Jurnal Farmasi Teknologi Sediaan Dan Kosmetika
Volume 3, Nomor 1, Tahun 2026
Homepage : https://ejurnaljlm.com/index.php/jftsk/index *E-ISSN 3063-2226*
DOI : 10.70075/jftsk.v3i1.287

Characterization of Tannic Acid Chemically Crosslinked PVA Chitosan Hydrogel

Nana Nurindah Upara[1*], Muhammad Taupik[2], Multiani S. Latif[3], Nur Rasdianah[4], A. Mut'hi Andy Suryadi[5]

[1,2,3,4,5] *Department of Pharmacy, Faculty of Sport and Health, Universitas Negeri Gorontalo*
General Sudirman Street No. 06 Gorontalo City 96128, Indonesia

** Penulis Korespondensi. Email: nananurindah@gmail.com*

ABSTRAK

Hidrogel adalah material hidrofilik yang dapat menyerap dan menahan sejumlah besar air melalui pembentukan ikatan hidrogen antara gugus fungsional polimer untuk membentuk jaringan tiga dimensi yang stabil. Karakteristik fisikokimia hidrogel sangat dipengaruhi oleh proses ikatan silang yang menentukan kepadatan jaringan dan stabilitas preparasi. Studi ini bertujuan untuk menentukan pengaruh variasi konsentrasi asam tanat sebagai pengikat silang kimia terhadap karakteristik fisikokimia hidrogel berbasis polivinil alkohol (PVA)-kitosan. Hidrogel diformulasikan dengan berbagai konsentrasi asam tanat sebesar 1%, 3%, dan 5%, kemudian dievaluasi termasuk uji organoleptik, pH, viskositas, daya sebar, adhesi, indeks pembengkakan, uji iritasi, dan karakterisasi menggunakan Fourier Transform Infrared (FTIR). Hasil menunjukkan bahwa peningkatan konsentrasi asam tanat menyebabkan pembentukan jaringan hidrogel yang lebih padat, yang ditandai dengan penurunan nilai indeks pembengkakan dan peningkatan adhesi serta perubahan viskositas. Nilai pH semua formula berada dalam kisaran pH fisiologis kulit. Uji iritasi pada 20 panelis menunjukkan bahwa semua formula tidak menyebabkan reaksi iritasi. Analisis FTIR mengkonfirmasi adanya interaksi molekuler antara PVA, kitosan, dan asam tanat melalui pembentukan ikatan hidrogen. Secara keseluruhan, variasi konsentrasi asam tanat sebagai pengikat silang kimia mempengaruhi karakteristik fisikokimia hidrogel PVA-kitosan dan menghasilkan sediaan yang stabil dengan kualitas yang baik.

Kata Kunci: Hidrogel; PVA-Kitosan; Asam Tanat; Ikatan Silang Kimia; Karakterisasi.

Diterima:	*Disetujui:*	*Online:*
02-02-2026	27-02-2026	27-02-2026

ABSTRACT

Hydrogel is a hydrophilic material that can absorb and retain large amounts of water through the formation of hydrogen bonds between polymer functional groups to form a stable three-dimensional network. The physicochemical characteristics of hydrogels are greatly influenced by the cross-linking process that determines the network density and stability of the preparation. This study aims to determine the effect of variations in the concentration of tannic acid as a chemical crosslinker on the physicochemical characteristics of polyvinyl alcohol (PVA)-chitosan-based hydrogels. Hydrogels were formulated with varying concentrations of tannic acid of 1%, 3%, and 5%, then evaluated including organoleptic tests, pH, viscosity, spreadability, adhesion, swelling index, irritation test, and characterization using Fourier Transform Infrared (FTIR). The results showed that increasing the concentration of tannic acid caused the formation of a denser hydrogel network, which was characterized by a decrease in the swelling index value and an increase in adhesion and changes in viscosity. The pH values of all formulas were within the physiological pH range of the skin. Irritation tests on 20 panelists showed that all formulas did not cause irritation reactions. FTIR analysis confirmed the presence of molecular interactions between PVA, chitosan, and tannic acid through hydrogen bond formation. Overall, variations in the concentration of tannic acid as a chemical *crosslinker affected the physicochemical characteristics of the PVA–chitosan hydrogel and resulted in stable preparations with good quality.*

