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Catechol-Modified Polyvinyl Alcohol Denture Adhesives with Enhanced Mucoadhesion for Improved Full-Denture Retention

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Abstract. The study aimed to develop and evaluate a novel denture adhesive patch based on catechol-functionalized polyvinyl alcohol (Cat-PVA), inspired by mussel adhesion mechanisms, to overcome the limitations of conventional denture adhesives and improve denture retention under moist and dynamic oral conditions. An edentulous jaw model with complete dentures was employed to compare Cat-PVA adhesives with a commercial product (Polident®). Biocompatibility was investigated using human gingival epithelial cells (HGECs) with CCK-8 assays. Spectroscopic analysis confirmed successful catechol incorporation, with a dose-dependent increase in catechol content. Cat-PVA adhesives exhibited significantly higher tensile and shear adhesion compared with unmodified PVA ($P < 0.05$). Cat-PVA3 denture adhesive demonstrated the strongest adhesion, maintaining nearly complete retention for 8 h under simulated saliva flow, while showing controlled degradation (~80% at 8 h). In the complete denture model, Cat-PVA3 achieved significantly greater tensile and shear bond strengths than Polident® ($P < 0.05$). Cat-PVA3 exhibited optimal performance, combining robust adhesion, prolonged retention, controlled disintegration, and excellent biocompatibility. Cat-PVA patches represent a promising, clinically translatable bioinspired strategy for denture fixation.

Keywords: denture adhesive, polyvinyl alcohol (PVA), catechol modification, mussel-inspired adhesion, mucoadhesion

1. Introduction

Population aging has increased edentulous patients, a condition sometimes managed as chronic. While conventional dentures are standard treatment, alveolar ridge resorption, especially in severe mandibular cases, compromises their retention and stability. This impairs mastication, speech, confidence, and quality of life.

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[1]. Multiple strategies exist, including denture adhesives. Implant-supported prostheses, though ideal, face barriers like cost, surgery, and bone volume. In contrast, denture adhesives provide a simple, cost-effective, and non-invasive way to improve retention and satisfaction, serving as a key prosthetic adjunct [2].

Despite their benefits, conventional denture adhesives have notable limitations. Specifically, paste and powder forms can leave oral residues, making cleaning difficult and posing hygiene concerns [3]. Second, their adhesive effect is typically short-lived, necessitating multiple applications per day and reducing patient compliance [4]. Recent material science explores polymers like chitosan, alginate, and PVA for their biocompatibility and mucoadhesion. PVA is particularly promising due to its FDA approval, hydroxyl groups for hydrogen bonding, and film-forming ability, though its inherent adhesion requires enhancement for oral use [5].

Inspired by mussel adhesion, their proteins enable strong attachment in wet, turbulent settings via byssal threads, primarily due to DOPA-rich mussel foot proteins (Mfp1–6), especially Mfp-3 and Mfp-5 [6]. DOPA's catechol groups are central to adhesion: they form hydrogen bonds, oxidize to quinones for self-polymerization and metal ion chelation, and undergo covalent reactions. Reversible noncovalent catechol interactions also enable self-healing [7]. Mussel-inspired systems are applied in bioadhesives, self-healing materials, coatings, and nanoparticles. Adhesive patches, offering controlled release and easy cleaning, present a favorable alternative to traditional forms by providing sustained retention without frequent reapplication [8].

A novel PVA-catechol denture adhesive patch was developed. The enhanced mucoadhesion, attributed to catechol's hydrogen bonding and covalent interactions, was confirmed by testing Cat-PVA against a commercial adhesive, demonstrating a promising bioinspired strategy to overcome current limitations.

2. Materials and methods

2.1. Materials

Poly(vinyl alcohol) (PVA, Mw 85–124 kDa, hydrolysis degree >99%) and dimethyl sulfoxide (DMSO) were purchased from Shanghai Aladdin Biochemical Technology Co., Ltd. (China). Sodium bisulfate monohydrate ($NaHSO_4 \cdot H_2O$) was obtained from Macklin Reagent (China). 3,4-Dihydroxy-L-phenylalanine (DOPA) was purchased from Sigma-Aldrich (USA).

2.2. Cell culture

Human gingival epithelial cells (HGECs) were obtained from GuangZhou Jennio Biotech Co., Ltd. (China). The cultures were maintained in Minimum Essential Medium (MEM; Gibco), supplemented with 10% fetal bovine serum (FBS; Gibco) and 1% penicillin–streptomycin (100 IU/ml). All cells were incubated at 37 °C in a humidified atmosphere containing 5% CO_2.

