

ORIGINAL ARTICLE

Tannic Acid Loaded Conductive PVA/Gelatin/NaCl Hydrogel For Adhesive Wearable Health Monitoring Devices

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ABSTRACT

Introduction: Polyvinyl alcohol (PVA), gelatin and sodium chloride (NaCl) (PVA/gelatin/NaCl)-based hydrogel exhibits optimal electrical conductivity, making it a promising candidate for wearable healthcare monitoring applications. However, the lack of adhesive properties limits the practical applications, especially in biomedical applications. Hence, this study aimed to enhance the adhesive properties of PVA/gelatin/NaCl-based hydrogel by loading tannic acid (TA), a natural polyphenol widely known for its adhesive properties and biocompatibility. **Materials and methods:** PVA/gelatin-based hydrogels were prepared and immersed in 10% and 20% NaCl solution, with a 0% NaCl hydrogel acting as a positive control. Subsequently, these hydrogels were immersed in 1%, 3% and 5% of TA solution for 24 h. The adhesive properties of the hydrogels were then evaluated by adhering to gloves-worn fingertips, onto a chicken's muscle, liver and bone, and on different materials, such as metal, plastic and rubber. **Results:** The results revealed that the PVA/Gel/NaCl₂₀/TA₅ exhibited strong adhesion across varying surfaces, such as fingertips, muscle, polyethylene terephthalate (PET), metal and gloves compared to the samples with other concentrations of NaCl and TA. Notably, the hydrogel exhibited sufficient adhesion to the biological tissues, which is crucial for health monitoring devices. **Conclusion:** The loading of TA enhanced the adhesive properties of the PVA/gelatin/NaCl-based hydrogel. Therefore, this hydrogel is well-suited for wearable health monitoring devices, where reliable attachment to both the components and biological tissues is critical.

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INTRODUCTION

In recent years, wearable technology or electronics has gained notable interest among researchers, owing to their capability to be implemented in countless applications, including healthcare for monitoring pulses (1), rehabilitation involving interactive devices (2), and in sport to attain crucial physiological data (3). This benefits both the users, such as patients or athletes and the therapists, as they monitor their patient's condition in real-time, allowing the determination of personalized

treatment to the users (2). Thus, wearable electronics are receiving great interest regardless of age factor. According to Grand View Research, the global wearable technology market was valued at USD 71.91 billion in 2023 and is approximated to extend at a compound annual growth rate (CAGR) of 14.6% (4). Therefore, the researchers are continuously enhancing the wearable electronics' functions, performance, and ease of wear to meet user demands, as early devices, were rigid in structure, making them uncomfortable to be worn (5). To overcome this issue, researchers seek alternatives with thin, flexible and soft nature electronics that are similar to the skin structure to maintain comfort at the same time it is biocompatible (5). When comparing available materials, polymers are the preferred candidates, as they allow the integration of sensors or electronics on

stretchable polymers to achieve flexibility. In particular, hydrogel, three-dimensional (3D) networks of cross-linked polymers that are made up of hydrophilic polymers are widely explored to be integrated with electronics, owing to their distinct and appealing properties, offering a multitude of applications in healthcare (6). These properties include high stretchability, flexibility, biocompatibility, and adjustable mechanical properties (7, 8). Besides that, hydrogels have great electrical conductivity when combined with conductive materials, which is significant for implementing sensors in wearable applications (9). Lastly, hydrogels can be tailored to have characteristics such as self-healing and adhesiveness, which are crucial for the wearable as they need to adhere to the skin and be able to withstand external forces and movements without failure (10).

Typically, the hydrogels are prepared from two distinct sources, either from naturally existing sources, such as alginate, hyaluronic acid, gelatin, collagen, and chitosan or synthetic-based sources including polylactic acid, polyethylene glycol, and polyvinyl alcohol (PVA) (11, 12). Both natural and synthetic hydrogels possess different characteristics that make them suitable for various applications. For instance, synthetic hydrogels are known for their increased strength and durability, making them suitable for load-bearing applications (13). On the other hand, hydrogels made from natural sources possess properties such as biodegradability and biocompatibility, making them appropriate to be used for wearables and implantable (13). Currently, hydrogel is commonly prepared using hybrid polymers, a combination of both natural and synthetic polymers (14). This is because the fabrication of hydrogels with hybrid polymers enables the utilisation of the biocompatibility and bioactivity of the natural polymers and the ability to tune the mechanical properties, chemical functionality, and ease of fabrication of synthetic polymers enables development of hydrogels with desired properties, application-specific functions, and improved performance (14). Therefore, in this study, a hybrid polymeric hydrogel containing PVA and gelatin will be utilised.

Among various types of biocompatible synthetic polymers, PVA produced by the hydrolysis of polyvinyl acetate, is selected due to its non-toxicity property (15). In addition, the solubility and film-forming properties make PVA a promising polymer to use as wearables as it has been explored to develop wound dressings (16). Moreover, PVA is commonly used because of its biocompatibility (17). However, the PVA hydrogel's insufficient mechanical properties and flexibility restrict its implementation as a wearable. Hence, to improve the mechanical properties of the PVA hydrogel, gelatin is considered to be added into the formulation of PVA hydrogel (18). Gelatin derived from the collagen of the animals offers excellent biodegradability, biocompatibility, and is able to enhance the mechanical

properties of the PVA-based hydrogels (18, 19). Hence, the PVA/gelatin-based hydrogel provides optimal biological and mechanical properties that are crucial for wearable applications. However, the conductivity of the PVA/gelatin-based hydrogel does not satisfy the criteria for wearable applications.

