI (10 g, 0.059 mole) was added dropwise. Anhydrous DMF was added once the viscosity increased, approximately a half-hour later. This solution was stirred at 400 rpm for 2 hours at 80℃. Subsequently, the resulting solution was poured into PTEF dishes and stored in the oven at 100℃ for 12 hours without vacuum. The resulting polyurethane film was dried at 100℃ in the oven under vacuum for another 12 hours to remove residual solvent.

Synthesis of Poly(ethylene glycol glycerol crosslinked) urethane (PEGCU-50)

The PU-prepolymer (PEG 2000 with HDI) was synthesized in a three-necked round bottom flask equipped with a mechanical stirrer, temperature controller, and a nitrogen inlet. Before synthesis, PEG (20 g, 0.01 mole) was placed into a vacuum oven at 110℃ for 2 hours to remove moisture. Once the PEG 2000 was placed in the flask, HDI (3.36 g, 0.02 mole) was added dropwise. Anhydrous DMF was added once the viscosity increased, approximately a half-hour later. This solution was stirred at 400 rpm for 2 hours at 80℃ before glycerol (0.61 g, 6.67 mmole) was added dropwise. After another 30 minutes of stirring at the same rate and temperature, the mixed solution was poured into PTEF dishes and stored in the oven at 100℃ for 12 hours without vacuum. The resulting polyurethane film was dried at 100℃ in the oven under vacuum for another 12 hours to remove residual solvent.



Scheme S1. Synthetic route for DEAEA.

