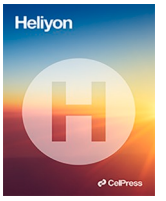
Heliyon 10 (2024) e26347

Contents lists available at ScienceDirect

Heliyon

journal homepage: www.cell.com/heliyon



Research article

Superior antibacterial surfaces using hydrophilic, poly(MPC) and poly(mOEGMA) free chains of amphiphilic block copolymer for sustainable use

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A R T I C L E I N F O

Keywords:
Antibacterial coatings
Block copolymers
Zwitterionic free chain
Environmental use

A B S T R A C T

Surface modification of electrically neutral hydrophilic polymers is one of the most promising methods for preventing biofouling and biological contamination by proteins and bacteria. Surface modification of inorganic materials such as silica-based glass can render them more durable and thus help in achieving the sustainable development goals. This study reports a novel method for the simple and effective surface modification of glass surfaces with amphiphilic block copolymers possessing the silane coupling segment composed of 3-(methacryloyloxy)propyltris (trimethylsilyloxy) silane and 3-methacryloxypropyltrimethoxysilane. The ability of hydrophilic segments composed of either 2-methacryloyloxyethyl phosphorylcholine (MPC) or poly(ethylene glycol) methyl ether methacrylate (mOEGMA) to prevent bacterial adhesion was investigated. The target block copolymers were prepared by reversible addition-fragmentation chain transfer polymerization and the monomer units of the hydrophilic segments were controlled to be either 120 or 160. The polymers were modified on the substrate by dip-coating. Contact angle measurements indicated that the block copolymer with the PMPC hydrophilic segment formed a hydrophilic surface without pre-hydration, while those with the PmOEGMA hydrophilic segment-coated surface became hydrophilic upon immersion in water. The block copolymer-coated surfaces decreased *S. aureus* adhesion, and a significant reduction was observed with the MPC-type block copolymer. The following surface design guidelines were thus concluded: (1) the block copolymer is superior to the random copolymer and (2) increasing the hydrophilic segment length further decreases bacterial adhesion.

1. Introduction

In light of the current global environmental issues, products with long lifetimes have been gaining significant attention for achieving sustainable development goals (SDGs) [1]. Most of the currently employed products are in contact with water or are exposed to biological agents (bacteria, viruses, and proteins) as well as organic (nutrients) and inorganic matter (mineral ions), because water dissolves various substances. Notably, bacterial adhesion initiates the formation of biofilms which can cause serious environmental

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https://doi.org/10.1016/j.heliyon.2024.e26347
Received 16 January 2024; Received in revised form 9 February 2024; Accepted 12 February 2024
Available online 16 February 2024

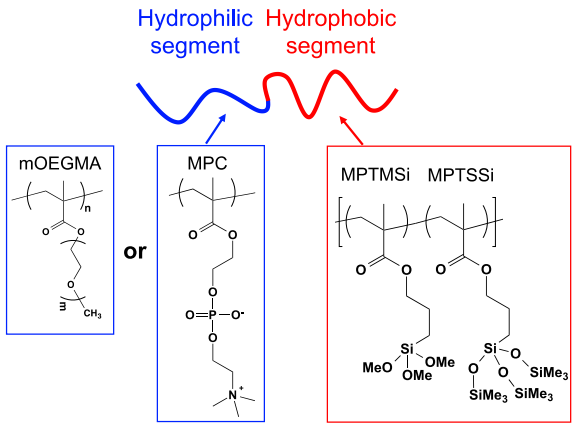

Fig. 1. Chemical structure of the amphiphilic block copolymers with the silane coupling segment. The hydrophilic segment was either PMPC or PmOEGMA. The silane coupling segment was P(MPTMSi-*r*-PMPTSSi).

problems [2–4]. Consequently, inhibiting the biofouling of materials to extend their lifetime while maintaining their original function can assist in achieving the SDGs. One promising method involves the modification of the surface of the material with anti-biofouling polymers so it can be employed in medical devices such as artificial hearts and joints [5–9].

