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N-Halamine-modified polyglycolide (PGA) multifilament as a potential bactericidal surgical suture: *In vitro* study

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ABSTRACT: In this study, new cationic homopolymer and anionic copolymer were synthesized, and deposited onto polyglycolide sutures using a layer-by-layer assembly technique. The coated sutures were rendered antibacterial by chlorinating with dilute solution of household bleach solution at pH 7. The chlorination treatment transformed the N—H groups of anionic copolymer into *N*-halamine structures. The *N*-halamine-modified sutures were challenged with *Staphylococcus aureus* and *Escherichia coli* O157:H7 bacteria at different contact times. The suture with chlorine loading of 0.22% completely inactivated both bacterial strains in 30 min contact time. Fourier transform infrared spectroscopy, scanning electron microscopy, and analytical titration confirmed the successful deposition of the *N*-halamine multilayers. The effect of layer-by-layer coatings of polyelectrolytes on the chlorine loading and antibacterial efficacy of sutures was evaluated. The straight-pull and knot-pull strength tests performed on the sutures reported slight decline in tensile properties after chlorination treatment. The *in vitro* hemolysis and cytocompatibility tests revealed that the *N*-halamines-based antibacterial sutures were biocompatible. © 2015 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2015**, *132*, 42483.

KEYWORDS: biocompatibility; biodegradable; coatings

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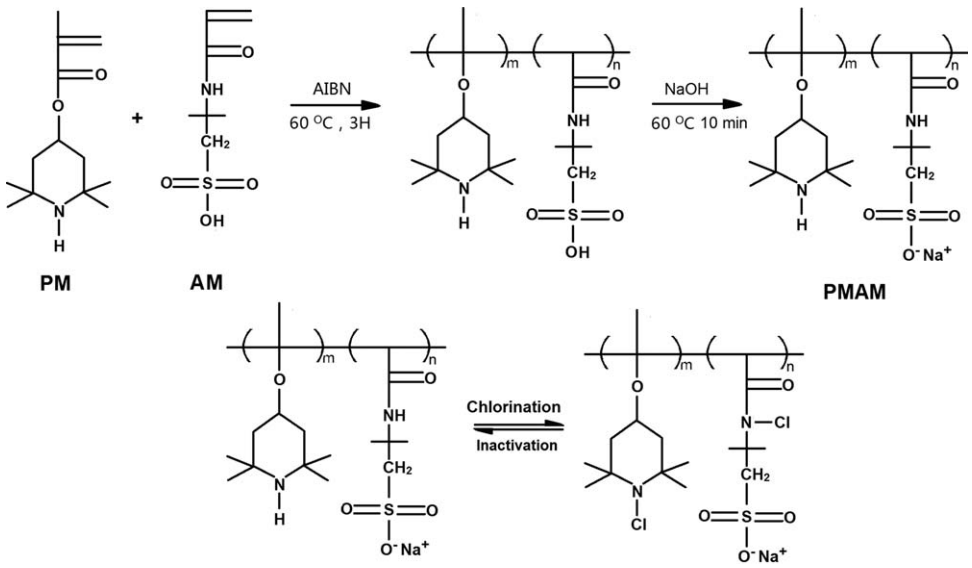
INTRODUCTION

The applications of polymeric antimicrobial in medical textiles have prominently increased in the past few years. One of the important applications is in the field of surgical sutures. The presence of suture at the site of incision can cause microbial adherence to its surface. Various microbes may contaminate not only the tissue in the surgical wound but also the actual suture materials.[1–4] The microorganisms associated with the wound infection include both Gram-positive and Gram-negative bacteria. Once suture materials become contaminated, antibacterial agents or other mechanisms of wound decontamination become ineffective owing to biofilm formation.[3] From the first decade of twenty-first cemtury, antibacterial sutures are developed by coating various antimicrobial agents and embedding antibiotics in polymer networks.[5,6] The studies reported the antimicrobial activity of silver nanoparticles,[7] triclosan,[8] chitosan,[9] antimicrobial peptides,[10] and grapefruit seed extract[11] immobilized on sutures. To control the surgical infections, Coated VICRYL Plus from Johnson & Johnson was the first commercially available antibacterial suture based on triclosan. Triclosan is a widely used antibacterial agent in cosmetics, household, and textile products. But the extensive use of triclosan has caused the bacteria to develop resistance against it.[12,13] Triclosan is also adversely affecting the human health as it bioaccumulates in human milk, fat tissues, and urine.[14] In addition, Li *et al.*[10] studied the antibacterial coating of suture based on triclosan and stated that triclosan coating functioned as bacteriostatic at low concentration. It was reported that suture coated with 2.4% triclosan provided a small log reduction of *Staphylococcus aureus* bacteria in 30 min contact time. Therefore, appropriate substitutes with better stability, biocompatibility, and biocidal activity are urgently required.