2.3. Synthesis of denture adhesives

PVA was dissolved in DMSO at 100 °C, followed by the addition of $NaHSO_4 \cdot H_2O$ and different amounts of DOPA (1/3, 1, 2 mmol) at 80 °C under nitrogen to synthesize Cat-PVA1–3 [9]. For adhesive film preparation, 0.2 g of lyophilized polymer obtained from the above step was dissolved in 2 mL PBS under heating, poured into a mold, and freeze-dried for 24 h, yielding denture adhesive films with ~1 mm thickness and ~1 cm diameter, which were stored under vacuum until further use.

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2.4. Characterization of denture adhesives and adhesive strength testing

The surface morphology of PVA and Cat-PVA films was examined by scanning electron microscopy (SEM). For ^{1}H NMR analysis, 1 mg of dried PVA or Cat-PVA samples was dispersed in 1 mL D_2O and analyzed at 400 Hz. UV–Vis absorption spectra were recorded for PVA and Cat-PVA solutions (0.05 mg/mL) to confirm successful incorporation of DOPA moieties.

Fresh porcine buccal mucosa was cut into strips (1.5 × 5 cm). The measurement of shear adhesion strength was conducted on a universal testing machine, with the crosshead set to advance at a speed of 10 mm/min. and maximum shear stress was recorded (n = 3). For tensile adhesion testing, a fresh buccal mucosa sample was fixed to a PTFE mold groove with cyanoacrylate glue, and an adhesive film was attached to a rectangular mold (2 × 2 × 5 cm)(n = 3).

2.5. Retention of denture adhesives under simulated saliva flow

Film retention was tested on fresh porcine buccal mucosa. The mucosa was collected, trimmed, and mounted on glass slides. Films were applied and immersed in PBS to simulate saliva flow. Detachment was recorded at 2, 4, 6, and 8 h, with five films per group in triplicate runs.

2.6. Edentulous jaw model and complete denture fabrication

An edentulous jaw model was reproduced in self-curing resin. Porcine buccal mucosa was attached to mimic oral tissue, and complete dentures were clinically fabricated and polymerized. Cylindrical resin handles were added at key positions for testing.

2.7. Retentive performance of denture adhesives in complete dentures

Adhesive films (0.3 × 1.0 cm) were placed on the fitting surfaces of maxillary complete dentures (bilateral first molar regions and palatal center) and mandibular complete dentures (bilateral first molar regions). Shear and tensile bond strengths were measured using a universal testing machine at 10 mm/min, and maximum stresses were recorded (n = 3). Commercial denture adhesive (Polident®) was used as a control under identical conditions for comparison.

2.8. Statistical analysis

All quantitative data are reported as the mean ± standard deviation (SD). Initial data processing and graphing were conducted in GraphPad Prism, while subsequent statistical analysis was carried out using SPSS (Version 21.0; IBM Corp.). To compare multiple groups, data were subjected to one-way analysis of variance (ANOVA) followed by Tukey's test for post hoc comparisons. Inter-group differences between two samples were analyzed with an independent-samples t-test. Statistical significance was designated at three levels: $P < 0.05$, $*P < 0.01$, and $**P < 0.001$.

3. Results

3.1. Characterization of denture adhesives

To verify the successful synthesis of denture adhesives, ^{1}H NMR spectroscopy was performed. As shown in Figure 1A, all Cat-PVA samples exhibited three characteristic peaks at 6.5–7.0 ppm, corresponding to the protons on the aromatic ring of DOPA, which were absent in pure PVA spectra.

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The intensity of these peaks increased with the DOPA content, confirming the gradual increase of catechol groups from Cat-PVA1 to Cat-PVA3. UV–Vis spectroscopy further supported the successful synthesis (Figure 1B). The macroscopic view of the dried adhesive is shown in Figure 1C, and the SEM images of the surface and cross-section are presented in Figures 1D and 1E, respectively.

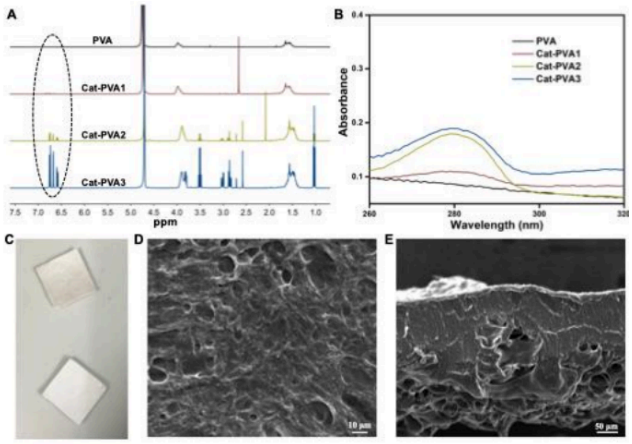


Figure 1. Characterization of denture adhesives: (A) ^{1}H-NMR spectra; (B) UV–Vis spectra; (C) macroscopic appearance; (D) SEM surface morphology; (E) SEM cross-sectional morphology