Silica-based glass is an inorganic solid material which has been widely employed in electronic and optical devices in addition to construction materials owing to its lifetime which is much longer than that of organic materials. The glass surface is hydrophilic and negatively charged because the material mostly consists of oxides, and water droplets pass beneath both water-soluble and oil-based contaminants and wash them away. Consequently, hydrophilicty can induce anti-fouling properties [10]; however, suppressing protein and bacteria biofouling on glass surfaces remains challenging. Several studies have investigated the use of antibiofouling materials on glass surfaces to modify electrically neutral hydrophilic polymer chains [11–14]. For example, coatings of polymeric materials comprising poly (polyethylene glycol) (PEG) have been intensively investigated [15,16]. Yoshikawa et al. demonstrated that a poly[poly(ethylene glycol)methyl ether methacrylate] (PPEGMA) brush can prevent biofouling and have blood compatibility [17]. Zwitterionic polymers, including poly(sulfobetaine methacrylate), poly(carboxybetaine methacrylate), and poly (2-methacryloyloxyethyl phosphorylcholine) (PMPC) have excellent hydrophilicity and anti-biofouling properties [18–21].

Surface chemistry enables the modification of hydrophilic polymers on material surfaces [22]. Controlled radical polymerizations such as surface-initiated atom transfer radical polymerization (SI-ATRP) enables the formation of densely packed polymer brush surfaces with the grafting density over 0.1 chains nm^{-2} [23]. Moreover, the protein adsorption in the zwitterionic PMPC brush surfaces decreased significantly with increasing graft density and/or chain length [24]. In these ways, dense polymer brush surfaces are favorable for obtaining anti-biofouling properties [12,24–26]; however, this approach is expensive and cannot be performed on a large scale. Hence, the "grafting to" method using an anchoring moiety has been gaining attention as a promising method to coat hydrophilic polymers. Our group has previously reported the synthesis of a block copolymer composed of a PMPC segment and hydrophobic silane coupling segment of poly(3-methacryloxypropyl trimethoxysilane-*random*-3-(methacryloyloxy)propyl-tris(tri(methylsilyloxy))silane) (P(MPTMSi-*r*-MPTSSi)) [27]. The P(MPTMSi-*r*-MPTSSi) segment enabled a stable bonding with the glass surface via silane coupling and the polymer (PMPC-*b*-P(MPTMSi-*r*-MPTSSi)) was successfully coated onto glass substrates by simple dip coating. Moreover, the obtained surface exhibited hydrophilic and protein-repellent properties which can suppress bacterial adhesion. Nevertheless, the polymer design strategy requires further investigation.

This study reports the synthesis of a polymer coating for the suppression of bacterial adhesion. The polymer was composed of a hydrophilic polymer segment and a hydrophobic silane coupling P (MPTMSi-*r*-MPTSSi) segment (Fig. 1). The hydrophilic segment was designed to be either PMPC or poly(oligo(ethylene glycol) methyl ether methacrylate) (PmOEGMA), and the segment length was controlled by reversible addition–fragmentation chain transfer (RAFT) polymerization. The surface wettability and antibiofouling properties were investigated by comparing them with those of random copolymers. From these investigations, surface design guidelines for suppressing bacterial adhesion were deduced.

2. Experimental section

2.1. Materials

MPC was purchased from NOF (Tokyo, Japan) while MPTSSi and MPTMSi were purchased from Shin-Etsu Chemical Co., Ltd. (Tokyo, Japan). The mOEGMA (molecular weight:300 Da) was purchased from Sigma-Aldrich (St. Louis, MO, USA). 4-Cyano-4-(phenyl-carbonothioylthio)pentanoic acid (CPD) was purchased from Strem Chemicals, Inc. (Newburyport, MA, USA). An Si wafer (10 nm-thickness SiO_2 (Si/SiO_2)) was purchased from Furuuchi Chemical Co. (Tokyo, Japan). A μ-bicinchoninic acid (BCA) protein assay kit was purchased from Thermo Fisher Scientific Inc. (IL, USA). All other chemicals were purchased from FUJIFILM Wako Pure

Table 1
Characterization of the amphiphilic block copolymers.

Code[a]	MPC or mOEGMA/MPTMSi/MPTSSi		M_w[c]	M_w/M_n[c]
	In feed	In copolymer[b]		
r120-MPC	120/40/40	112/36/34	9.5×10^4	1.6
b120-MPC	120/40/40	114/36/31	8.4×10^4	1.5
b160-MPC	160/20/20	158/18/16	8.3×10^4	1.5
r120-PEG	120/40/40	147/27/27	2.5×10^4	1.3
b120-PEG	120/40/40	119/39/40	1.5×10^4	1.3
b160-PEG	160/20/20	155/24/24	2.1×10^4	1.2

[a] The samples are referred to as b (or r)X–Y, where X and Y indicate the monomer unit number of hydrophilic segment, and the hydrophilic segment composition, respectively. "r" and "b" indicate "random copolymer" and "block copolymer", respectively.
[b] bDetermined by ^{1}H NMR.
[c] Determined using GPC.