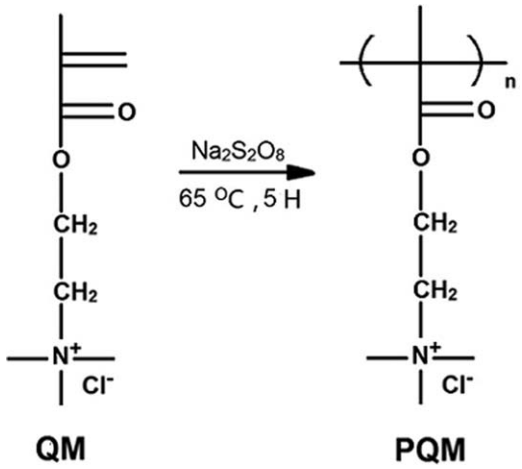
N-halamines have been proved as promising antimicrobial agents due to their stability and broad-spectrum biocidal activity.[15–17] *N*-halamines have demonstrated efficacy in killing of Gram-positive and Gram-negative microorganisms without causing the microorganisms to develop resistance to them.[18] The inactivation of bacteria occurs through direct transfer of oxidative halogen to the cell membrane. The biocidal activity depends on the stability of the halamine bonds, which is



Scheme 1. Synthesis and function of copolymer (PMAM).

directly related to the *N*-halamine structure. The rates of inactivating bacteria follow the order imide > amide > amine halamines, which is the reverse of the order of their stabilities.[19] In recent studies by Cerkez *et al.*, *N*-halamines polyelectrolytes were synthesized and deposited on cotton to create bactericidal surfaces.[20–22] The *N*-halamines can become a desirable substitute in on-going research on antibacterial coating of suture.

Herein, for the first time, polyglycolide (PGA) sutures were antibacterially functionalized by *N*-halamines polyelectrolytes. We synthesized water-soluble polycation and a new polyanion with amide groups (Schemes 1 and 2). The polyelectrolytes were immobilized on PGA suture via layer-by-layer (LbL) technique without any initial surface modification (Figure 1). The LbL method was found to be a simple and effective technique to create antibacterial coating onto the suture. The biocidal function of *N*-halamines using low solution concentration and fairly good biocompatibility are the key findings of this research. The straight-pull and knot-pull strength tests were performed to study the variation in tensile properties of the coated suture before and after chlorination. The antibacterial efficacies of chlorinated suture were evaluated against *S. aureus* and *Escherichia coli* O157:H7 bacteria in different contact times. To find shelf life of the coated suture, storage stability test was performed.



Scheme 2. Synthesis of homopolymer (PQM).

EXPERIMENTAL

Materials

The braided and sterilized PGA sutures were purchased from Jinhuan Medical Products Co, Ltd, China. 2,2,6,6-tetramethyl-4-piperidyl methacrylate (TM), 2-acrylamido-2-methyl-1-propanesulfonic acid (AM), and trimethyl-2-methacryloxyethylammonium chloride (QM) were purchased from J&K Chemicals, Shanghai, China. Other chemicals were purchased from Sinopharm Chemical Reagent Co., Ltd., Shanghai, China.

Instruments

The FTIR spectra of the treated PGA sutures were recorded by Thermo Scientific Nicolet iS10 spectrometer in the optical range of 400–4000 cm^{-1}. The ^1H NMR spectra of polymers were recorded on a Broker AV-300 spectrometer. Surface morphologies of uncoated and coated suture were analyzed by SU-1510 (Hitachi, Tokyo, Japan) field-emission scanning electron microscopy (SEM) and CSPM-5000 (BenYuan Co. China) AFM in tapping mode equipped with silicon tip. The DCAT 21 (Dataphysics) dynamic contact angle measuring instrument was used to analyze the surface hydrophobicity of sutures. The elemental analysis was made by vario EL III elemental analyzer. The gel permeation chromatography (GPC) analysis of synthesized polymers was performed on a Waters 1515 instrument using THF at room temperature.

Preparation of Anionic Polyelectrolyte

An equimolar (20 mmol) mixture of 2,2,6,6-tetramethyl-4-piperidyl methacrylate (PM) and 2-acrylamido-2-methyl-1-propanesulfonic acid (AM) was dissolved in 50 mL of ethanol (EtOH), and then 1 wt % azobis (isobutyronitrile) (AIBN) was added into the solution, which was stirred until all the ingredients were fully dissolved (Scheme 1). Nitrogen gas was

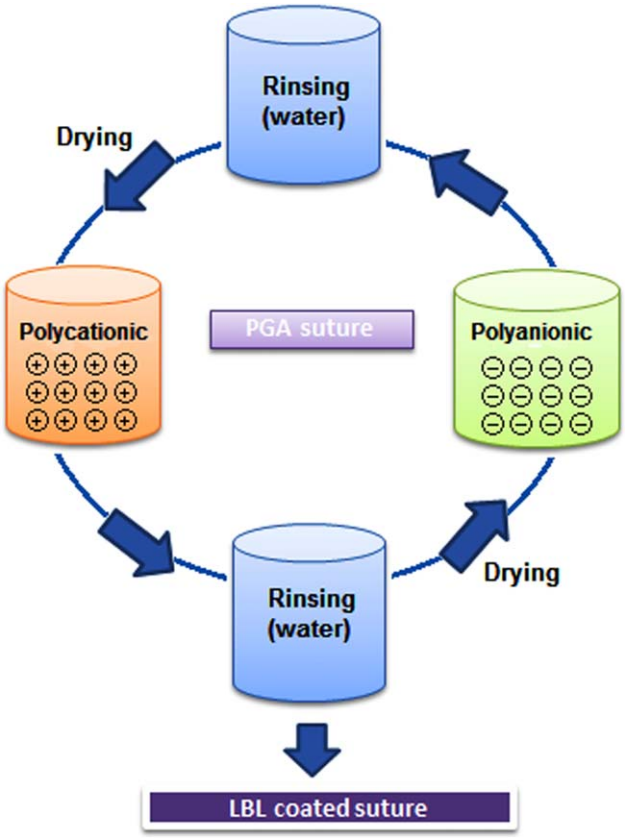


Figure 1. LbL assembly technique adapted for coating sutures. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

