Published by :
https://www.ijert.org/
An International Peer-Reviewed Journal

International Journal of Engineering Research & Technology (IJERT)
ISSN: 2278-0181
Vol. 15 Issue 03 , March - 2026

# Preparation and Antibacterial Application of pH-Responsive dB-PEG-PVA Hydrogel Dressin

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***Abstract*—Chronic wounds fail to heal in a timely manner due to disruption of the normal healing process, among which bacterial biofilm formation is a major barrier. Inhibiting biofilms while promoting wound repair has important clinical significance. Traditional antibacterial dressings often suffer from uncontrolled antibiotic release, and excessive antibiotic use can lead to drug resistance and additional health risks. Polyvinyl alcohol (PVA)-based hydrogels have attracted considerable attention because of their good biocompatibility and ability to maintain a moist wound environment; however, pure PVA hydrogels show limited bioactivity and insufficient functionality for biomedical applications. To overcome these limitations, a chemically/physically double-crosslinked hydrogel was constructed using PVA as the matrix and phenylboronic acid-terminated polyethylene glycol (PEG) as a functional component. Vancomycin hydrochloride (Van) was further loaded to obtain a pH-responsive antibacterial hydrogel dressing. The introduction of PEG regulated the crosslinking density and improved swelling, water absorption, and retention properties, while dynamic boronated ester bonds between phenylboronic acid and PVA hydroxyl groups endowed the hydrogel with pH responsiveness. In the weakly acidic microenvironment of bacterial biofilms, the hydrogel enabled controlled drug release and enhanced antibacterial efficacy. The prepared hydrogel exhibited excellent swelling, adhesion, and moisture-retention ability, as well as significant antibacterial activity against Staphylococcus aureus, indicating its potential as a promising wound dressing for chronic wound treatment.**

***Keywords*—pH-responsive hydrogel, Polyvinyl alcohol (PVA), Phenylboronic acid, Controlled drug release, Antibacterial wound dressing**

## I. INTRODUCTION

Chronic wound infection has become a major clinical challenge due to persistent inflammatory response, bacterial colonization, and biofilm formation, which severely interfere with the normal healing process [1,2]. Bacterial biofilms provide a protective microenvironment for pathogens, significantly enhancing antibiotic resistance and reducing the therapeutic efficacy of conventional antibiotics, thereby making chronic wounds difficult to heal [3-5]. Therefore, the development of functional dressings with controllable antibacterial activity and responsiveness to the infection microenvironment is of great significance for improving the treatment of chronic wounds.

Hydrogels have attracted extensive attention in wound healing because of their three-dimensional network structure, high water content, and excellent biocompatibility, which can provide a moist environment for wounds and promote tissue regeneration [6-8]. Among them, polyvinyl alcohol (PVA) hydrogels are widely used in biomedical applications due to their low toxicity, good film-forming ability, and tunable mechanical properties [9]. However, pure PVA hydrogels often suffer from insufficient mechanical strength, unstable swelling behavior, and limited biological activity, which restrict their further biomedical applications [10]. Therefore, introducing functional polymers and constructing double-crosslinked network structures has been considered an effective strategy to improve the stability and functionality of PVA hydrogels.

Polyethylene glycol (PEG) is a hydrophilic and biocompatible polymer that can regulate the crosslinking density of hydrogels and improve their swelling behavior, water retention, and structural stability [11]. In addition, phenylboronic acid groups can form reversible boronated ester bonds with polymers containing cis-diol structures, thereby endowing the material with pH-responsive properties [12]. Since bacterial biofilms usually exhibit a weakly acidic microenvironment, the introduction of boronated ester bonds into hydrogel systems enables infection-responsive drug release [13]. Loading antibiotics, such as vancomycin, into pH-responsive hydrogels can increase the local drug concentration while reducing systemic side effects and minimizing the risk of bacterial resistance.

Based on the above considerations, in this study, phenylboronic acid-functionalized PEG was introduced into the PVA network to

International Journal of Engineering Research & Technology (IJERT)
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Vol. 15 Issue 03 , March - 2026

construct a pH-responsive PVA/PEG double-network hydrogel through physical–chemical synergistic crosslinking, followed by loading vancomycin as an antibacterial agent. The dynamic boronated ester bonds endowed the hydrogel with infection-responsive drug release capability, while the double-crosslinked network significantly improved the mechanical properties, swelling performance, and stability of the hydrogel. The prepared hydrogel exhibited good water absorption, adhesion, and significant antibacterial activity against Staphylococcus aureus, indicating its potential as an antibacterial dressing for chronic wound treatment.