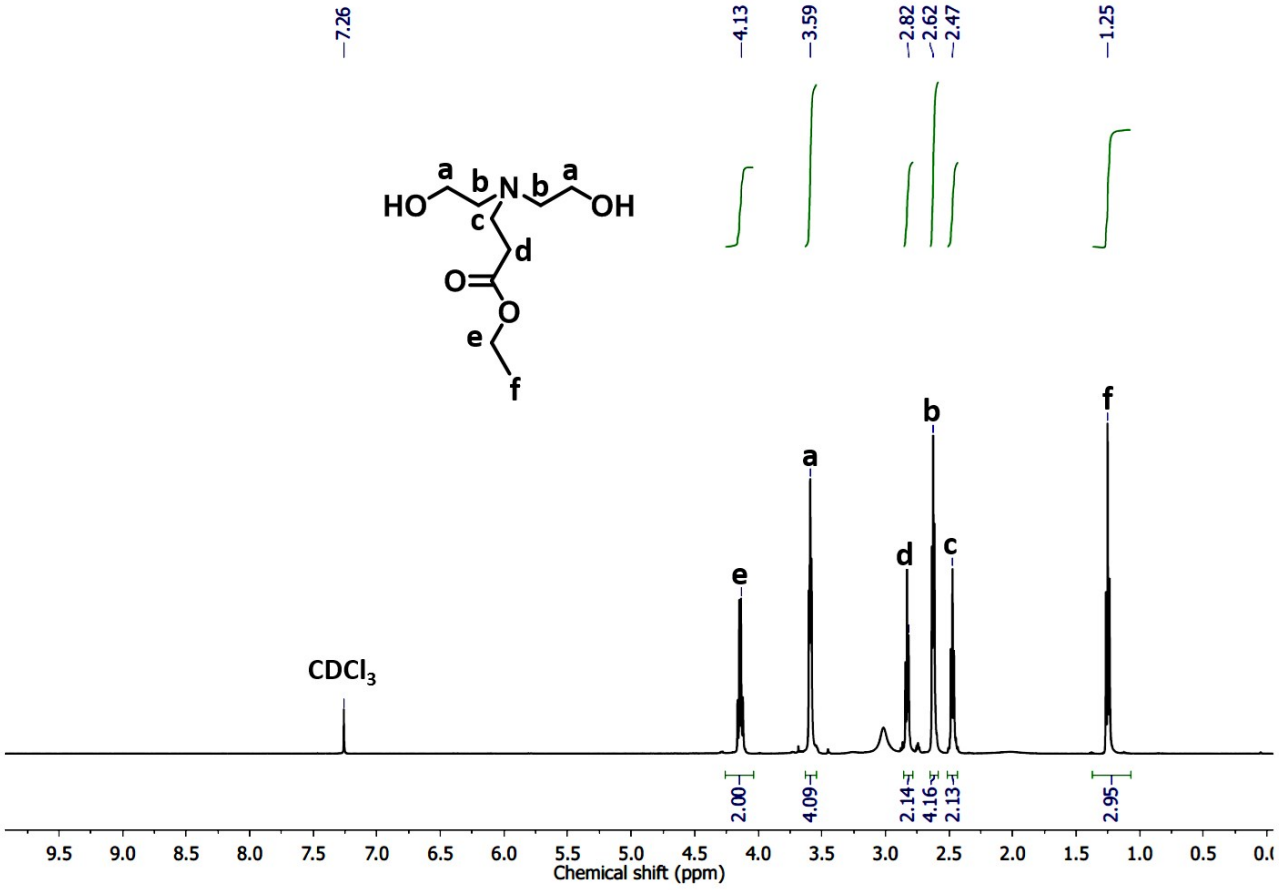


Figure S1. ^{1}H NMR spectrum for DEAEA.

Table S1. Mole percentage and weight percent of diol in each sample.

	CB-diol (mol%)	Glycerol (mol%)	PEG_{2000} (mol%)	CB-diol (wt%)	Glycerol (wt%)	PEG_{2000} (wt%)
PCBGU-100	100	\	\	100	\	\
PCBGU-75	75	16.67	\	90.93	9.07	\
PCBGU-50	50	33.33	\	76.98	23.02	\
PCBGU-25	25	50	\	52.71	47.29	\
PGU	\	100	\	\	100	\
PEGCU-50	\	33.33	50	\	2.98	97.02