bubbled into the solution for first 15 min. The polymerization reaction was carried out under a nitrogen atmosphere at 60°C for 3 h. Once the polymerization was completed, 20 mmol of NaOH was added into the copolymer solution, and the mixture was refluxed for 10 min in order to produce the sodium salt of the 2,2,6,6-tetramethyl-4-piperidyl methacrylate-co-2-acrylamido-2-methyl-1-propanesulfonic acid (PMAM). The reaction vessel was cooled to room temperature, and EtOH was removed by evaporation. ^{1}H NMR (400 MHz, DMSO-d6, δ): 1.21(18H), 1.53(7H), 2.10(4H), 2.55(1H), 3.39(1.91), 5.13(1H).

Preparation of Cationic Polyelectrolyte

Trimethyl-2-methacryloxyethylammonium chloride (QM) (50 mmol) and sodium persulfate (0.40 g) were added in a three-necked round-bottomed flask equipped with a gas inlet and reflux condenser containing 150 mL water (Scheme 2). The solution was stirred and nitrogen gas was bubbled for 30 min before starting the reaction, and continued for 5 h at 65°C. After the completion of polymerization reaction, water was removed by evaporation to get the homopolymer, poly-trimethyl-2-methacryloxyethylammonium chloride (PQM). ^{1}H NMR (400 MHz, DMSO-d6, δ): 1.10–1.22(5H), 3.31(9H), 3.80(2H), 4.54(2H).

Layer-By-Layer Coating of Sutures

The dilute solutions of anionic and cationic polyelectrolytes (2% w/v) were prepared in water in different glass containers, and the sutures were immersed alternatively in the solutions starting with cationic polymer. The immersion time was 1 min for each dip but the first dip was for 5 min to ensure the reasonable attraction for subsequently anionic polymer layer. Between each immersion, the sample was rinsed and dried at 100°C for 10 min to remove loosely attached polyelectrolytes on the surface of sutures. This procedure was repeated until the desired number of layers was reached.

Chlorination and Titration

Coated samples were chlorinated with 10% aqueous solution of household bleach at pH 7 for 1 h and then rinsed with distilled water. The samples were dried at 45°C for 1 h to remove any free chlorine from the surface. The chlorine loading (Cl^+%) of the sutures was determined by iodometric/thiosulfate titration, and was calculated according to equation:

$$Cl^+(\%) = \frac{N \times V \times 35.45}{W \times 2} \times 100$$

Where Cl^+% is the wt % of the oxidative chlorine on the samples, N and V are the normality (equiv/L) and volume (L) of $Na_2S_2O_3$ (titrant), respectively, and W is the weight of the suture sample in grams.

Antibacterial Efficacy Test

The antimicrobial efficacy of coated sutures was evaluated by modification of test method reported elsewhere.[23] The suspensions of *S. aureus* (ATCC 6538) and *E. coli* (ATCC 43895) were prepared in sterile phosphate buffer saline (PBS) with concentration of 10^6 (cfu/mL). Each sample of 1 g was challenged for 15, 30, and 60 min in Eppendorf tubes containing 200 μL bacterial suspensions ($\sim 10^6$ CFU/mL). After the challenge, sample was quenched with 5.0 mL solution of sodium thiosulfate (0.02 N) to neutralize the loosely attached oxidative chlorine on the surface of suture. Serial dilutions of this quenched solution were prepared and plated on Trypticase agar plates. After incubation for 24 h at 37°C, the resultant viable colonies were counted and bactericidal activity was determined.

In Vitro Hemolysis Test

The hemolytic activity of the coated sutures was determined using protocol as previously reported with few modifications.[24] Briefly, erythrocytes (red blood cells) were separated from fresh human blood sample and rinsed three times with 0.9% NaCl. The samples weighing 1 g were incubated at 37°C for 60 min with the erythrocyte (5% v/v) suspension in PBS (35 mM PBS and 0.15 M NaCl at pH 7.4). The erythrocytes were removed by centrifugation for 10 min and hemolysis was assessed by measuring the optical density at 540 nm of the supernatant. For negative and positive controls, erythrocytes in PBS (A_{blank}) and in 0.1% Triton X-100 (A_{triton}) were used. The test was repeated three times for each sample and the mean values were recorded. The percentage of hemolysis was calculated according to the equation:

$$Hemolysis = \left[\left(A_{sample} - A_{blank}\right)/\left(A_{triton} - A_{blank}\right)\right] \times 100\%$$