## II. EXPERIMENTAL

### A. Preparation and characterization of phenylboronic acid-terminated PEG (dB-PEG)

HO-$PEG_{6K}$-OH (10.2 g, 1.7 mmol) was first placed in an oil bath at 110℃ and heated under vacuum for 1 h until completely melted and dissolved. The temperature was then reduced to 60℃ under nitrogen protection, followed by the addition of tetrahydrofuran (10–20 mL), isochrone diisocyanate (0.8 g, 3.6 mmol), and a drop of dibutyltin dilaurate as catalyst. The reaction mixture was stirred at a constant speed for 4 h. Subsequently, 3-aminophenylboronic acid (0.5 g, 3.6 mmol) was added and the reaction was continued for another 8 h. After completion, excess tetrahydrofuran was removed by rotary evaporation to obtain the crude product, which was further purified by precipitation in excess cold ether. The obtained product was dried in a vacuum oven at 40℃ to yield phenylboronic acid-terminated polyethylene glycol (dB-PEG), and its structure was confirmed by $^1H$ NMR spectroscopy.

### B. Synthesis of dB-PEG-PVA and PEG-PVA hydrogels

PVA was dissolved in deionized water at 90℃ and stirred in a 90℃ water bath for 4 h to obtain a 15 wt% PVA solution. dB-PEG was dissolved in deionized water using a 60℃ oil bath to prepare a 3 wt% dB-PEG solution. The dB-PEG and PVA solutions were then mixed at a 3 wt% dB-PEG : 15 wt% PVA volume ratio of 1:1, 1:2, 1:3, and 1:4 to obtain 12 mL of the mixture. After thorough stirring, the mixed solution was poured into polytetrafluoroethylene (PTFE) molds and subjected to a freeze–thaw process to form hydrogels, which were labeled as dB-PEG-PVA1, dB-PEG-PVA2, dB-PEG-PVA3, and dB-PEG-PVA4, respectively. As a blank control, 12 mL of 15 wt% PVA solution was similarly processed via the freeze–thaw method and labeled as dB-PEG-PVA0. For drug-loaded hydrogels, vancomycin (Van, 1.2 mg, 100 μg/mL in 3 wt% dB-PEG) was added according to the total volume, and the hydrogels were prepared following the same procedure.

### C. Characterization of PEG-PVA and dB-PEG-PVA Hydrogels

The crystalline structure and crystallinity of the lyophilized PEG-PVA hydrogel samples were characterized using X-ray diffraction (XRD). For the measurement, the freeze-dried samples were evenly placed at the center of the sample holder and fixed on the diffractometer stage. The scanning range was set from 10° to 80° with a scan rate of 4°/min, and each measurement took approximately 17 minutes. After testing, the sample holder was removed, cleaned with ethanol, dried, and returned to its position. The positions and intensities of the diffraction peaks were recorded.

To confirm the synthesis of dB-PEG-PVA hydrogels and investigate the interactions among chemical groups, Fourier transform infrared (FTIR) spectroscopy was performed using the KBr pellet method. PVA, TAPBA, PEG, IPDI, and PEG-PVA or dB-PEG-PVA hydrogels with different ratios were dried, ground into a fine powder with a mortar, and pressed into thin pellets. FTIR spectra were collected over the range of 4000-400 $cm^{-1}$ to analyze their chemical structures.

### D. Mechanical Properties and Physical Characterization of Hydrogels

Rectangular hydrogel samples (20×13×4 $mm^3$) were prepared in clean PTFE molds using the freeze–thaw method. The initial distance between the upper and lower grips was measured with a vernier caliper and recorded as the gauge length $L_0$, while the sample width and thickness were measured to calculate the cross-sectional area. Tensile tests were performed using a universal testing machine at a stretch rate of 5 mm per minute. The tensile strength and elongation at break were determined from the maximum point on the stress–strain curve.

To evaluate the surface wettability of the hydrogels, samples were first equilibrated in PBS solution until fully swollen, gently wiped dry, and placed flat on the sample stage. A drop of deionized water was applied onto the hydrogel surface using a micro-syringe. After the droplet stabilized, images were captured using a high-speed camera. Each sample was tested in triplicate, and the average contact angle was calculated.