3.2. Adhesive strength of denture adhesives

Figures 2A–C illustrate the tensile testing process of Cat-PVA adhesives using porcine buccal mucosa. As shown in Figure 2D, the tensile bond strength increased with catechol content, with all Cat-PVA groups showing significantly higher values compared with PVA ($P < 0.0001$). Similarly, the shear bond strength results (Figure 2E) revealed that Cat-PVA2 and Cat-PVA3 exhibited significantly greater adhesion than PVA ($P < 0.01$). Among them, Cat-PVA3 demonstrated the highest bond strength and was therefore selected for subsequent experiments.

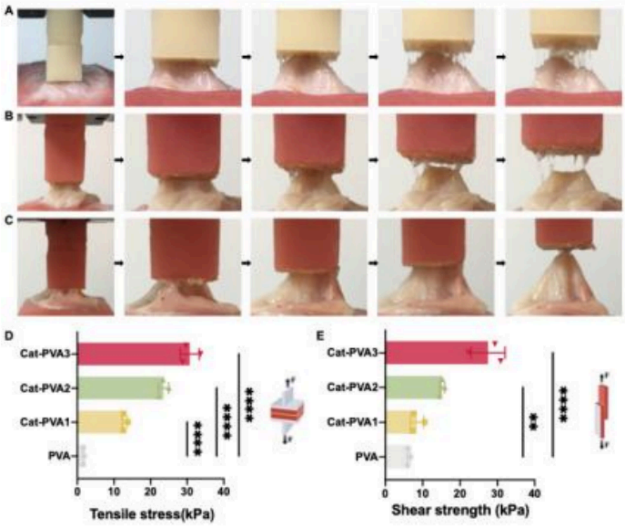


Figure 2. Adhesive strength of denture adhesives: (A–C) tensile testing process; (D) tensile bond strength; (E) shear bond strength

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3.3. Retention time and in vitro degradation

Retention performance of adhesive films was evaluated under simulated fluid flow conditions. Figures 3A–B show the testing setup using a rotating fluid system to mimic salivary flow. As shown in Figure 3C, PVA films partially detached after 1 h, with almost complete detachment by 6–8 h. In contrast, Cat-PVA3 films maintained nearly full retention up to 8 h, with only one film detaching across three independent tests. The in vitro degradation behavior was further assessed (Figure 3D). PVA films showed negligible degradation within 8 h, whereas Cat-PVA3 films exhibited progressive mass loss, reaching ~50% degradation at 6 h and ~80% at 8 h.

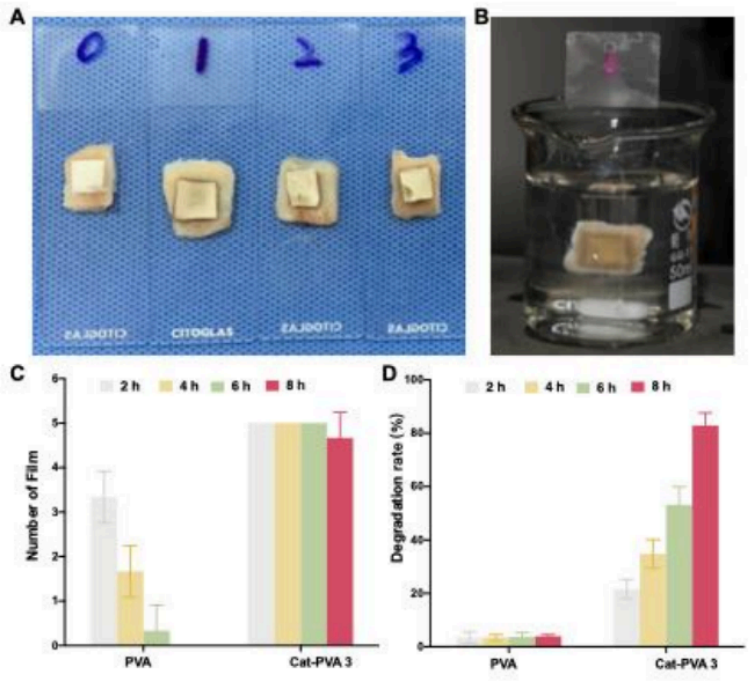


Figure 3. Retention and degradation of denture adhesives: (A–B) retention testing method; (C) retention results; (D) in vitro degradation profiles