In Vitro Cytocompatibility Test

The cytocompatibility of *N*-halamine-modified suture was evaluated by using NIH 3T3 mouse fibroblasts. Briefly, 3T3 cells were cultured in Dulbecco's Modified Eagle Medium (DEME) with

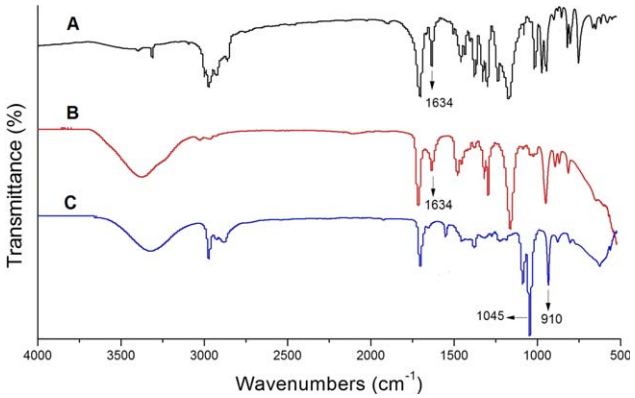


Figure 2. FTIR spectra of (A) trimethyl-2-methacryloxyethylammonium chloride, (B) 2-acrylamido-2-methyl-1-propanesulfonic acid, and (C) PMAM. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

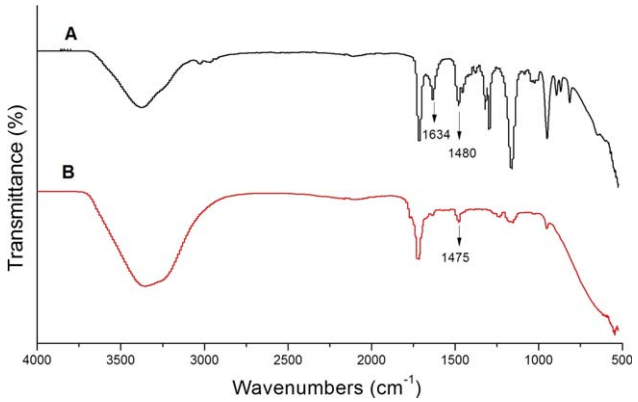


Figure 3. FTIR spectra of (A) trimethyl-2-methacryloxyethylammonium chloride and (B) PQM. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

10% fetal bovine serum (FBS) and 1% Penicillin Streptomycin (Pen Strep) at 37°C with 95% humidity and 5% CO_2. Ten thousand cells in 200 µL DMEM were seeded into each well of a 98-well plate. After cells adhered to the plate, a piece of suture fiber with 1 cm length was placed in each well, followed by 24 h incubation. After 24 h, a Cell Counting Kit-8 (CCK8) assay was performed to determine the cell viability. The tissue culture plate itself was served as the control. The cell viability was reported as the percentage to the control ($n = 6$ for all samples).

Tensile Strength Test

The tensile properties were measured as per ASTM D2256 test method using universal testing system (YG-B026G, China). The distance between the jaws was 8 cm, and the gauge speed was 10 mm/min. The straight-pull and knot-pull strength tests were performed. In straight-pull test, the suture was clamped in between the fixed jaw and moving jaw of the tester and pulled until they broke. However, for the knot-pull test, the suture was tied into a square knot by a single throw around a 6-mm-diameter rod. The rod was removed and the ends of the suture were positioned around the grip mandrels such that the knot was at the center of the gauge area. The tests were repeated three times for each sample and mean values were recorded.

Storage Stability Test

The storage stability of chlorinated sutures at room temperature was evaluated. The sutures placed in airtight plastic bags were stored in dark place. The chlorine contents were tested periodically over 10 weeks storage period. The sutures samples were titrated to assess the stability of active chlorine to storage.

RESULTS AND DISCUSSIONS

Characterization of Polymers

The Fourier transform infrared spectroscopy (FTIR) spectra of trimethyl-2-methacryloxyethylammonium chloride (TM), 2-acrylamido-2-methyl-1-propanesulfonic acid (AM), and 2,2,6,6-tetramethyl-4-piperidyl methacrylate-co-2-acrylamido-2-methyl-1-propanesulfonic acid sodium salt (PMAM) are shown in Figure 2. The characteristic band recorded in TM and AM monomers at 1634 cm^{-1} due to C═C double bond vibrational mode was disappeared after copolymerization in PMAM spectrum.[22] Also, bands of the stretching of S—O at 910 cm^{-1} and the symmetric S═O stretching at 1045 cm^{-1} are observed and indicated the presence of sulphonate groups of monomer AM in the structure of PMAM copolymer.[25]

In Figure 3, spectra of trimethyl-2-methacryloxyethylammonium chloride (QM) and poly-trimethyl-2-methacryloxyethylammonium chloride (PQM) are shown. The vinyl bond vibrational stretching band (1634 cm^{-1}) of the monomer (QM) disappeared in the spectrum of homopolymer PQM. The C—H asymmetric deformation of the *N*-trimethyl groups of the monomer was observed at 1475 cm^{-1} in the spectrum of PQM.[26] The characteristics bands recorded in the FTIR spectra suggested successful formation of desired products.