The water content of the hydrogels was measured by soaking samples in PBS (pH 7.4) for 24 hours until swelling equilibrium was reached. Excess surface water was gently blotted with a cotton cloth, and the wet mass was recorded as $m_b$. Samples were then lyophilized to constant weight, and the dry mass was measured as $m_a$. The water content was calculated to assess the hydrogel's water absorption capacity and application potential.

$$W\% = \frac{m_a - m_b}{m_a} \times 100\%$$

To study the physical and chemical properties of dB-PEG-PVA hydrogels and optimize their structure and functionality, the swelling

International Journal of Engineering Research & Technology (IJERT)
ISSN: 2278-0181
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ratio was measured gravimetrically. Each sample′ s initial mass was accurately weighed and recorded as $W_0$, and then immersed in PBS solutions (0.02 M, pH 5.5 and 7.4). Samples were weighed at time points of 1, 2, 4, 6, 8, 10, 12, 24, and 48 hours until swelling equilibrium was reached, recording the mass at each time as $W_t$. Each measurement was repeated at least three times, and the average value was reported. The swelling ratio was calculated according to the standard formula.

$$SR\,(\%) = \frac{W_t - W_o}{W_o} \times 100\%$$

E. Drug Loading and Release Studies

To determine the Vancomycin (Van) content in Van@dB-PEG-PVA1, a UV-visible spectrophotometer was used. A Van standard curve was established over a concentration range of 1-25 μg/mL. The drug loading of Van@PEG-PVA1 and Van@dB-PEG-PVA1 was then calculated by measuring the absorbance of the solution. To evaluate the drug release behavior of Van@dB-PEG-PVA1 hydrogel under different conditions, the drug-loaded dB-PEG-PVA1 and PEG-PVA1 hydrogels were first immersed in PBS (pH 7.4) for 12 hours until swelling equilibrium was reached. Subsequently, individual hydrogels were transferred into 50 mL centrifuge tubes, and six samples each of Van@dB-PEG-PVA1 and Van@PEG-PVA1 with equal mass were prepared. Three samples from each group were then placed in 5 mL of PBS at pH 7.4 and pH 5.5, respectively, and incubated in a 37°C thermostatic shaker. At predetermined time points of 1, 2, 3, 4, 5, 6, 12, 24, 36, and 72 hours, the release medium was collected and replaced with fresh PBS. The absorbance of the collected medium was measured to calculate the amount of Van released at each time point. Finally, the drug concentration remaining in the hydrogels was determined to calculate the cumulative release.

F. *In Vitro* Antibacterial Assay

Bacterial biofilm formation significantly enhances antibiotic resistance, thereby increasing the difficulty of clinical treatment. In vitro antibacterial tests of hydrogels are commonly used to evaluate their ability to inhibit or kill specific bacteria. To assess the antibacterial performance of Van@dB-PEG-PVA1 hydrogel under mildly acidic conditions, methicillin-resistant Staphylococcus aureus (MRSA) was selected as the test strain. The antibacterial activity was evaluated using the plate coating method and the standard plate count method. Prepared hydrogel samples were placed in 24-well cell culture plates and surface-sterilized under UV light for 30 minutes. Under aseptic conditions in a biosafety cabinet, 1 mL of MRSA suspension was added to each well, ensuring complete coverage of the hydrogel surface. The plates were then incubated at 37°C for 18 hours to allow bacterial adhesion and biofilm formation. The antibacterial effect was assessed using the standard plate count method. Sterile TSA culture medium was prepared in advance, cooled to 45–50°C, and poured into sterile Petri dishes to form plates. The cultured bacterial suspension was subjected to $10^{-4}$ serial dilutions (100 μL of bacterial suspension + 900 μL PBS, repeated four times), and 100 μL of each dilution was evenly spread onto TSA plates. The plates were incubated upside down at 37°C for 18 hours, after which colony-forming units (CFUs) were counted to evaluate the inhibitory effect of the hydrogel on *S.aureus*. Three parallel samples were included in each group to ensure data reliability.