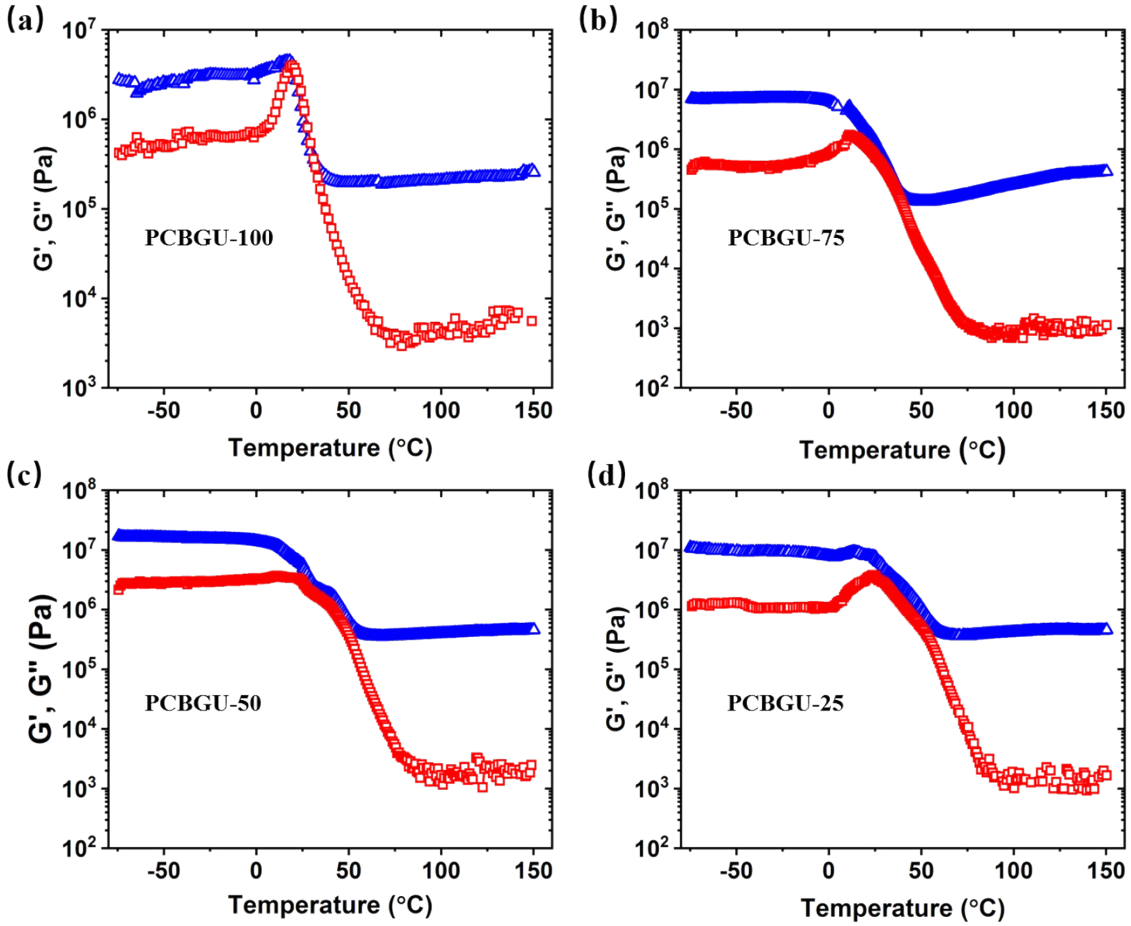


Figure S2. Temperature-sweep rheological behavior of (a) PCBGU-100 (b) PCBGU-75 (c) PCBGU-50 and (d) PCBGU-25 from -75 to 150 ^{o}C.

Table S2. Surface coverage of biofilm on PCBGUs and control surfaces with different times immersed in *P. aeruginosa* solution.

	Surface coverage (%)					
	API-PU	PEGCU-50	PCBGU-100	PCBGU-75	PCBGU-50	PCBGU-25
3 Day	85.41	28.53	0	0	0	0
1 Week	95.26	45.93	0	0	0	0
2 Week	96.95	92.115	0	0	0	0
3 Month	98.86	96.917	0	0	0	0
6 Month	99.68	98.51	0	0	0	0

Table S3. Surface coverage of biofilm on PCBGUs and control surfaces with different times immersed in *S. epidermidis* solution.

	Surface coverage (%)					
	API-PU	PEGCU-50	PCBGU-100	PCBGU-75	PCBGU-50	PCBGU-25
3 Day	91.76	11.76	0	0	0	0
1 Week	92.20	80.97	0	0	0	0
2 Week	96.48	91.01	0	0	0	0
3 Month	95.72	95.09	0	0	0	0
6 Month	99.93	99.56	0	0	0	0