Chemical Industry (Osaka, Japan) unless otherwise noted.

Block and random copolymers were synthesized and characterized based on a previously described protocol [27]. Briefly, hydrophilic and hydrophobic silane coupling segments were synthesized by RAFT polymerization using CPD as the RAFT agent (Scheme S1). The average molecular weight and polydispersity index were determined by gel permeation chromatography (GPC, Jasco). Poly (methyl methacrylate) was used as a standard to calculate the molecular weight. The monomer composition was determined by ^{1}H NMR (ECS-400, JEOL).

2.2. Modification of the copolymers onto the surface of substrates

The Si/SiO_2 and glass substrates were successively washed via ultrasonication in hexane, ethanol, and acetone. The substrates were further cleaned by an oxygen plasma treatment at 600 mTorr (PDC-001, Harrick Plasma). An ethanol solution containing the polymer (0.2 wt%) was mixed with an aqueous acetic acid solution (0.1 M) to prepare a coating solution with a ratio of 9:1 (v/v). The cleaned substrates were immersed in the coating solution for 2 h. After removing the coating solution, the substrates were dried and then heated at 70 °C for 3 h to proceed the silane coupling reaction. The substrates were washed with ethanol to remove the unreacted polymers.

2.3. Characterization of the polymer-coated surfaces

The thicknesses of the polymer-coated surfaces on the Si/SiO_2 substrates were quantitated using spectroscopic ellipsometry (alpha-SE, J. A. Woollam) in air. The obtained data were fitted using the Cauchy layer model to calculate the thickness. The static contact angles of water on the polymer-coated surfaces were measured using a contact-angle meter (CA-W, Kyowa Interface Science Co.). Dynamic contact angles were measured using the extension/contraction method.

2.4. Protein adsorption assay

Polymer-coated glass substrates (1 × 4 cm) were immersed in a phosphate-buffered saline (PBS) solution containing fibrinogen (0.1 mg/mL) at 37 °C for 3 h. The substrates were then rinsed with PBS, followed by ultrasonication in PBS containing sodium dodecyl sulfate (0.5 wt%) as to completely collect the adsorbed protein from the substrates. The μ-BCA protein assay was performed to determine the protein concentration. A plate reader (ARVO X3, PerkinElmer) was used to measure the absorbance at 560 nm.

2.5. Bacterial adhesion test

Staphylococcus aureus (*S. aureus*) (NBRC13276, Biotechnology Center, National Institute of Technology and Evaluation) was used to analyze the suppression of bacterial adhesion. The bacteria stock were kept at −80 °C before use, and transferred to a Trypticase soy broth (TSB) medium (500 μL) to proceed pre-culture at 37 °C overnight. The bacteria were further cultured in a TSB medium (25 mL) with gentle shaking, and the cell density was adjusted to 10^7 cells/mL ($OD_{600} = 0.02$). The polymer-coated glass substrates were then immobilized in a culture dish. The bacterial suspension of 10^7 cells/mL in TSB was loaded into it, followed by incubation at 37 °C for 3 h. The culture medium was repeatedly replaced with PBS. Adhered bacteria were fixed in PBS containing paraformaldehyde (4%), followed by methylene blue staining. The samples were observed by optical microscopy (CKX53FLPH/DP74, Olympus). Bacterial coverage was analyzed using an ImageJ software.

2.6. Observation of adhered bacteria by scanning electron microscopy (SEM)

The *S. aureus* suspension in TSB (10^7 cells/mL) was loaded onto a polymer-coated glass substrate in a culture dish and then incubated at 37 °C for 24 h. The substrates were rinsed with TSB. Adhered *S. aureus* cells were fixed in a glutaraldehyde solution (2.5

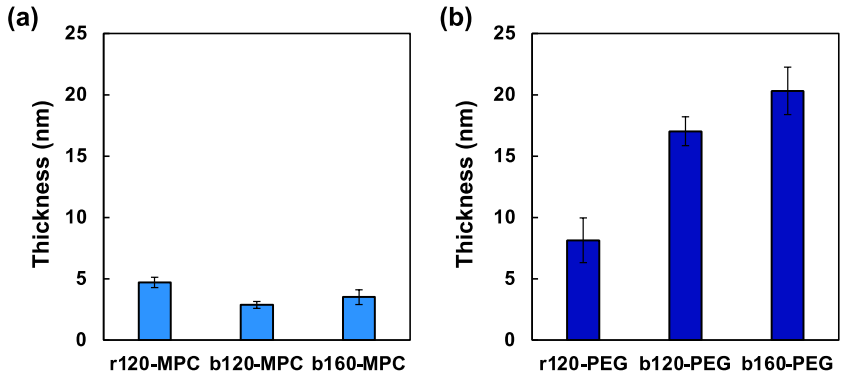

Fig. 2. Thicknesses of the obtained block copolymer-modified surfaces, (a) MPC-type and (b) PEG-type. The data are presented as the mean ± SD (n = 3).