## III. RESULTS AND DISCUSSION

A. Characterization of Phenylboronic Acid-Terminated Polyethylene Glycol (dB-PEG)

The structure of dB-PEG was characterized using a Varian Mercury Plus-400 NMR spectrometer. Deuterated chloroform ($CDCl_3$) was used as the solvent, and both $^1H$ NMR and $^{11}B$ NMR spectra were recorded at room temperature, as shown in Figure1, confirming the successful synthesis of dB-PEG.

$^1H$ NMR of dB-PEG: $^1H$ NMR (600 MHz, $CDCl_3$) showed peaks at 7-8 ppm corresponding to the hydrogens on the phenyl ring of dB-PBA, indicating the successful grafting of TAPBA.

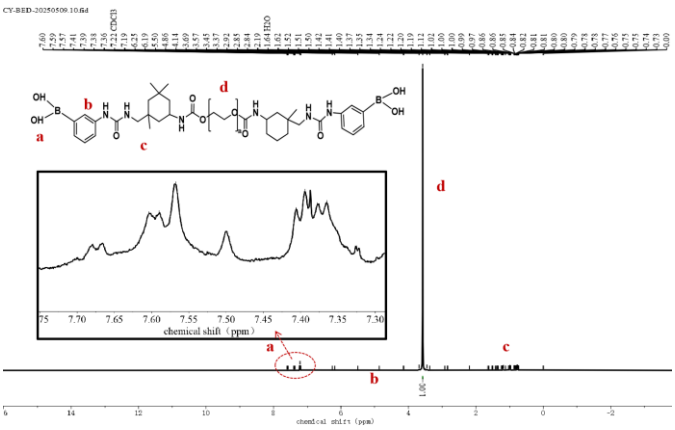


Figure1 $^1H$ NMR Spectrum of dB-PEG in Deuterated DMSO-$d_6$

B. X-Ray Diffraction (XRD) Analysis.

As shown in Figure 2, PVA exhibits a distinct diffraction peak at 19° -20° , which is one of the characteristic peaks of the PVA crystalline structure, indicating that PVA possesses a certain degree of crystallinity. Compared with pure PVA, the main characteristic diffraction peaks of the dB-PEG-PVA sample show no significant shift in position, suggesting that the crystal structure of the composite material remains fundamentally unchanged and that key structural features such as interplanar spacing largely retain the characteristics of PVA. In terms of peak intensity, changes in the degree of crystallinity

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are observed, as crystallinity affects diffraction peak intensity. Similarly, PEG-PVA samples exhibit main characteristic diffraction peaks at positions comparable to those of pure PVA, indicating that the crystal structure largely retains PVA features. Compared with pure PVA, the main crystalline structural features, such as the crystal planes corresponding to the diffraction peak positions, remain essentially unchanged, although factors such as the PEG content in different materials can affect the degree of crystallinity and thereby alter the peak intensity. The results also show a sharp peak near 20 ° , representing the formation of physical microcrystalline regions after PVA undergoes freeze-thaw treatment. This indicates the presence of a physically crosslinked network formed by PVA in the dB-PEG-PVA hydrogel.

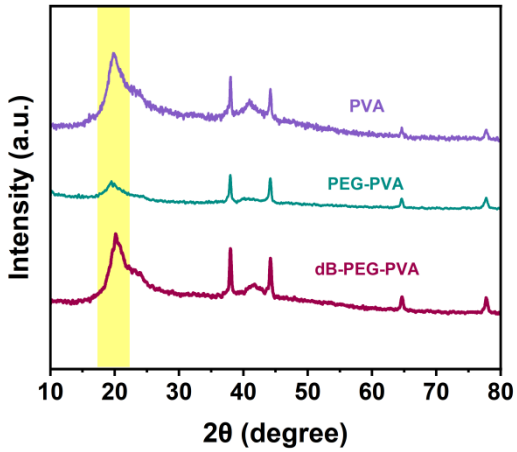


Figure 2 XRD Spectrum of PEG-PVA and dB-PEG-PVA hydrogels

C. Fourier Transform Infrared (FTIR) Spectroscopy Analysis.

As shown in Figure 3, TAPBA exhibits distinct absorption peaks in the regions of 3000–3500 $cm^{-1}$ and 1000-1500 $cm^{-1}$. At 1110 $cm^{-1}$ and 1370 $cm^{-1}$, the stretching vibration peaks of C–O and B–O in B–O–C indicate the formation of boronated ester bonds between PVA and TAPBA in the hydrogel. This demonstrates the presence of a chemically crosslinked network between dB-PEG and PVA in the dB-PEG-PVA hydrogel.