Keywords: *Hydrogel; PVA-Chitosan; Tannic Acid; Chemical Crosslink; Characterization.*

Received:	*Accepted:*	*Online:*
2026-02-02	2026-02-27	2026-02-27

1. Introduction

Gels are semisolid systems consisting of suspensions formed from small inorganic particles or large organic molecules dispersed within and penetrated by a liquid [1]. In biomedical and pharmaceutical applications, gels are widely used as formulation bases because they are soft, easy to apply, and can provide sustained release of active ingredients [2]. A more advanced gel system that plays an important role in drug delivery is the hydrogel.

Hydrogels are materials capable of absorbing large amounts of water without dissolving, owing to a hydrophilic polymer network that binds water molecules through hydrogen bonding. Their structure resembles that of biological tissues, making hydrogels highly suitable for medical applications such as wound dressings, drug delivery systems, and tissue engineering [3]. Hydrogels are generally composed of one or more polymers, either natural or synthetic. Polymers play a key role in forming the network structure because their long chains can interact and connect with each other, producing a stable three dimensional system. The choice of polymer strongly determines hydrogel characteristics, including elasticity, mechanical strength, and biocompatibility (4). One of the most widely used polymers in hydrogel formulations is poly(vinyl alcohol) (PVA), often in combination with chitosan.

Poly(vinyl alcohol) (PVA) is a hydrophilic polymer rich in hydroxyl (–OH) groups, which confer water solubility, high flexibility, and good chemical stability. PVA is also non-toxic and compatible with various natural materials. PVA-based hydrogels exhibit strong mechanical stability and high water-retention capacity, making them promising candidates for biomedical applications [(5). Chitosan is a natural polymer obtained by deacetylation of chitin, which is abundant in the exoskeletons of shrimp and crabs. Chitosan is known for its antibacterial activity, biodegradability, and good biocompatibility. When combined with PVA, chitosan can enhance the bioactivity and mechanical performance of hydrogels, making them suitable for applications such as wound healing and drug delivery (6).

To obtain a stable hydrogel network, the two polymers must be interconnected through a crosslinking process. Crosslinking forms bridges between polymer chains, preventing the system from dissolving easily and providing adequate structural strength. In general, crosslinking mechanisms are classified into two categories: physical crosslinking and chemical crosslinking (7).

Physical crosslinking involves non covalent interactions such as hydrogen bonding, ionic interactions, or freeze thaw cycling. The main advantages of physical crosslinking are its reversible nature and the absence of additional chemical crosslinkers. However, the resulting hydrogels often exhibit relatively low mechanical strength. In contrast, chemical crosslinking involves the formation of covalent bonds using crosslinking agents such as glutaraldehyde, epichlorohydrin, or formaldehyde. Although this approach generally produces stronger networks, these agents may leave toxic residues that can reduce the biocompatibility and safety of the hydrogel (8). As a safer alternative, researchers have increasingly explored chemical crosslinkers such as tannic acid.

Tannic acid is a polyphenolic compound widely found in tea, tree bark, and various fruits. It contains multiple hydroxyl groups that can form hydrogen bonds and complexes with the amine groups of chitosan and the hydroxyl groups of PVA. These interactions can generate a robust hydrogel network without the need for toxic chemical reagents (9). In addition, tannic acid exhibits antibacterial, antioxidant, and anti-inflammatory activities, which may promote wound healin (10).