To resolve this issue, various methods have been explored by incorporating conductive materials within hydrogels. Some of the methods are the addition of conductive polymers, conductive nanoparticles, inorganic fillers, and ionic salts (20-24). Among these methods, the addition of ionic salt, in particular, sodium chloride (NaCl) is preferred due to its high solubility in water over a wide temperature range, making it a versatile and effective choice for various applications (25). In addition, NaCl is readily available at a low cost and is safer compared to other commonly used inorganic metal salts like LiCl, ZnCl₂, and FeCl₃ (26). Besides that, studies have demonstrated a significant increase in the conductivity of hydrogels with the incorporation of NaCl, with reported values reaching up to 7.26 S/m (27). On the other hand, NaCl also improves hydrogels' mechanical properties, such as toughness, resilience, and stretchability (28-30). Therefore, adding varying concentrations of NaCl to PVA/gelatin-based hydrogel can increase the conductivity, aiding in the development of efficient and functional wearable hydrogels.

Despite the PVA/gelatin/NaCl-based hydrogel exhibiting optimum electrical conductivity, it possesses one of crucial shortcomings, which is a lack of adhesive properties. This limits their implementation for wearable devices as they tend to detach or fall when subjected to movement. Hence, the PVA/gelatin/NaCl-based hydrogels must possess some degree of attachment to the skin to realize wearable health monitoring applications. To address this issue, tannic acid (TA), a natural polyphenol compound that is extracted from plants will be loaded into the PVA/gelatin/NaCl-based hydrogel. The TA is preferred in this study due to its excellent biocompatibility that causes the hydrogel to remain non-toxic and safe to be used for long term. For instance, in a study conducted by Cheng et al. (2022), the loading of TA had no adverse effect on biocompatibility, as the cell viability remained above 70% (31). In addition, the loading of TA contributes to strong adhesive properties of the hydrogel. The adhesion ability is provided by the presence of the catechol group in TA, which causes the formation of strong hydrogen bonds and hydrophobic interaction with the biological tissues. This enhances the adhesive properties of the hydrogels, ensuring a firm attachment to the skin regardless of subjecting to movement or the presence of moisture (32). It is evident from the study conducted by Yao et al. (2022), in which the PVA/HACC/TA hydrogel exhibited good adhesion to porcine skin, with an adhesion strength of 23.3 kPa, and adhered to human skin without detaching, even resisting wrist movement (33).

Apart from that, the TA consists of a large number of hydroxyl groups, which elevates the number of hydrogen bonds with the PVA/gelatin/NaCl-based hydrogel, hence, enhancing the mechanical strength of the hydrogel (32). For instance, the PVA/TA hydrogel demonstrated a tensile stress of 0.8 MPa, significantly higher than that of the PVA-only hydrogel, which exhibited approximately 0.4 MPa (34). This improvement is attributed to the phenolic groups in TA, which form a greater number of hydrogen bonds with PVA (34). On the other hand, the TA also possess antimicrobial properties that further improve the capability of hydrogel by preventing them from microbial contamination and oxidative degradation, thus, prolonging the longevity of the hydrogel (35). Therefore, the loading of TA into PVA/gelatin/NaCl-based hydrogel is anticipated to enhance the adhesive property of the hydrogel.

In this study, the combined effect of PVA/gelatin/NaCl-based hydrogel loaded with TA is anticipated to resolve the lack of adhesive property of the hydrogel. Various mediums were utilised to evaluate the adhesive property of the PVA/gelatin/NaCl-based hydrogel loaded with TA. This is followed by the analysis of the results on how the different loading of NaCl and TA affects the adhesive strength of the hydrogel. Lastly, the suitable TA concentration and PVA/gelatin/NaCl-based hydrogel were determined.

MATERIALS AND METHODS

In this section, the materials and the methods involved in preparing the adhesive PVA/gelatin/NaCl-based hydrogel with different TA concentrations were discussed. In addition, the various methods utilised to evaluate the adhesive properties of the samples were presented.

Materials

In this study, the required chemicals, such as the tannic acid (ACS reagent), gelatin powder, PVA, NaCl and deionised water were purchased from Sigma-Aldrich.

Preparation of PVA/gelatin-based Hydrogel with Varying NaCl Concentrations

A 7% mw/v gelatin solution was prepared by dissolving the gelatin powder into deionized water. Subsequently, the solution was heated at 50°C and stirred continuously for 1 hour until complete dissolution. Meanwhile, 10% w/v PVA solution was prepared by dissolving PVA powder into the deionized water for 1 hour at a temperature of 90°C. Then, the PVA and gelatin solutions were mixed with a ratio of 1:1 and stirred until a homogenous solution was obtained. The prepared formulation is then poured into a petri dish prior to the freeze-thawing process. The PVA/gelatin was frozen for 22 hours at -20°C followed by a thawing process for 2 hours at room temperature and this process was repeated for 3 cycles. Lastly, the prepared samples were immersed in 10%

and 20% NaCl solution at room temperature and the samples without the NaCl (0%) were served as positive control. According to a study conducted by Lei et al. (2022) moderate NaCl concentrations enhance both ionic conductivity and gel strength, whereas excessive levels may lead to protein aggregation and structural irregularities, ultimately compromising the hydrogel's integrity (36). Hence, in this study, 10% and 20% NaCl were selected to optimize conductivity while preserving structural integrity.