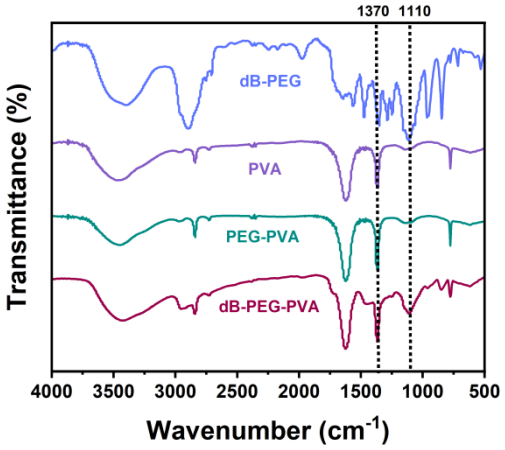


Figure 3 FTIR Spectrum of PEG-PVA and dB-PEG-PVA hydrogel.

D. Mechanical Properties Analysis

The tensile stress – strain curve is an important indicator of a material's mechanical properties, reflecting its ability to resist fracture under tensile load. As shown in Figure 4, the mechanical properties of PEG-PVA and dB-PEG-PVA hydrogels were evaluated through tensile testing. From the curves, it can be seen that the physically crosslinked hydrogel formed by pure PVA exhibits the highest tensile strength, reaching approximately 250 kPa, with a fracture elongation of around 400%, demonstrating good mechanical performance.

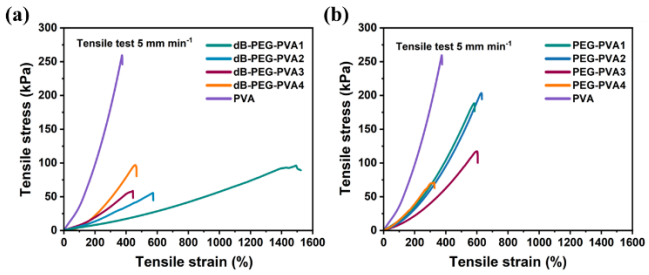


Figure 4 Tensile test curves of dB-PEG-PVA (a) and PEG-PVA (b).

After the introduction of dB-PEG or PEG, the tensile strength of the hydrogels decreased significantly, while the fracture elongation showed little change. This is likely because the incorporation of PEG increases the material's water absorption, thereby reducing the density of physical crosslinks. Notably, the fracture elongation of dB-PEG-PVA1 increased dramatically to 1500%. This may be attributed to the formation of additional boronated ester bonds between phenylboronic acid and PVA hydroxyl groups, while the original hydrogen-bonded crosslinking points of PVA are disrupted, resulting in a decrease in overall crosslink density. Under tensile stress, the long PEG chains orient and stretch along the direction of the applied force, significantly enhancing the material's fracture elongation.

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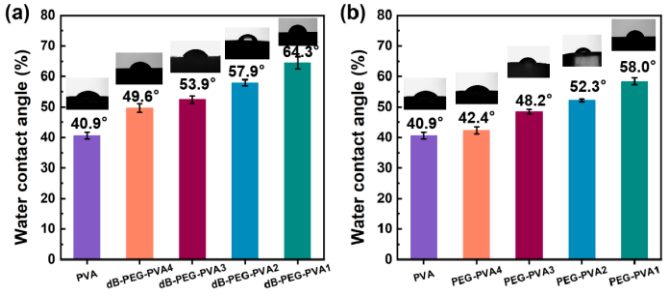


Figure 5 Contact angle measurements of dB-PEG-PVA (a) and PEG-PVA (b).

Water contact angle is an important measure of surface hydrophilicity or hydrophobicity. As shown in Figure 5, the contact angles of dB-PEG-PVA and PEG-PVA hydrogels are higher than that of pure PVA hydrogel and gradually increase with higher PEG content, but remain below 60 ° , indicating that the hydrogels still retain considerable hydrophilicity suitable for wound dressing applications. The hydrophilicity of a material is largely determined by the number of hydrophilic groups on its surface. The introduction of PEG slightly increases crosslink density, which marginally reduces hydrophilicity, but it remains within an appropriate range. Moderate hydrophilicity helps maintain a moist wound environment while preventing excessive water uptake that could compromise the gel structure.