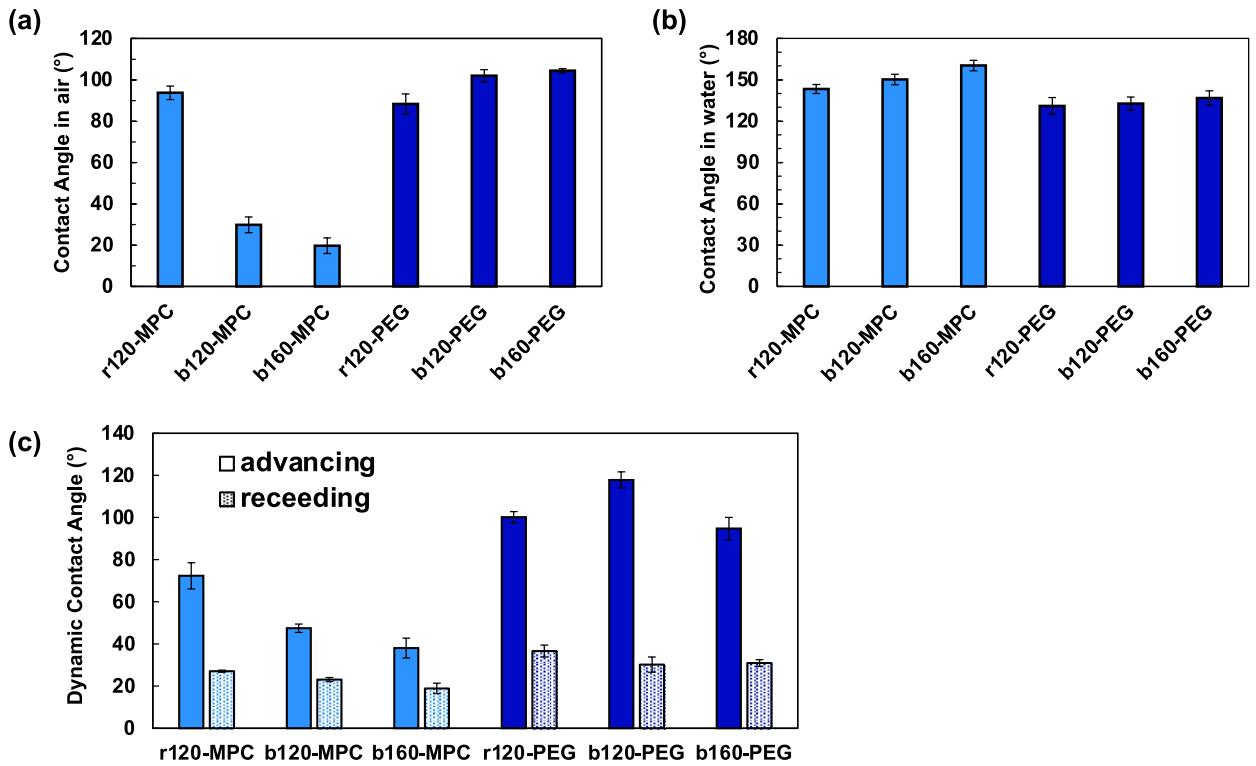

Fig. 3. (a) Static contact angles of water in air, (b) static contact angles of air in water, and (c) dynamic contact angles of water of the block copolymer-modified surfaces. For each sample, the left and right bars indicate the advancing and receding contact angles, respectively. The data are presented as the mean ± SD (n = 3).

vol%). The samples were immersed in 50, 75, 95, and 100% (v/v) ethanol solutions, followed by vacuum drying. The samples were stained with osmium (VIII) oxide and observed using SEM (JSM-7000F, JEOL).