Preparation of PVA/gelatin/NaCl-based Hydrogel with Varying TA Concentrations

The prepared PVA/gelatin-based hydrogel with different salt concentrations (0%, 10% and 20%) were cut into small pieces of desirable dimension. The samples were named according to the concentrations of NaCl and TA. For instance, samples lacking NaCl with 1%, 3% and 5% TA loading were named PVA/Gel/NaCl₀/TA₁, PVA/Gel/NaCl₀/TA₃, and PVA/Gel/NaCl₀/TA₅. These samples serve as the positive control group. Similarly, the samples loaded with 10% NaCl and 1%, 3% and 5% TA were represented by PVA/Gel/NaCl₁₀/TA₁, PVA/Gel/NaCl₁₀/TA₃, and PVA/Gel/NaCl₁₀/TA₅ whilst, the samples loaded with 20% NaCl were named as PVA/Gel/NaCl₂₀/TA₁, PVA/Gel/NaCl₂₀/TA₃, and PVA/Gel/NaCl₂₀/TA₅. Prior to the immersion in the TA solution, the thickness of each sample was measured using a digital vernier calliper. The thickness was measured at three different positions of the samples and the average was recorded (37). Subsequently, the samples were immersed in different concentrations of TA solution, such as 1%, 3% and 5% for 24 h at room temperature (31). The samples were removed from the petri dish and washed repeatedly with deionised water to remove the residual TA that failed to form stable hydrogen bonds with the hydrogels (31). Lastly, the thickness of each sample was measured and the average was recorded.

Adhesive Analysis of Samples with Varying TA Concentration

The adhesive properties of the TA-loaded samples were qualitatively evaluated via the digital camera in different scenarios to evaluate their performance for biomedical applications (38). The adhesive strength was tested based on the ability of the samples to attach without complete detachment to the medium under test. For this purpose, the samples were tested by attaching to different mediums, such as the fingertip, onto a polyethylene terephthalate (PET) sheet, muscle, liver and bone of the chicken. Finally, the samples' adherence to different materials, such as metal, plastic and rubber (38) were tested. Each sample was gently pressed onto the target surface and held for approximately 1 minute to ensure sufficient contact and allow for adhesion. The test was repeated three times per condition to ensure consistency and reproducibility. All of these evaluations were conducted at room temperature (~ 25 °C) under ambient pressure.

Ethical Clearance

Ethical exemption for this study has obtained from Universiti Teknologi Malaysia Research Ethics Committee (UTM REC) with the ethical exemption approval number of UTMREC-2025-E3. The samples were sourced from commercially available poultry products intended for human consumption. No live animal handling or experimentation was conducted.

RESULTS

The adhesive properties of the samples were tested in various conditions to evaluate the effect of loading of different concentrations of TA to PVA/Gel/NaCl-based hydrogels. From Fig.1 it is noteworthy that the colour of the samples changed from white to brownish as the concentration of TA increases from 1% to 5%. Among the samples, the intensity of colour change in PVA/Gel/NaCl20 is greater, followed by PVA/Gel/NaCl10 and the least intensity of brownish is observed in PVA/Gel/NaCl10 samples. On the other hand, the samples immersed in 5% TA exhibit darker colour compared to the samples immersed in other TA concentrations.

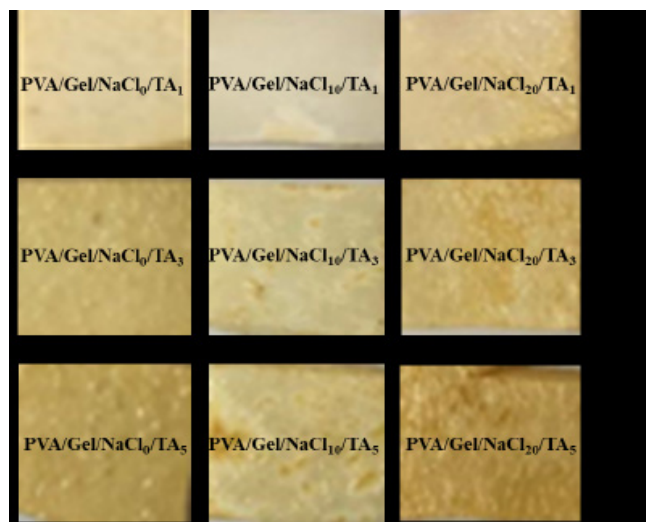


Fig. 1: Samples with 0%, 10% and 20% NaCl loaded with different concentration of (1%, 3%, and 5%) TA

Besides that, the thickness of the samples was measured before and after immersing in the TA solution. The average readings and standard deviation obtained before and after are presented in Table I. From Table I it can be seen that the thickness of the samples immersed in 1% TA is higher compared to the samples immersed in 3% and 5% TA regardless of the NaCl concentration. In addition, all measurements exhibited very low standard deviation values (ranging from 0.0055 to 0.0058 mm), indicating good measurement consistency and minimal variability across replicates. Subsequently, the adhesive strength of the samples was tested in distinct conditions. Firstly, the adherence of the samples was tested by attaching them to the gloves at the fingertip. Based on that, the adhesive properties of each sample were analyzed and summarised in Table II. According to Table II, the PVA/

Gel/NaCl0TA1 lacks adhesive properties compared to the PVA/Gel/NaCl0TA3 and PVA/Gel/NaCl0TA5. Concurrently, the PVA/Gel/NaCl0TA3 exhibited weak adhesion that requires strong pressed against the fingertip. It was observed that the edges were detaching, as shown in Fig.2.