The molecular weight (M_n) and molecular weight distribution (M_w/M_n) of synthesized polymers were measured by GPC analysis. GPC recorded the values of copolymer PMAM ($M_n = 15{,}100$, $M_w/M_n = 1.70$) and of homopolymer PQM ($M_n = 9300$, $M_w/M_n = 1.41$). The composition of copolymer PMAM was investigated by CHNS elemental analysis. The calculated result was determined as C, 55.57%; H, 8.33%; N, 6.42%; S, 7.4%, and with the observed result as C, 55.45%; H, 8.24%; N, 6.31%; S, 6.88%.

Characterization of LbL Coating

The formation of the coatings on sutures was characterized by FTIR spectroscopy (Figure 4). The bands at 1725 and 1649 cm^{-1} corresponded to ester carbonyl stretching band modes and N—H in-plane vibrational bending of the acyclic amide group, respectively.[26,27] The asymmetric and symmetric bands of SO_2 were observed at 1189 and 1043 cm^{-1}, respectively, indicated the presence of top layer of copolymer PMAM on suture surface.[25,28] The bands observed in spectra of coated sutures at 1721 and 3410 cm^{-1} were ester carbonyl and N—H vibrational stretching modes of the copolymers, respectively. The FTIR data indicated the successful coating of *N*-halamines polyelectrolytes.

Surface Morphology of Coated Suture

The SEM images of sutures were recorded with 200× magnification [Figure 5(A,B)]. Clearly, the image showed uneven and

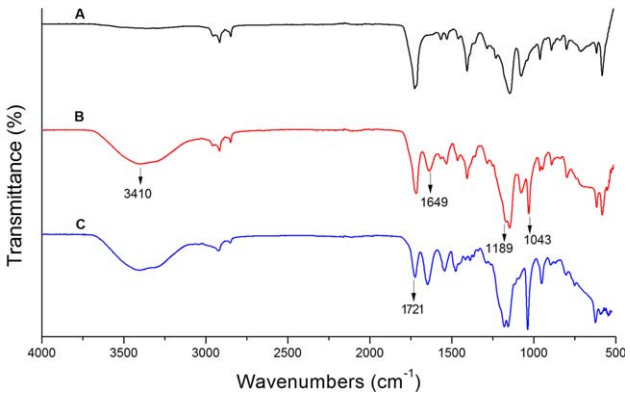


Figure 4. FTIR spectra of (A) untreated suture, (B) LbL coated suture, and (C) chlorinated suture. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

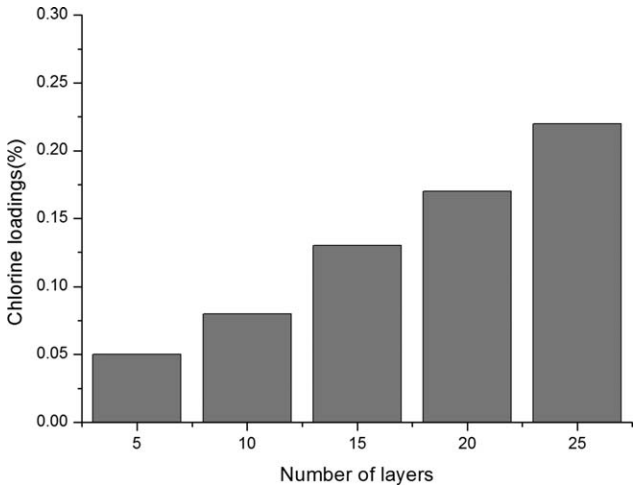


Figure 6. Effect on chlorine loadings (Cl^+%) of sutures by coated layers.

dense coating of polyelectrolytes on treated suture as compared to untreated suture.

The surface topography was conducted on PGA suture by AFM microscopy in order to point out the morphological change obtained after the layer-by-layer treatment [Figure 5(C,D)]. AFM analysis showed the average roughness of 21.7 nm of untreated suture, whereas increment of 64.5 nm was observed in coated suture. There was a simultaneous gain in surface roughness of coated suture with increase in number of deposited layers. The SEM and AFM analysis demonstrated successful coating of *N*-halamine polyelectrolytes on suture.

Chlorine Loadings of Sutures

The LBL deposition of polyelectrolytes was further affirmed by measuring chlorine content of the chlorinated sutures through analytical titration. The chlorination of coated sutures with aqueous solution of sodium hypochlorite transformed the N—H groups of polyanion (PMAM) into N—Cl bonds. A linear increase in chlorine loadings (Cl^+%) was recorded with increase in number of deposited layers on the sutures (Figure 6). It was also observed that the release of chlorine from bottom layers of the coating was retarded as the number of layers on suture was increased.