3. Results and discussion

The target amphiphilic block copolymers were composed of a hydrophilic segment (either PMPC or PmOEGMA) and a hydrophobic-silane coupling segment (P(MPTMSi-*r*-MPTSSi)). The block copolymers were synthesized by the two-step RAFT polymerization [27]. The hydrophilic segment (PMPC or PmOEGMA) was firstly polymerized, followed by the second RAFT polymerization of MPTMSi and MPTSSi (the detail shown in Supporting Information). To compare the copolymer structures, ternary random copolymers composed of hydrophilic monomers (MPC or mOEGMA), MPTMSi, and MPTSSi were synthesized via RAFT polymerization. Table 1 summarizes the monomer composition, weight-averaged molecular weight (M_w), and polydispersity index (M_w/M_n, M_n:

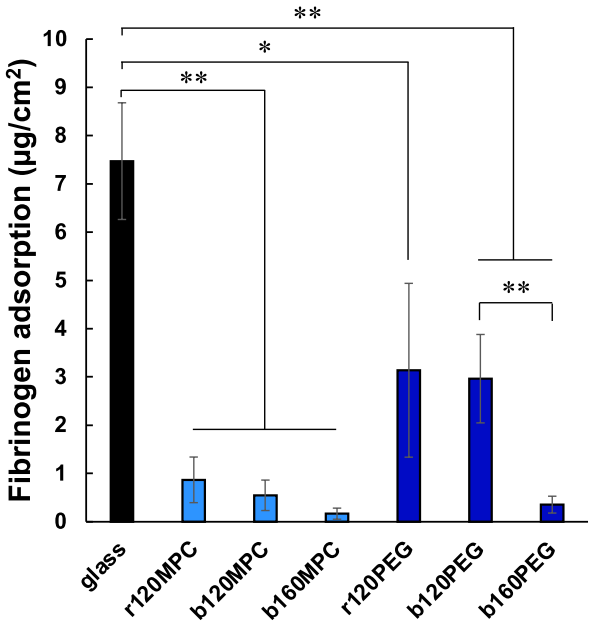

Fig. 4. Amount of the fibrinogen adsorbed on the block copolymer-modified surfaces (*$p < 0.05$, **$p < 0.01$.). The data are presented as the mean ± SD ($n = 3$).

number-averaged molecular weight) of each polymer. The samples were annotated as b (or r) X–Y, where X and Y indicate the monomer unit number of the hydrophilic segment and hydrophilic segment composition, respectively, while "r" and "b" indicate "random copolymer" and "block copolymer," respectively. The compositions and molecular weight distributions of the obtained copolymers were successfully controlled. In this manner, block and random copolymer structures were designed, and the monomer units in the hydrophilic segment of the block copolymer were controlled to be either 120 or 160. These parameters were investigated to clarify the effects of the polymer coatings on the surface properties and bacterial adhesion suppression.

The obtained copolymers were coated onto the surfaces of Si/SiO_2 and glass substrates. The thickness of the coating obtained on Si/SiO_2 was quantitatively determined using spectroscopic ellipsometry (Fig. 2). Regardless of the polymer structure, the thickness of the MPC-type copolymer was approximately 5 nm (Fig. 2(a)). The block copolymers with the PmOEGMA hydrophilic segment were thicker than those with the PMPC hydrophilic segment (Fig. 2(b)). The zwitterionic group can interact with charged surfaces, such as SiO_2 or glass surfaces, via dipole-dipole or ion-dipole interactions [28,29]. Unlike the PEG-type copolymers, the MPC-type copolymers could interact with substrates, which resulted in a several-nanometer-thick coating, regardless of the polymer structure of the MPC copolymers. Thus, nanoscale polymer coatings were successfully prepared.

The hydrophilic properties of a surface play a pivotal role in the potential application of the antifouling coating. Fig. 3(a) shows the water contact angles in air on the polymer-coated surfaces. For the MPC-type copolymers, the contact angle on the r120-MPC-coated surface was approximately 90° in air, whereas those on the b120-MPC- and b160-MPC-coated surfaces decreased to 30° and 20°, respectively. The r120-MPC-coated surface was hydrophobic because the hydrophobic moiety of MPTSSi was expressed on the surface in air to minimize the surface energy. The block copolymer-coated surfaces were hydrophilic because a sufficiently long PMPC segment covered the surface. For the PEG-type copolymers, the contact angles of the polymer-coated surfaces were approximately 100° regardless of the polymer structure. The PmOEGMA segment is thought to be expressed on the surface; however, the hydrophobic moiety in the ethylene glycol unit affects the hydrophobicity of the surface. The air contact angles of the polymer-coated surfaces in water (Fig. 3(b)) revealed that all the polymer-coated surfaces were hydrophilic in water. Importantly, PEG-type block copolymer surfaces exhibited hydrophilic properties in water. Although the surface coated with the amphiphilic copolymers was hydrophilic in air, the hydrophilic moieties in the copolymer were present on the surface in water. Overall, the hydrophilicity in water increased in the order of r120, b120, and b160.