Table I: Average thickness and standard deviation of the samples before and after immersing in TA solutions

Samples	Before		After	
	Average (mm)	Standard deviation (mm)	Average (mm)	Standard deviation (mm)
PVA/Gel/NaCl ₀ /TA ₁	2.90		3.18	0.0055
PVA/Gel/NaCl ₀ /TA ₃		0.0058	3.10	0.0057
PVA/Gel/NaCl ₀ /TA ₅			2.30	0.0058
PVA/Gel/NaCl ₁₀ /TA ₁	1.94		2.95	0.0058
PVA/Gel/NaCl ₁₀ /TA ₃		0.0057	2.53	0.0056
PVA/Gel/NaCl ₁₀ /TA ₅			1.31	0.0058
PVA/Gel/NaCl ₂₀ /TA ₁	2.23		3.38	0.0057
PVA/Gel/NaCl ₂₀ /TA ₃		0.0058	2.62	0.0058
PVA/Gel/NaCl ₂₀ /TA ₅			1.90	0.0056

Table II: Interpretation of adhesive capability of the samples on gloves at finger tip

Samples	Tannic Acid Loading		
	1 %	3 %	5 %
PVA/Gel/NaCl ₀	No adhesion property	<ul style="list-style-type: none"> • Weak adhesion even after strong press • The edges are detached 	Adhere with light press
PVA/Gel/NaCl ₁₀	Adhere with strong press	<ul style="list-style-type: none"> • Weak adhesion even after strong press • The edges are detached 	Adhere upon placement
PVA/Gel/NaCl ₂₀	Adhere with light press	Adhere upon placement	Strongly adhere upon placement



Fig. 2: PVA/Gel/NaCl0 with 3% TA detached at the edge

Meanwhile, the adhesive strength of the samples with NaCl is greater than the samples without NaCl regardless of the concentration of TA. For instance, the PVA/Gel/NaCl₁₀/TA₁ and PVA/Gel/NaCl₂₀/TA₁ samples were able to attach to the fingertip, however, a strong and light press was required, respectively. Similarly, the PVA/

Gel/NaCl₂₀/TA₃ and samples PVA/Gel/NaCl₂₀/TA₅ were attached to the fingertip without detaching and strongly adhered, respectively.

On the other hand, the adhesive properties of the samples were tested on different biological tissues, such as on chicken's muscle, liver and bone. This is done to evaluate the capability of the samples to adhere to the biological tissues as it is crucial for biomedical applications. Based on the testing, it was noteworthy that the samples with 3% and 5% TA loading were able to attach to the muscle, regardless of NaCl concentration, as depicted in Fig.3(a) and presented in Table III. Lastly, the ability of the samples to attach to different materials including plastic, represented by PET with a thickness of 0.1 mm, metal and rubber gloves were evaluated and the resulting adhesive properties were summarised and depicted in Table IV and Fig. 3(b),(c) and (d), respectively. It can be concluded that the samples with 5% TA and 20% NaCl can adhere strongly to the PET, and metal.

Table III: Summary of adhesive capability of the samples on different biological tissue

Samples	Biological tissues		
	Muscle	Bone	Liver
PVA/Gel/NaCl ₀ /TA ₁	Not adhere	Not adhere	Not adhere
PVA/Gel/NaCl ₀ /TA ₃	Not adhere	Not adhere	Not adhere
PVA/Gel/NaCl ₀ /TA ₅	Adhere	Not adhere	Not adhere
PVA/Gel/NaCl ₁₀ /TA ₁	Not adhere	Not adhere	Not adhere
PVA/Gel/NaCl ₁₀ /TA ₃	Adhere	Not adhere	Not adhere
PVA/Gel/NaCl ₁₀ /TA ₅	Adhere	Not adhere	Not adhere
PVA/Gel/NaCl ₂₀ /TA ₁	Not adhere	Not adhere	Not adhere
PVA/Gel/NaCl ₂₀ /TA ₃	Adhere	Not adhere	Not adhere
PVA/Gel/NaCl ₂₀ /TA ₅	Adhere	Not adhere	Not adhere

Table IV: Summary of adhesive capability of the samples on different materials

Samples	Materials		
	PET	Metal	Rubber gloves
PVA/Gel/NaCl ₀ /TA ₁	Adhere	Weakly adhere	Not adhere
PVA/Gel/NaCl ₀ /TA ₃	Adhere	Adhere	Adhere with strong press
PVA/Gel/NaCl ₀ /TA ₅	Strongly adhere	Strongly adhere	Adhere with light press
PVA/Gel/NaCl ₁₀ /TA ₁	Adhere	Weakly adhere	Adhere with strong press
PVA/Gel/NaCl ₁₀ /TA ₃	Adhere	Adhere	Weak adhesion
PVA/Gel/NaCl ₁₀ /TA ₅	Strongly adhere	Strongly adhere	Adhere with gentle press
PVA/Gel/NaCl ₂₀ /TA ₁	Adhere	Weakly adhere	Adhere with light press
PVA/Gel/NaCl ₂₀ /TA ₃	Adhere	Adhere	Adhere
PVA/Gel/NaCl ₂₀ /TA ₅	Strongly adhere	Strongly adhere	Strongly adhere