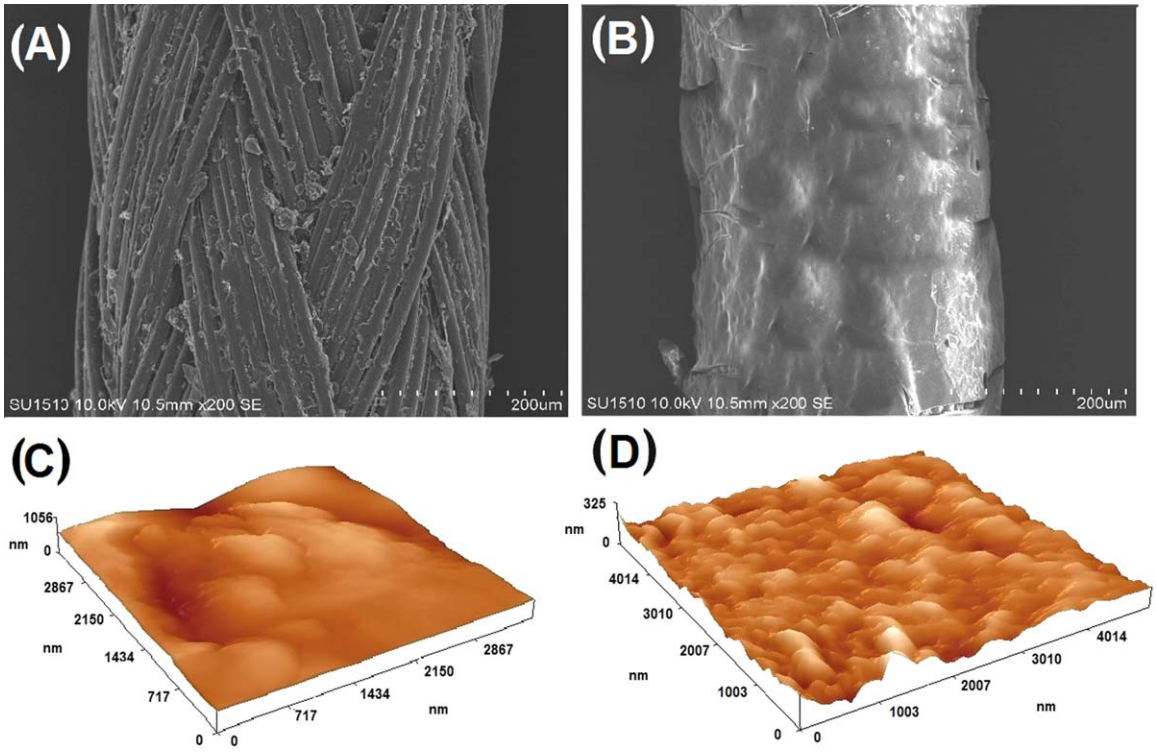


Figure 5. SEM images of (A) uncoated and (B) coated suture at 200× magnification. 3D AFM images of (C) uncoated and (D) coated suture. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table I. Antibacterial Efficacy of the Sutures

Samples	Contact time (min)	Bacterial reduction (log)	
		S. aureus[a]	*E. coli*[a]
LBL (control)	30	0.02	0.03
LBL (chlorinated)	15	1.48	1.79
	30	5.86	5.95
	60	5.86	5.95

LBL: layer-by-layer
[a]The inoculum concentrations were 5.86 logs.
The inoculum concentrations were 5.95 logs.

trol sample could be due to the adhesion of the bacteria on the surface. All the chlorinated samples showed bactericidal activity against *S. aureus* and *E. coli* O157:H7. The sample with 25 layers killed all the bacteria within 30 min contact time as seen in Figure 7. The chlorinated sutures successfully killed both bacterial strains; coated with low concentration solution (2% w/v) of *N*-halamine polyelectrolytes. It was also deduced that increase in contact time for inactivation of bacteria may be caused by slow chlorine diffusion from the bottom layers of coated suture; similar phenomenon was observed during analytic titration. The experimental results also provided tangible evidence in support of the hypothesis that the active chlorine in *N*-halamines structure (N—Cl) is a source of bactericidal activity as confirmed in previous studies.[21,22]

Antibacterial Efficacy Testing

The quantitative determination of antibacterial activity of *N*-halamine-functionalized sutures was achieved through a modified test method. The sutures were challenged for different contact times, by direct contact with suspension of *S. aureus* (5.86 logs) and *E. coli* O157:H7 (5.95 logs). The antibacterial efficacy results of unchlorinated (control) and chlorinated suture (0.22% Cl^+) are displayed in Table I. The control sample did not exhibit significant reduction against *S. aureus* and *E. coli* O157:H7 as expected. The small reduction experienced by con-

Hemolytic Activity Test

The *in vitro* hemolytic activity of the sutures was investigated using fresh human erythrocytes. As seen in Figure 8, the untreated suture provided no hemolytic toxicity. The unchlorinated and chlorinated sutures showed 12.1 and 16.8% hemolysis activity, respectively. According to several studies, *in vitro* percent hemolysis is rated as "no concern" when it varies from 5 to 25%.[29,30] Therefore, below 25% hemolytic activity makes *N*-halamine-modified sutures nontoxic and compatible with blood. The observed increase in the hemolysis activity of