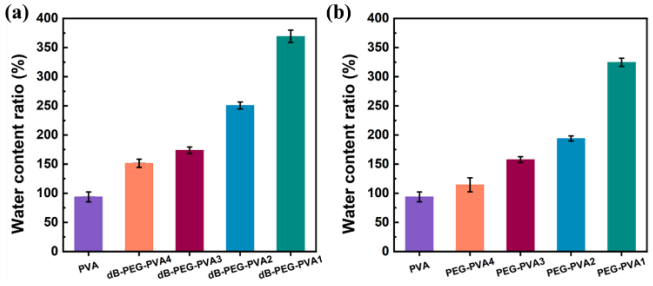


Figure 6 Water content measurements of dB-PEG-PVA (a) and PEG-PVA (b).

The water content of hydrogels is a critical property, as high-water content provides a moist environment conducive to wound healing. As shown in Figure 6, water absorption of the hydrogels increases significantly with PEG content, rising from 97% to 369%, indicating that PEG enhances the hydrogel's water retention capacity. The results demonstrate that both PEG-PVA and dB-PEG-PVA hydrogels exhibit good hydrophilicity, while dB-PEG-PVA hydrogels, due to chemical crosslinking, have higher crosslink density and superior water retention, making them highly promising as wound dressing materials.

To further investigate swelling behavior and optimize the hydrogel crosslinker ratio for improved performance in specific applications, PVA hydrogels with different PEG contents were tested. As shown in Figure 7, compared with conventional PVA hydrogels, PEG-PVA hydrogels containing boronate ester bonds exhibit superior swelling properties, enhancing biocompatibility and making them well-suited for wound dressing applications.

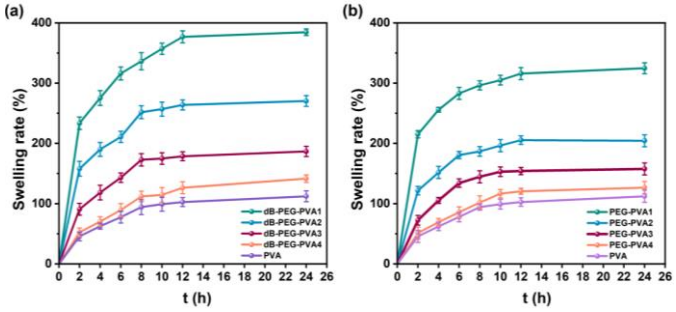


Figure 7 Swelling behavior curves of dB-PEG-PVA (a) and PEG-PVA (b).

E. Drug Loading and Release Analysis

As shown in Figure 8, a standard curve was established at 190 nm for Vancomycin (Van) over a concentration range of 1-25 μg/mL, with the equation y = 0.05983x-0.0691 and $R^2$ = 0.9994. By measuring the absorbance of Van@PEG-PVA1 and Van@dB-PEG-PVA1 and substituting into the equation, the drug release rate can be calculated. The release rate of Van from Van@dB-PEG-PVA1 at pH 5.5 reached a maximum of 86.3%. Based on these results, Van@dB-PEG-PVA1 hydrogel was selected for subsequent studies. To investigate its release behavior under different pH conditions, the hydrogel samples were immersed in environments with pH 5.5 and 7.4. The cumulative release over time is shown in Figure 13. Under acidic conditions (pH 5.5), the cumulative release of Van from Van@dB-PEG-PVA1 was significantly higher than at pH 7.4. After 72 hours at pH 5.5, the cumulative release reached a maximum of 86.3%, indicating that this hydrogel possesses excellent pH-responsive drug release capability.

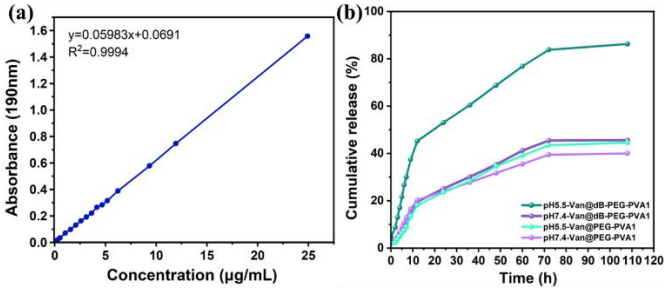


Figure 8 Standard curve of Vancomycin (Van) (a) and cumulative release profiles of Van from hydrogels (b).