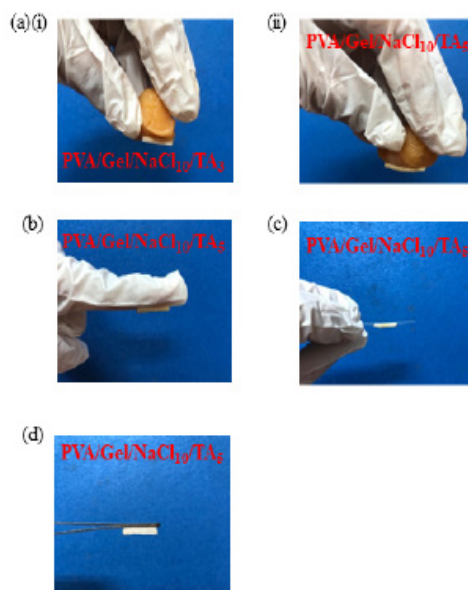


Fig. 3: (a) (i) PVA/Gel/NaCl₀/TA₃ and (ii) PVA/Gel/NaCl₁₀/TA₅ adhere to the muscle tissue (b) PVA/Gel/NaCl₁₀/TA₅ adhere to the rubber glove (c) PVA/Gel/NaCl₁₀/TA₅ adhere to the PET (d) PVA/Gel/NaCl₁₀/TA₅ adhere to the metal

DISCUSSION

The adhesive strength of the hydrogel is a crucial feature required for biomedical applications, especially for wearables. Hence, in this study, the TA was loaded into the PVA/Gel/NaCl-based hydrogels and the resulted adhesive properties were tested. Prior to the adhesive properties evaluation the colour and the thickness of the hydrogel samples were observed and recorded. Notably, the colour change in the samples was influenced by the presence of NaCl and depends on the factors, such as salting-out effect, and ionic strength. For instance, the high concentration of NaCl (20%) in the sample causes the salting-out effect that precipitates out the gelatine, thus, increasing the interaction between the TA and gelatin, resulting in intense colour change. Meanwhile, the absence of NaCl indicates no salting-out effect, hence, maximizes the interaction between the TA and gelatin, causing intense colour change but less than the sample with 20 % NaCl. In samples with 10% NaCl, the salting-out effect is balanced. Therefore, the gelatin is precipitated minimally, reducing the interaction between both TA and gelatin, leading to the least colour change. Besides that, the samples with 20% NaCl have high ionic strength which enhances the interactions between TA and gelatin, contributing to darker colour. Similarly, the absence of NaCl creates stronger interaction leading to intense colour change (39). However, the moderate ionic strength of 10% NaCl samples has sufficient ionic strength for shielding the charges on the gelatin to reduce the interaction's extent. Thus, a more stable hydrogel structure with the least colour change was

obtained (39).

On the other hand, the samples immersed in 5% TA exhibit darker colour due to the availability of a higher number of TA molecules that can interact with the hydroxyl groups in PVA and the amino groups in gelatin. As a result, there will be intense interaction including hydrogen bonding and hydrophobic interaction between the TA and hydrogel structure (40, 41). The increase in the interaction will saturate the hydrogels, maximizing the binding, thus, greater the colour change. Lastly, the high concentration of TA contributes to increased crosslinking between the TA and hydrogel (42). Therefore, alters the hydrogel's optical properties significantly and leads to a darker colour. Furthermore, aggregation on the surface of samples with 10% NaCl were observed regardless of TA concentration. This is due to the synergic effect of the ionic environment and introduction of TA, which causing precipitation of TA, resulting in aggregation (39).

Next, the thickness of the samples was reduced as the TA concentration increased. This is because of the presence of lower TA molecules in 1% TA to crosslink with the hydrogel. Therefore, the hydrogel can absorb more water, enabling it to have greater swelling capacity than other samples. Conversely, at 3% and 5% TA, more crosslinking will be occurred, thus, reducing the swelling ability of the hydrogel. Moreover, the fewer the TA molecules in 1% TA, the lower the binding between the TA and the hydrogels. Hence, there will be more free sites for water absorption that contribute to greater swelling ability. Meanwhile, the hydrogels immersed in 3% and 5% TA bind extensively, reducing the free sites for water absorption, and resulting in a reduction in swelling ability. Finally, the hydrophobic interactions between the TA and hydrogel are less in 1% TA compared to 3% and 5% TA. Thus, the hydrogel can absorb more water from the TA solution.

Subsequently, the PVA/Gel/NaCl0TA1 samples have very weak adhesive properties, which are not adequate to adhere to the fingertip, owing to the lower number of TA molecules in 1% TA, which is not sufficient to be crosslinked with the samples, leading to minimal hydrogen bonding. Meanwhile, the weak adhesion of PVA/Gel/NaCl0TA3 is contributed by the moderate crosslinking and hydrogen bonding, which slightly enhances the mechanical strength of the PVA/Gel/NaCl0TA3 hydrogels. However, the degree of crosslinking and hydrogen bonding are not optimal for forming strong adhesion. For PVA/Gel/NaCl0TA5 samples, a light press is required upon adhesion though a high number of TA molecules were present to provide greater adhesive strength.