To further evaluate the structural changes in air and water, the dynamic contact angles of the surfaces were evaluated (Fig. 3(c)). For the MPC-type copolymers, the advancing contact angle decreased in the order of r120-MPC, b120-MPC, and b160-MPC and so did the hysteresis which is the difference between the advancing and receding contact angles. This result indicated that the MPC-type block copolymer-coated surfaces were hydrophilic in air. The PEG-type copolymer-coated surfaces exhibited larger hysteresis compared with those of the MPC-type thus further confirming the structural change in the PEG-type block copolymer-coated surface between air and water. The changes in thickness between air and water were also investigated for the PEG-type copolymers (Fig. S1). The increase in the water thickness indicated that the polymer chains were swollen and extended in water, and consequently the b120-PEG and b160-PEG coated surfaces exhibited large hysteresis and became hydrophilic in water.

Subsequently, the adsorption of fibrinogen as a plasma protein was investigated. The copolymer coating successfully decreased fibrinogen adsorption compared to the bare glass (Fig. 4), as expected from the hydrophilic surfaces obtained by coating the

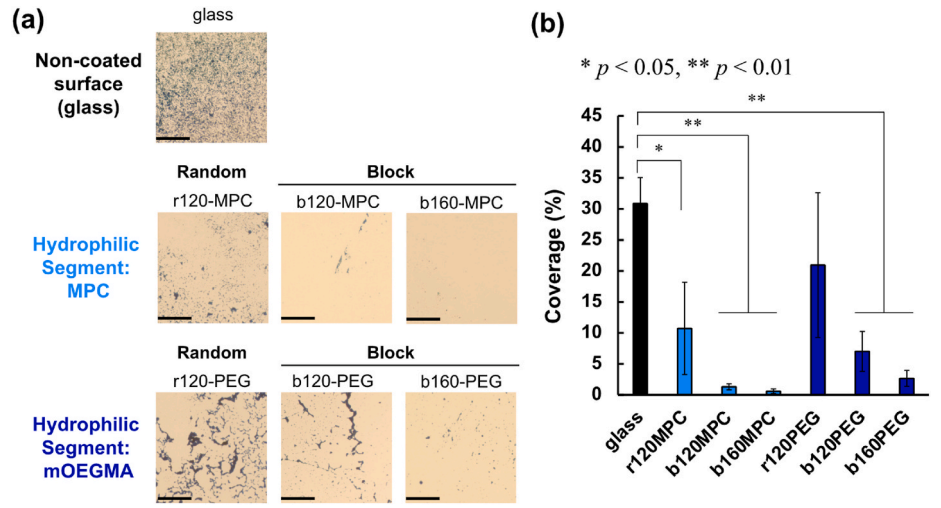

Fig. 5. (a) The microscope images of the adhered *S. aureus* on the block copolymer-modified surfaces incubated in TSB. Scale: 200 μm. (b) Bacterial coverage on the block copolymer-modified surfaces (*$p < 0.05$, **$p < 0.01$.). The data are presented as the mean ± SD ($n = 3$).

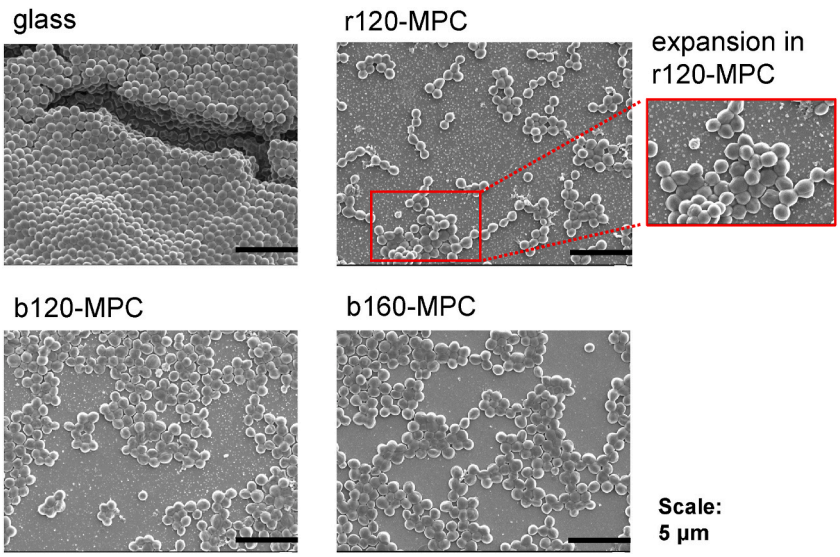

Fig. 6. SEM images of the adhered *S. aureus* on the MPC-type block copolymer-modified surfaces. The incubation was proceeded at 37 °C for 24 h. Scale: 5 μm.