F. In Vitro Antibacterial Performance Analysis

It is well known that the bacterial microenvironment is often mildly acidic. As shown in Figure 9, the Van@dB-PEG-PVA group

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exhibited the lowest number of bacterial colonies, indicating the strongest inhibitory effect against Staphylococcus aureus. This effect can be attributed to the following: in the acidic microenvironment formed by bacteria, the dynamic boronated ester bonds between the phenylboronic acid groups and PVA hydroxyls are cleaved, triggering pH-responsive release of Vancomycin (Van). Additionally, the phenylboronic acid groups themselves possess intrinsic antibacterial activity, which disrupts bacterial cell membrane integrity and exerts a bactericidal effect. This mechanism works synergistically with Van, significantly enhancing the overall antibacterial effect. Compared with the control group, the pH-responsive release mechanism allows the drug to be rapidly and selectively released within the infection microenvironment, ensuring local concentrations remain above the minimum inhibitory concentration. In contrast, the Van@PEG-PVA and Van@PVA groups showed similar and relatively high colony counts because they lack phenylboronic acid groups and therefore do not exhibit pH-responsive behavior. Drug release in these groups relies primarily on diffusion, resulting in low release efficiency. Moreover, PEG-PVA is only physically crosslinked, which limits both drug loading and release rate, making effective antibacterial action difficult to achieve.

These results visually confirm that pH-responsive, controlled Van release can reduce bacterial resistance. Specifically, Van@dB-PEG-PVA achieved a cumulative release of 86.3% under mildly acidic conditions, ensuring sustained antibacterial activity against S. aureus. The introduction of PEG reduces crosslink density, while dB-PEG, through its phenylboronic acid groups, enhances drug binding, synergistically improving antibacterial efficiency. Overall, Van@dB-PEG-PVA hydrogels achieve controlled drug release and high antibacterial efficiency through a pH-responsive dual-crosslinked network, providing an innovative strategy for addressing chronic wound biofilm infections. The low efficacy of PEG-PVA and pure PVA carriers further underscores the necessity of functional group incorporation, and offers guidance for future studies to fine-tune boronated ester bond density to balance drug release rate and long-term antibacterial activity.

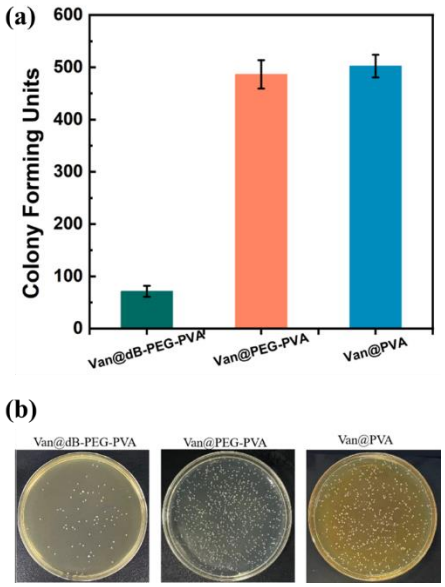


Figure 15 Colony count histogram (a) and antibacterial performance of drug-loaded hydrogels (b).

## IV CONCLUSION

In this study, a novel pH-responsive dB-PEG-PVA composite hydrogel dressing was developed to address the challenge of bacterial biofilm formation during chronic wound healing. Through a chemical – physical dual crosslinking strategy, TAPBA, PEG, and Vancomycin (Van) were introduced into a PVA-based hydrogel, endowing the material with excellent water absorption, water retention, and pH-responsive properties. Under mildly acidic conditions, the hydrogel enables controlled drug release, and the dynamic boronate ester bonds formed between phenylboronic acid groups and PVA hydroxyl groups provide a pH-responsive structure that works synergistically to enhance antibacterial activity, significantly inhibiting the growth of Staphylococcus aureus. The results demonstrate that the hydrogel exhibits good mechanical stability, swelling ability, and antibacterial activity, as well as favorable biocompatibility and the capability to maintain a moist environment. This novel hydrogel dressing provides an effective strategy for chronic wound treatment and is expected to overcome the limitations of traditional antibacterial dressings, such as uncontrolled antibiotic release and drug resistance, while promoting wound healing. Therefore, it shows great potential for clinical application and further research.

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