In addition, the differences in the force required to adhere the samples were due to the distinct ionic strength, where the 20% NaCl significantly increases

the interactions between the TA and samples, resulting in some degree of adhesive strength that is greater than the samples with 10% NaCl. Meanwhile, the synergic effect of increment in the number of TA molecules and ionic strength that contribute to strong adhesion of PVA/Gel/NaCl₂₀/TA₃ and samples PVA/Gel/NaCl₂₀/TA₅ to the fingertip. Therefore, it can be concluded that PVA/Gel-based hydrogel with 20% NaCl and 5% TA loading significantly improves the adhesive strength.

For the evaluation with biological tissues, the sufficient adhesive property enables the samples to adhere to the muscle tissue. However, the samples failed to attach to the liver and bone, owing to the insufficient adhesive property (43). Lastly, the ability to adhere to PET, metal and rubber gloves owing to the high and moderate surface energy of these materials, respectively. Concurrently, the difference in the adhesion of the samples on gloves was discussed earlier.

CONCLUSION

In this paper, the PVA/Gel-based hydrogels with 0%, 10% and 20% of NaCl were loaded with 1%, 3% and 5% TA by immersing the samples for 24 h. The adhesive strength of these samples was evaluated under varying conditions including attaching the samples to gloves-worn fingertip, onto a chicken's muscle, liver and bone, and on different materials, such as metal, plastic and rubber. After the immersion in 24 h, it was observed that the PVA/Gel/NaCl0 and PVA/Gel/NaCl20 samples with 5% TA exhibited darker brownish colour than the samples with 1% and 3% TA. In addition, the samples with 10% NaCl exhibited less colour change regardless of TA concentrations. Besides that, the PVA/Gel/NaCl₂₀/TA₅ exhibited strong adhesion to the fingertip, muscle, PET sheet, metal and gloves compared to the other samples. From the findings, it is concluded that the PVA/Gel/NaCl₂₀/TA₅ hydrogel is appropriate to be used for wearable biomedical applications, as the synergic effect of NaCl and TA contributes to conductive and adhesivable hydrogels, thus, paving the pathway for the wearable health monitoring technologies, where both the properties are crucial. To further advance this research, future studies should focus on assessing the hydrogel's adhesion under dynamic movements, varying temperatures, and moist environments to better mimic real-world biomedical applications. Lastly, this study provided a qualitative evaluation of adhesion, future work should include quantitative assessments, such as peel strength and lap shear tests to deliver more comprehensive insights into adhesive behavior.