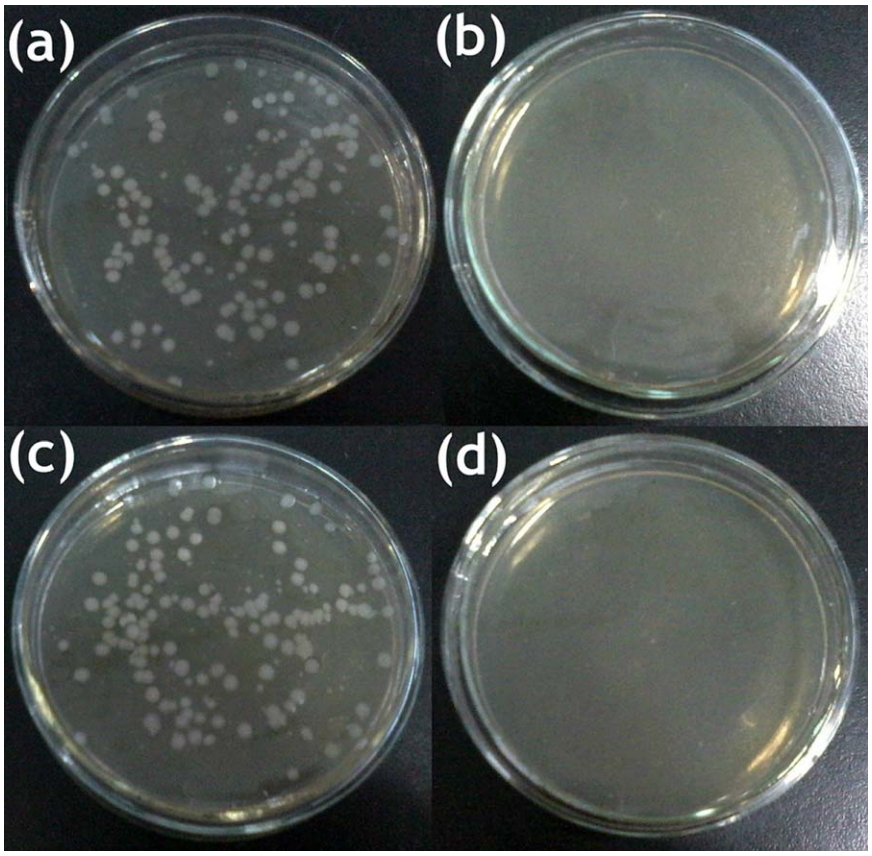


Figure 7. Plates with *E. coli* colonies for (a) unchlorinated and (b) chlorinated suture, and *S. aureus* colonies for (c) unchlorinated and (d) chlorinated suture. The challenge time was 30 min. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

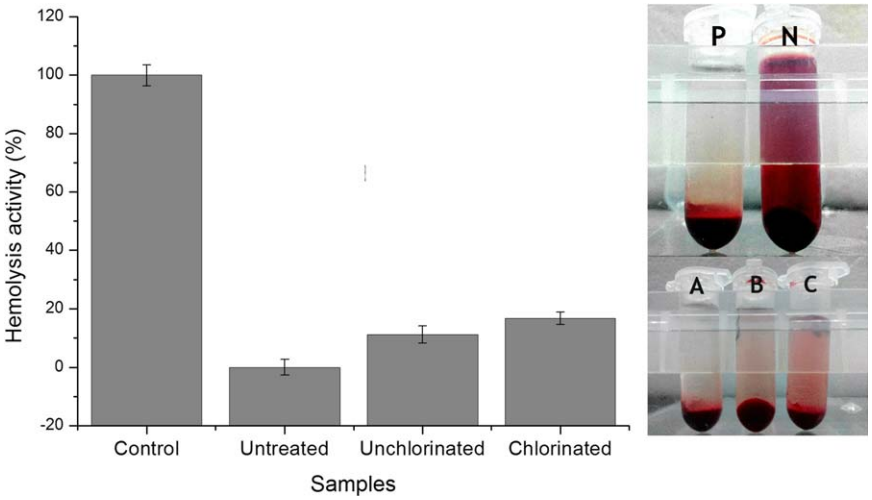


Figure 8. Hemolysis activity (%) of treated sutures; whereas (P) positive control, (N) negative control, (A) untreated suture, (B) unchlorinated suture, and (C) chlorinated suture. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

chlorinated suture is caused by enhanced hydrophobicity due to the formation of nonpolar N—Cl covalent bond.[31] The results support the notion that greater hydrophobicity is prone to inducing hemolysis against human erythrocytes.[32] The chlorinated suture with high chlorine loadings can lead to hemolytic toxicity.

To further investigate the effect of chlorination on surface hydrophobicity of coated suture, the water contact angle was measured. The contact angles of unchlorinated and chlorinated samples were $124 \pm 1°$ and $129 \pm 1°$, respectively. The top layer of anionic copolymer on coated suture containing amide and sodium sulphonate groups made the surface hydrophilic. After chlorination, N—H bonds were transformed into N—Cl bonds (*N*-halamine structures) causing a small increase in contact angle of chlorinated sutures.[33] The decrease in hydrophilic groups on the coated suture due to chlorination resulted in surface hydrophobicity. Therefore, chlorinated sutures showed higher level of hemolytic toxicity than unchlorinated sutures (Figure 8).