amphiphilic copolymers. Among the MPC-type copolymers, fibrinogen adsorption decreased in the following order: r120-MPC b120-MPCand b160-MPC. Among the PEG-type copolymers, fibrinogen adsorption was significantly reduced by coating with b160-PEG. These results indicated that block copolymers with long hydrophilic segments can effectively decrease fibrinogen adsorption. Since the block copolymer coating decreased plasma protein adsorption, we expected it to decrease bacterial adhesion to the substrates.

Fig. 5(a) shows the microscopic images and coverage of *S. aureus* after the contact of the polymer-coated substrates with *S. aureus* in nutrient-rich TSB. The r120-MPC coating decreased *S. aureus* adhesion to some extent compared to bare glass. Moreover, MPC-type block copolymer (b120-MPC and b160-MPC) coatings dramatically suppressed *S. aureus* adhesion (Fig. 5(b)). The block copolymer also more effectively decreased the *S. aureus* adhesion than r120-PEG. Although the PEG-type block copolymer-coated surfaces were hydrophobic in air, they exhibited hydrophilic properties owing to the extension of the polymer chain in water, which enabled bacterial adhesion suppression. Bacterial adhesion further decreased upon increasing the hydrophilic segment length from b120-PEG to b160-PEG. This result is consistent with the decrease in fibrinogen adsorption (Fig. 4). From these investigations, the following surface design guidelines for bacterial adhesion suppression were obtained: (1) the block copolymer is superior to the random copolymer and (2) increasing the hydrophilic segment length further decreases bacterial adhesion.

We further investigated the anti-adhesive properties of bacteria on the polymer-coated surfaces. Fig. 6 shows the SEM images of adhered *S. aureus*, which was incubated in TSB at 37 °C for 24 h. A large number of *S. aureus* adhered and formed multiple layers on the

bare glass substrate; however, the bacterial adhesion dramatically decreased on the polymer-coated substrates even after incubation for 24 h. On the random copolymer-coated surface, *S. aureus* partially adhered and formed a multilayer (indicated by the red box in Fig. 6), while the adhesion of *S. aureus* was a monolayer on the block copolymer-coated surfaces. Thus, the block copolymer coatings successfully exhibited superior antibacterial properties.

4. Conclusions

This study reports the fabrication of bacterial adhesion-suppression coatings from amphiphilic block copolymers. The hydrophilic free chain (PMPC or PmOEGMA) acts as an antibacterial property, and hydrophobic polymer with a silane coupling unit (P(MPTMSi-*r*-MPTSSi)) works for bonding the substrate to become stable. The monomer units of the hydrophilic segments were controlled to be either 120 or 160 by RAFT polymerization. Characterization of the obtained polymer-coated surfaces indicated that the block copolymer with the hydrophilic PMPC segment formed a hydrophilic surface without prehydration, whereas those with the PmOEGMA hydrophilic segment-coated surface became hydrophilic upon immersion in water. The block-copolymer-coated surfaces successfully decreased *S. aureus* adhesion, whereas the MPC-type block copolymer resulted in a significant decrease in adhesion. By comparing the polymer structures, the following guidelines to suppress bacterial adhesion were indicated: (1) the block copolymer is superior to the random copolymer, and (2) increasing the hydrophilic segment length further decreases bacterial adhesion. Our amphiphilic block copolymer coating will be useful for silica-based glass in social infrastructure applications for long-term use, which can contribute to solving current global environmental issues for SDGs.

Data availability statement

Data will be available on request.

CRediT authorship contribution statement

Tsukuru Masuda: Writing – original draft, Visualization, Validation, Methodology, Investigation, Funding acquisition. **Shoichi Yoshizawa:** Validation, Methodology, Investigation. **Aya Noguchi:** Validation, Methodology, Investigation. **Yuta Kozuka:** Validation, Methodology, Investigation. **Norifumi Isu:** Validation, Methodology, Conceptualization. **Madoka Takai:** Writing – review & editing, Visualization, Validation, Supervision, Project administration, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

We thank the Japan Society for the Promotion of Science (JSPS) for providing support through a Grant-in-Aid for Scientific Research (B) (No. 20H02795 to T.M.). This work was partially supported by Grant-in-Aid for Transformative Research Areas (A) -KAKENHI- 21H05231. We thank Editage for the English grammar editing.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.heliyon.2024.e26347.