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REFERENCES

- Perez MV, Mahaffey KW, Hedlin H, Rumsfeld JS, Garcia A, Ferris T, et al. Large-Scale Assessment of a Smartwatch to Identify Atrial Fibrillation. *N Engl J Med*. 2019;381(20):1909-17. <https://doi.org/10.1056/NEJMoa1901183>
- De Fazio R, Mastronardi VM, De Vittorio M, Visconti P. Wearable Sensors and Smart Devices to Monitor Rehabilitation Parameters and Sports Performance: An Overview. *Sensors (Basel)*. 2023;23(4). <https://doi.org/10.3390/s23041856>
- Aroganam G, Manivannan N, Harrison D. Review on wearable technology sensors used in consumer sport applications. *Sensors*. 2019;19(9):1983. <https://doi.org/10.3390/s19091983>
- Research GV. Wearable technology market size, share & trends analysis report by product (head & eyewear, wristwear), by application (consumer electronics, healthcare), by region (Asia pacific, Europe), and segment forecasts, 2023 - 2030. n.d.
- Peng H. Wearable electronics. *Natl Sci Rev*. 2023;10(1):nwac193. <https://doi.org/10.1093/nsr/nwac193>
- Nuutila K, Eriksson E. Moist Wound Healing with Commonly Available Dressings. *Adv Wound Care (New Rochelle)*. 2021;10(12):685-98. <https://doi.org/10.1089/wound.2020.1232>
- Wu S, Xu C, Zhao Y, Shi W, Li H, Cai J, et al. Recent Advances in Chitosan-Based Hydrogels for Flexible Wearable Sensors. *Chemosensors*. 2023;11(1):39. <https://doi.org/10.3390/chemosensors11010039>
- Chen C-K, Chen P-W, Wang H-J, Yeh M-Y. Alkyl Chain Length Effects of Imidazolium Ionic Liquids on Electrical and Mechanical Performances of Polyacrylamide/Alginate-Based Hydrogels. *Gels*. 2021;7(4):164. <https://doi.org/10.3390/gels7040164>
- Liu K, Zhao Z, Zheng S, Liu A, Wang Y, Chen L, et al. High Ion-Conducting PVA Nanocomposite Hydrogel-Based Wearable Piezoelectric and Triboelectric Sensors for Harsh Environments. *Biomacromolecules*. 2024;25(7):4384-93. <https://doi.org/10.1021/acs.biomac.4c00436>
- Yang Y, Yao C, Huang W-Y, Liu C-L, Zhang Y. Wearable Sensor Based on a Tough Conductive Gel for Real-Time and Remote Human Motion Monitoring. *ACS Applied Materials & Interfaces*. 2024;16(9):11957-72. <https://doi.org/10.1021/acsami.3c19517>
- Patel L, Worch JC, Dove AP, Gehmlich K. The Utilisation of Hydrogels for iPSC-Cardiomyocyte Research. *International Journal of Molecular Sciences*. 2023;24(12):9995. <https://doi.org/10.3390/ijms24129995>
- Chen A, Deng S, Lai J, Li J, Chen W, Varma SN, et al. Hydrogels for Oral Tissue Engineering: Challenges and Opportunities. *Molecules*. 2023;28(9). <https://doi.org/10.3390/molecules28093946>
- Gan X, Wang X, Huang Y, Li G, Kang H. Applications of Hydrogels in Osteoarthritis Treatment. *Biomedicines*. 2024;12(4):923. <https://doi.org/10.3390/biomedicines12040923>
- Islam MR, Rahman MM, Dhar PS, Nowrin FT, Sultana N, Akter M, et al. The Role of Natural and Semi-Synthetic Compounds in Ovarian Cancer: Updates on Mechanisms of Action, Current Trends and Perspectives. *Molecules*. 2023;28(5). <https://doi.org/10.3390/molecules28052070>
- Delavari MM, Stiharu I. Preparation and Characterization of Eco-Friendly Transparent Antibacterial Starch/Polyvinyl Alcohol Materials for Use as Wound-Dressing. *Micromachines (Basel)*. 2022;13(6). <https://doi.org/10.3390/mi13060960>
- Boonsuk P, Sukolrat A, Kaewtatip K, Chantarak S, Kellarakis A, Chaibundit C. Modified cassava starch/poly(vinyl alcohol) blend films plasticized by glycerol: Structure and properties. *Journal of Applied Polymer Science*. 2020;137. <https://doi.org/10.1002/app.48848>
- Burgalassi S, Zucchetti E, Ling L, Chetoni P, Tampucci S, Monti D. Hydrogels as Corneal Stroma Substitutes for In Vitro Evaluation of Drug Ocular Permeation. *Pharmaceutics*. 2022;14(4):850. <https://doi.org/10.3390/pharmaceutics14040850>
- Sun M, Wang Y, Yao L, Li Y, Weng Y, Qiu D. Fabrication and Characterization of Gelatin/Polyvinyl Alcohol Composite Scaffold. *Polymers (Basel)*. 2022;14(7). <https://doi.org/10.3390/polym14071400>
- Andreazza R, Morales A, Pieniz S, Labidi J. Gelatin-Based Hydrogels: Potential Biomaterials for Remediation. *Polymers*. 2023;15(4):1026.
- Han IK, Song K-I, Jung S-M, Jo Y, Kwon J, Chung T, et al. Electroconductive, Adhesive, Non-Swelling, and Viscoelastic Hydrogels for Bioelectronics. *Advanced Materials*. 2023;35(4):2203431. <https://doi.org/https://doi.org/10.1002/adma.202203431>
- Wang J, Li Q, Li K, Sun X, Wang Y, Zhuang T, et al. Ultra-High Electrical Conductivity in Filler-Free Polymeric Hydrogels Toward Thermoelectrics and Electromagnetic Interference Shielding. *Advanced Materials*. 2022;34(12):2109904. <https://doi.org/https://doi.org/10.1002/adma.202109904>
- Sun S, Xu M, Zhao Y, Cheng T, Xiang Y, Liu X, et al. Nucleobase-Modified Adhesive and Conductive Hydrogel Interface for Bioelectronics. *ACS Applied Nano Materials*. 2023;6(22):21226-35. <https://doi.org/10.1021/acsanm.3c04282>
- Chen K, Lai W, Xiao W, Li L, Huang S, Xiao X. Low-Temperature Adaptive Dual-Network MXene