In Vitro Cytocompatibility Test

The sutures were incubated within the media of mouse 3T3 fibroblasts for 24 h to determine the *in vitro* cytocompatibility. Figure 9 showed 100% cell viability of untreated suture. The LbL coated suture exhibited toxicity with about 82% showing significant difference to control ($p < 0.05$). After chlorination, the suture with chlorine loading of 0.22 wt % showed some toxicity of about 79% cell viability, which was significantly lower than the control ($p < 0.05$). The result is consistent with the former findings that chlorinated materials caused significant decrease in cell viability.[19,34] However, it was also observed in the studies that the consumption of oxidative chlorine with the time promoted the cell growth.

Tensile Strength Test

The tensile strength and knot security of sutures are very important in surgery to provide adequate tension for wound closure. The effect of LbL and chlorination treatment was studied on the inherent tensile strength of sutures. The results of

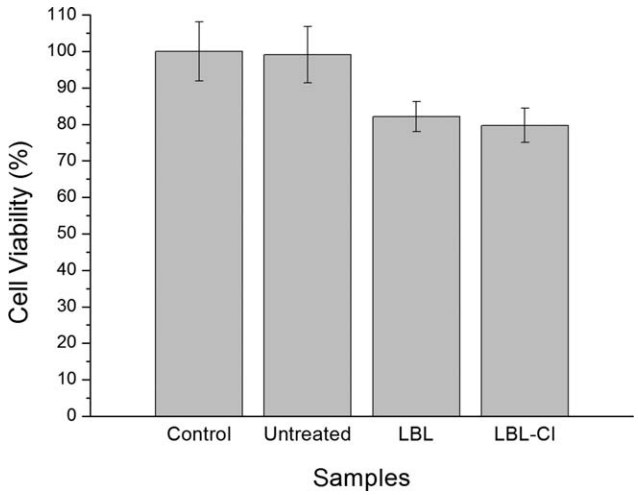


Figure 9. 3T3 mouse fibroblast viability after incubated with different sutures for 24 h. Blank tissue culture plate was the control. (*, $p < 0.05$).

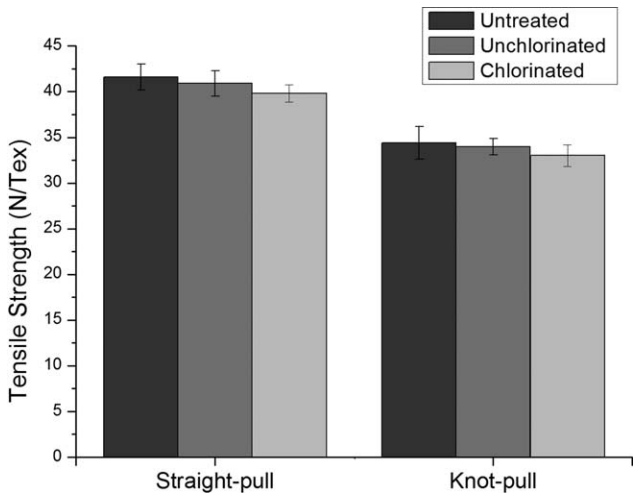


Figure 10. Effect of chlorination treatment on tensile properties of suture.

Table II. Storage Stability of Chlorinated Suture

Storage Time (weeks)	Cl^+ % wt
0	0.22
2	0.21
6	0.19
10	0.18
10 (rechlorination)	0.22

straight-pull and knot-pull strengths are graphically presented in Figure 10. The slight decrease in strength seen after chlorination was due to the oxidation of polyglycolide chains by sodium hypochlorite. The results showed that LbL coating technique has an advantage over the traditional coating process to produce *N*-halamine-based antimicrobial sutures. This technique is useful for coating medical devices especially when chemically inert surfaces are involved.

Storage Stability Test

To estimate the stability of *N*-halamines attached to suture surface, the storage test was performed at room temperature. The storage stability of the chlorinated sutures is periodically shown in Table II. After 10 weeks of storage, the samples retained 81% of the initial chlorine loading, which is still effective in inactivating bacteria. The chlorine lost was restored by rechlorination, showing that the chlorine losses were due to dissociation of N—Cl bonds. The results demonstrated that the N—Cl bonds loaded on sutures are very stable at room temperature.

CONCLUSIONS

Anionic and cationic polymers were synthesized and successfully coated on sutures via LbL assembly technique. The *N*-halamine coating demonstrated to be bactericidal at low solution concentration; whereas the widely used antimicrobial agent triclosan is only bacteriostatic. The *N*-halamine-loaded sutures inactivated *S. aureus* and *E. coli* O157:H7 within 30 min of contact time. The sutures with 0.22% chlorine loading showed 16.8% hemolytic activity and 79% cell viability. The *in vivo* studies for cytocompatibility and hemolysis activity will be needed to determine the availability of the antimicrobial *N*-halamine-functionalized suture in the future. The storage stability test indicated that the 0.18% wt active chlorine remained on the chlorinated after 10 weeks of storage. It was inferred that LBL technique is favorable to coat sutures as it modifies the surface without altering the polymer network of suture material. The biocompatible, biocidal, and bacteria-resistant *N*-halamine coatings can become a suitable substitute for developing antibacterial sutures.

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