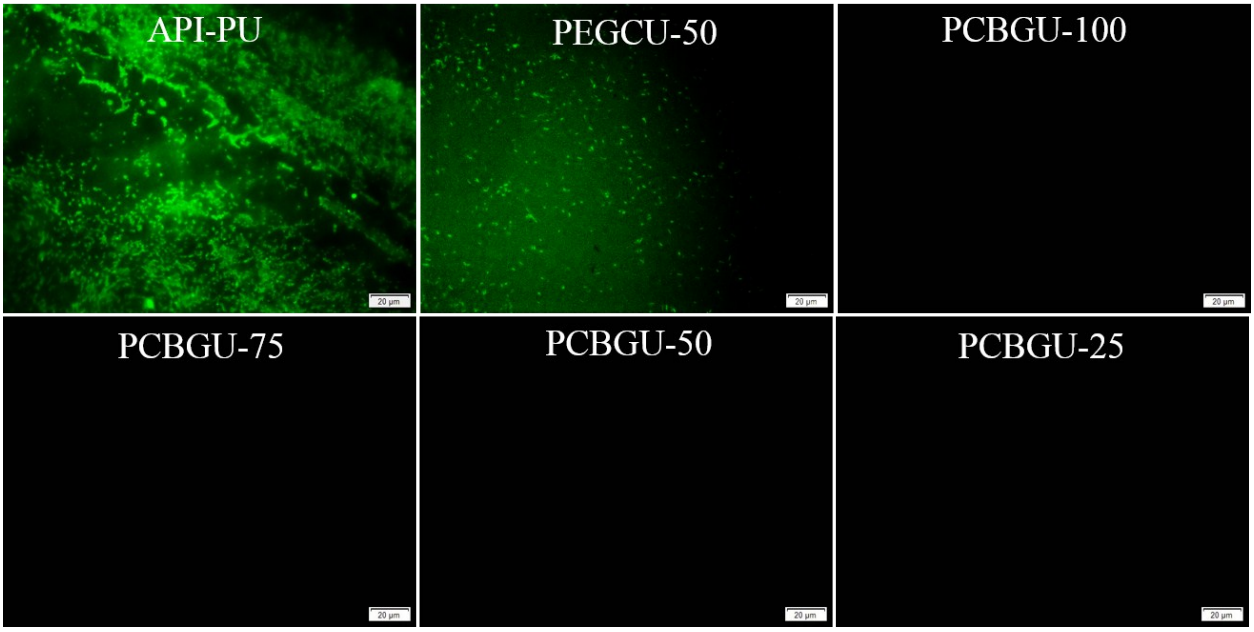


Figure S3. Fluorescence images for the biofilm formation *P. aeruginosa* PAO1 after 3 days on PCBGU-100, PCBGU-75, PCBGU-50, and PCBGU-25 hydrogels. Commercially available API-PU and PEGCU-50 were used as control materials.

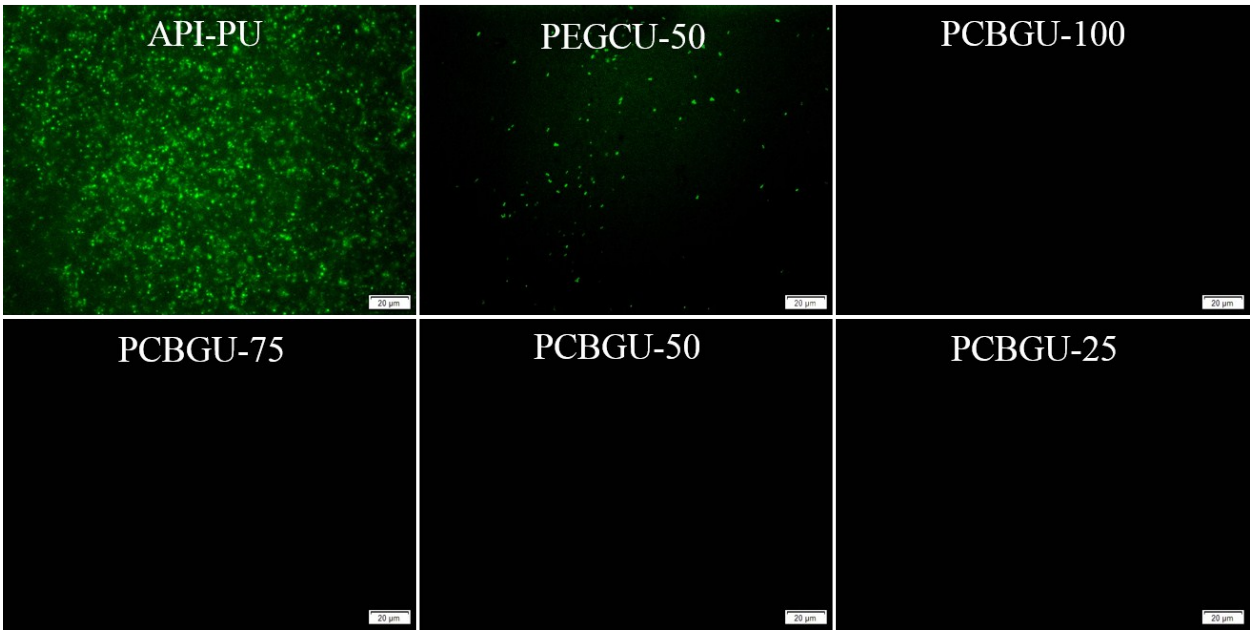


Figure S4. Fluorescence images for the biofilm formation *S. epidermidis* after 3 days on PCBGU-100, PCBGU-75, PCBGU-50, and PCBGU-25 hydrogels. Commercially available API-PU and PEGCU-50 were used as control materials.