- Nanocomposite Hydrogel as Flexible Wearable Strain Sensors. *Micromachines*. 2023;14(8):1563. <https://doi.org/10.3390/mi14081563>
24. Chen J, Li B, Ma X, Zhou S, Gu Q, Bian H, et al. Modified lignin-induced composite hydrogels with good mechanical properties, adhesion, and UV resistance for strain sensors. *Journal of Applied Polymer Science*. 2023;140. <https://doi.org/10.1002/app.54643>
 25. Qing X, Liu Z, Katsaounis A, Bouropoulos N, Taurino I, Fardim P. Poly(vinyl alcohol)/Pullulan/NaCl Conductive Hydrogels with High Strength and Sensitivity for Wearable Strain Sensors. *ACS Applied Polymer Materials*. 2024;6(14):8105-15. <https://doi.org/10.1021/acsapm.4c00935>
 26. Cui W, Zheng Y, Zhu R, Mu Q, Wang X, Wang Z, et al. Strong Tough Conductive Hydrogels via the Synergy of Ion-Induced Cross-Linking and Salting-Out. *Advanced Functional Materials*. 2022;32(39):2204823. <https://doi.org/https://doi.org/10.1002/adfm.202204823>
 27. Ge X, Guo Y, Gong C, Han R, Feng J, Ji J, et al. High-Conductivity, Low-Impedance, and High-Biological-Adaptability Ionic Conductive Hydrogels for Ear-EEG Acquisition. *ACS Applied Polymer Materials*. 2023;5(10):8151-8. <https://doi.org/10.1021/acsapm.3c01368>
 28. Wei J, Wang J, Shao Z. Bioinspired cellulose nanofibrils and NaCl composited polyacrylamide hydrogels with improved toughness, resilience, and strain-sensitive conductivity. *Journal of Applied Polymer Science*. 2022;139(47):e53188. <https://doi.org/https://doi.org/10.1002/app.53188>
 29. Wang X, Li X, Wang B, Chen J, Zhang L, Zhang K, et al. Preparation of Salt-Induced Ultra-Stretchable Nanocellulose Composite Hydrogel for Self-Powered Sensors. *Nanomaterials*. 2023;13(1):157. <https://doi.org/10.3390/nano13010157>
 30. Xin Y, Liang J, Ren L, Gao W, Qiu W, Li Z, et al. Tough, Healable, and Sensitive Strain Sensor Based on Multiphysically Cross-Linked Hydrogel for Ionic Skin. *Biomacromolecules*. 2023;24(3):1287-98. <https://doi.org/10.1021/acs.biomac.2c01335>
 31. Cheng C, Peng X, Xi L, Wan C, Shi S, Wang Y, et al. An agar-polyvinyl alcohol hydrogel loaded with tannic acid with efficient hemostatic and antibacterial capacity for wound dressing. *Food & Function*. 2022;13(18):9622-34. <https://doi.org/10.1039/D2FO02251F>
 32. Liu H, Qin S, Liu J, Zhou C, Zhu Y, Yuan Y, et al. Bio-Inspired Self-Hydrophobized Sericin Adhesive with Tough Underwater Adhesion Enables Wound Healing and Fluid Leakage Sealing. *Advanced Functional Materials*. 2022;32. <https://doi.org/10.1002/adfm.202201108>
 33. Yao Q, Zheng W, Tang X, Chen M, Liao M, Chen G, et al. Tannic acid/polyvinyl alcohol/2-hydroxypropyl trimethyl ammonium chloride chitosan double-network hydrogel with adhesive, antibacterial and biocompatible properties. *Reactive and Functional Polymers*. 2022;179:105384. <https://doi.org/https://doi.org/10.1016/j.reactfunctpolym.2022.105384>
 34. Guo Y, An X, Fan Z. Aramid nanofibers reinforced polyvinyl alcohol/tannic acid hydrogel with improved mechanical and antibacterial properties for potential application as wound dressing. *Journal of the Mechanical Behavior of Biomedical Materials*. 2021;118:104452. <https://doi.org/https://doi.org/10.1016/j.jmbbm.2021.104452>
 35. Michalska-Sionkowska M, Warzyńska O, Kaczmarek-Szczepańska B, Łukowicz K, Osyczka AM, Walczak M. Characterization of Collagen/Beta Glucan Hydrogels Crosslinked with Tannic Acid. *Polymers*. 2021;13(19):3412. <https://doi.org/10.3390/polym13193412>
 36. Lei Y, Ouyang H, Peng W, Yu X, Jin L, Li S. Effect of NaCl on the Rheological, Structural, and Gelling Properties of Walnut Protein Isolate-κ-Carrageenan Composite Gels. *Gels*. 2022;8(5). <https://doi.org/10.3390/gels8050259>
 37. Saha I, Roy S, Das D, Das S, Karmakar P. Topical effect of polyherbal flowers extract on xanthan gum hydrogel patch - induced wound healing activity in human cell lines and male BALB/c mice. *Biomedical Materials*. 2023;18. <https://doi.org/10.1088/1748-605X/acc8e9>
 38. Zhang Z, Zhang Y, Liu Y, Zheng P, Gao T, Luo B, et al. Water-retaining and separable adhesive hydrogel dressing for wound healing without secondary damage. *Science China Materials*. 2023;66(8):3337-46. <https://doi.org/10.1007/s40843-022-2466-7>
 39. Cortez-Trejo MC, Figueroa-Córdenas JD, Quintanar-Guerrero D, Baigts-Allende DK, Manríquez J, Mendoza S. Effect of pH and protein-polysaccharide ratio on the intermolecular

- interactions between amaranth proteins and xanthan gum to produce electrostatic hydrogels. *Food Hydrocolloids*. 2022;129:107648. <https://doi.org/https://doi.org/10.1016/j.foodhyd.2022.107648>
40. Zhang X, Do M, Casey P, Sulistio A, Qiao G, Lundin L, et al. Chemical Modification of Gelatin by a Natural Phenolic Cross-linker, Tannic Acid. *Journal of agricultural and food chemistry*. 2010;58:6809-15. <https://doi.org/10.1021/jf1004226>
41. Sesia R, Ferraris S, Sangermano M, Spriano S. UV-Cured Chitosan-Based Hydrogels Strengthened by Tannic Acid for the Removal of Copper Ions from Water. *Polymers*. 2022;14(21):4645. <https://doi.org/10.3390/polym14214645>
42. Li Z, Chen Z, Chen H, Chen K, Tao W, Ouyang X-k, et al. Polyphenol-based hydrogels: Pyramid evolution from crosslinked structures to biomedical applications and the reverse design. *Bioactive Materials*. 2022;17:49-70. <https://doi.org/https://doi.org/10.1016/j.bioactmat.2022.01.038>
43. George MN, Liu X, Miller AL, Zuiker E, Xu H, Lu L. Injectable pH-responsive adhesive hydrogels for bone tissue engineering inspired by the underwater attachment strategy of marine mussels. *Biomaterials Advances*. 2022;133:112606. <https://doi.org/https://doi.org/10.1016/j.msec.2021.112606>