3.4. Retentive performance in complete dentures and biocompatibility

Figure 4A shows the edentulous jaw model covered with porcine mucosa and the positioning of adhesive films on the denture fitting surface. Figures 4B and 4C depict the tensile and shear testing processes of Cat-PVA3 compared with a commercial denture adhesive (Polident®). As shown in Figures 4D and 4E, Cat-PVA3 exhibited significantly higher tensile and shear bond strengths in both maxillary and mandibular dentures compared with Polident® ($P < 0.05$). Biocompatibility was evaluated by co-culturing human gingival epithelial cells (HGECs) with different adhesives. As shown in Figure 4F, no significant cytotoxicity was observed after 1, 2, or 3 days of co-culture ($P > 0.05$), demonstrating good cytocompatibility of the prepared adhesives.

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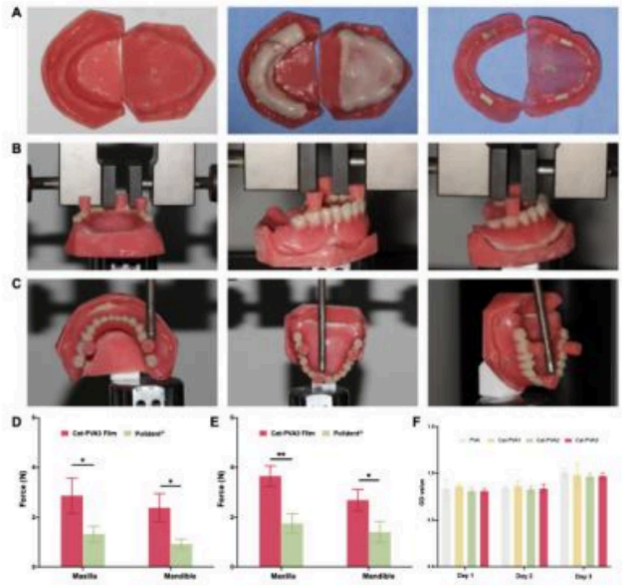


Figure 4. Retentive performance and biocompatibility of denture adhesives: (A) edentulous jaw model and adhesive positioning; (B) tensile testing method; (C) shear testing method; (D) tensile bond strength; (E) shear bond strength; (F) HGEC viability after co-culture with different adhesives

4. Discussion

In this study, Cat-PVA adhesives were synthesized and characterized. UV-vis and ^{1}H-NMR confirmed successful catechol incorporation, with UV-vis peak intensity and aromatic proton signals (6.5–7.0 ppm) both increasing proportionally with DOPA content.

Adhesive strength was quantitatively assessed via shear and tensile testing on fresh porcine buccal mucosa. While unmodified PVA showed some adhesion, catechol-modified Cat-PVA achieved over 30 kPa, with strength increasing with catechol content, providing robust and durable mucosal retention. Long-term adhesion of denture adhesives is crucial for denture stability. Using fresh porcine buccal mucosa for its structural similarity, detachment times were recorded. Results showed rapid detachment for PVA films but stable adhesion for catechol-modified adhesives [10]. Nevertheless, in vitro findings may not directly correlate with in vivo outcomes, as factors such as salivary secretion, oral motion, and enzymatic activity cannot be precisely replicated ex vivo, all of which may influence adhesive strength and residence time.

In addition, denture adhesives in the oral cavity are inevitably subjected to degradation [11]. The developed adhesives exhibited controlled disintegration (~50% at 6 h, ~80% at 8 h), balancing strong adhesion with convenient removal. In comparison with Polident®, Cat-PVA3 showed superior shear and tensile adhesion, significantly improving denture retention and demonstrating promising clinical potential.

Catechol modification dose-dependently enhances denture adhesive strength via hydrogen bonding and covalent interactions, including Michael addition and Schiff base reactions with mucosal components, significantly reinforcing PVA-DOPA adhesion [12]. Cytocompatibility with human gingival epithelial cells (HGECs), assessed via CCK-8 assays, showed negligible cytotoxicity, confirming favorable biocompatibility for further biomedical applications.

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5. Conclusion

This study successfully synthesized catechol-modified PVA (Cat-PVA) adhesives, which demonstrated superior adhesion, prolonged retention, and controlled disintegration compared to unmodified PVA and commercial adhesives. The enhanced performance stems from synergistic hydrogen bonding and covalent interactions between catechol groups and mucosal surfaces. Excellent cytocompatibility confirms its safety, highlighting Cat-PVA as a promising, clinically translatable bioinspired strategy for denture fixation.

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