References

[1] L.X. Yong, J.K. Calautit, Sustainability 15 (2023) 3394.
[2] M.P. Schultz, J.A. Bendick, E.R. Holm, W.M. Hertel, Biofouling 27 (2011) 87–98.
[3] O. Habimana, A.J.C. Semiaõ, E. Casey, J. Membr. Sci. 454 (2014) 82–96.
[4] D. Lebeaux, J.M. Ghigo, C. Beloin, Microbiol. Mol. Biol. Rev. 78 (2014) 510–543.
[5] K. Ishihara, Langmuir 35 (2019) 1778–1787.
[6] K.M. Kovach, J.R. Capadona, A. Sen Gupta, J.A. Potkay, J. Biomed. Mater. Res. 102 (2014) 4195–4205.
[7] K. Nagahashi, Y. Teramura, M. Takai, Colloids Surf. B Biointerfaces 134 (2015) 384–391.
[8] Y. Takatori, T. Moro, M. Kamogawa, H. Oda, S. Morimoto, T. Umeyama, M. Minami, H. Sugimoto, S. Nakamura, T. Karita, J. Kim, Y. Koyama, H. Ito, H. Kawaguchi, K. Nakamura, J. Artif. Organs 16 (2013) 170–175.
[9] R. Ukita, K. Wu, X. Lin, N.M. Carleton, N. Naito, A. Lai, C.C. Do-Nguyen, C.T. Demarest, S. Jiang, K.E. Cook, Acta Biomater. 92 (2019) 71–81.
[10] S. Nishimoto, B. Bhushan, RSC Adv. 3 (2013) 671–690.
[11] A. Erfani, J. Seaberg, C.P. Aichele, J.D. Ramsey, Biomacromolecules 21 (2020) 2557–2573.
[12] D. Nagasawa, T. Azuma, H. Noguchi, K. Uosaki, M. Takai, J. Phys. Chem. C 119 (2015) 17193–17201.
[13] Z. Feng, X. Feng, X. Lu, Environ. Sci. Technol. 57 (2023) 7298–7308.
[14] C.-H. Lin, S.-C. Luo, Langmuir 37 (2021) 12476–12486.

[15] C. Freij-Larsson, T. Nylander, P. Jannasch, B. Wesslén, Biomaterials 17 (1996) 2199–2207.
[16] J. Zheng, W. Song, H. Huang, H. Chen, Colloids Surf. B Biointerfaces 77 (2010) 234–239.
[17] C. Yoshikawa, S. Hattori, C.-F. Huang, H. Kobayashi, M. Tanaka, J. Mater. Chem. B 9 (2021) 5794–5804.
[18] A.B. Asha, Y. Chen, R. Narain, Chem. Soc. Rev. 50 (2021) 11668–11683.
[19] L. Mi, S.Y. Jiang, Angew. Chem., Int. Ed. 53 (2014) 1746.
[20] X.-Y. Zhang, Y.-Q. Zhao, Y. Zhang, A. Wang, X. Ding, Y. Li, S. Duan, X. Ding, F.-J. Xu, Biomacromolecules 20 (2019) 4171–4179.
[21] W. Yang, H. Xue, W. Li, J. Zhang S. Jiang, Langmuir 25 (2009) 11911–11916.
[22] T. Masuda, M. Takai, J. Mater. Chem. B 10 (2022) 1473–1485.
[23] T. Wu, K. Efimenko, J. Genzer, J. Am. Chem. Soc. 124 (2002) 9394–9395.
[24] W. Feng, J.L. Brash, S. Zhu, Biomaterials 27 (2006) 847–855.
[25] C. Chu, Y. Higaki, C.H. Cheng, M.H. Cheng, C.W. Chang, J.T. Chen A Takahara, Polym. Chem. 8 (2017) 2309–2316.
[26] A.R. Kuzmyn, A.T. Nguyen, L.W. Teunissen, H. Zuilhof, J. Baggerman, Langmuir 36 (2020) 4439–4446.
[27] A. Noguchi, T. Masuda, C. Chen, S. Yoshizawa, N. Isu, M. Takai, Mater. Adv. 1 (2020) 2737–2744.
[28] M. Kobayashi, A. Takahara, Polym. Chem. 4 (2013) 4987–4992.
[29] L. Wang, G. Gao, Y. Zhou, T. Xu, J. Chen, R. Wang, R. Zhang, J. Fu, ACS Appl. Mater. Interfaces 11 (2019) 3506–3515.