Several studies support the role of tannic acid as a chemical crosslinker. A study on PVA/PEI hydrogels crosslinked with tannic acid reported that increasing TA concentration improved tensile strength, swelling capacity, and antibacterial activity; however, excessive concentrations could render the hydrogel more brittle (11). Similar findings were reported by researchers who developed PVA–chitosan/tannic acid hydrogels with high self-healing ability and good structural stability (12). Based on the above rationale, this study aimed to design and characterize a PVA–chitosan hydrogel using tannic acid as a safer chemical crosslinker. The developed system is expected to serve as an effective alternative for pharmaceutical product development, particularly for hydrogel-based wound healing applications.

2. Methods

This study used a true experimental design conducted at the Pharmaceutical Technology Laboratory, Department of Pharmacy, Faculty of Sports and Health, Universitas Negeri Gorontalo, from September 2025 to January 2026. It was a true experimental study aimed at characterizing PVA–chitosan hydrogels chemically crosslinked with tannic acid.

Material and Equipment

The equipment used included an analytical balance, Petri dishes, porcelain crucibles, a climatic chamber, beakers, graduated cylinders (Pyrex), a hot plate magnetic stirrer (IKA® C-MAG HS 7), a pH meter (Hanna Instruments), droppers, and a digital thermometer (Thermo Scientific). The materials used were 70% ethanol, 1% acetic acid, distilled water, chitosan, poly(vinyl alcohol) (PVA), and tannic acid.

Formulation of PVA–Chitosan Hydrogels

PVA–chitosan hydrogels were prepared in three formulations (F1, F2, and F3), with a total weight of 30 g for each formulation. Chitosan was used as a polymer at a fixed concentration of 2% in all formulations, while poly(vinyl alcohol) (PVA) was also used at a fixed concentration of 7%. The formulation variable was the concentration of tannic acid as a chemical crosslinker, which was set at 1% for F1, 3% for F2, and 5% for F3. Distilled water was used as the solvent and added to a final volume of 100%.

Chitosan Preparation

A 2% chitosan solution was prepared by dissolving chitosan in 1% acetic acid. The dissolution process was carried out using a magnetic stirrer at room temperature for 3 h until a homogeneous solution was obtained. Stirring facilitated the interaction between the amino groups of chitosan and acetic acid, enabling optimal dissolution. The resulting preparation was a 2% chitosan solution with a slightly viscous appearance.

PVA Preparation

A 7% PVA solution was prepared by dissolving poly(vinyl alcohol) (PVA) in distilled water. Dissolution was carried out at 70–90°C with continuous stirring for 2 h until a homogeneous solution was obtained. The resulting 7% PVA solution was clear and slightly viscous.

Hydrogel Preparation (Mixing Process)

The mixing process was carried out by adding the PVA solution into the chitosan solution, followed by stirring with a magnetic stirrer for 2 h until a homogeneous mixture was obtained. The mixture was then poured into Petri dishes or ointment jars and covered with plastic wrap to prevent contamination.

Tannic Acid Crosslinking

Tannic acid crosslinking was performed by adding tannic acid solution at varying concentrations of 1%, 3%, and 5% (w/v) for each formulation. The mixtures were then stirred until homogeneous, cast into Petri dishes, frozen at −80°C for 24 h, and subsequently freeze-dried to obtain porous hydrogels with a stable three-dimensional structure.

Evaluation of Formulation Quality

Formulation quality evaluation included organoleptic assessment, spreadability, adhesiveness, pH, viscosity, swelling behavior, and irritation testing. Organoleptic evaluation was conducted visually and sensorially to assess shape, color, odor, texture, elasticity, and taste using a scoring method by 20 untrained panelists. Spreadability and adhesiveness tests were performed by placing 1 g of hydrogel between two glass slides under a specified load, then measuring the spread diameter and detachment time as indicators of consistency and adhesion. The pH was measured using a calibrated pH meter to ensure compatibility with the physiological skin pH range (4.5–6.5), while viscosity was determined using a Stormer viscometer at various rotational speeds. Swelling was evaluated by immersing the hydrogel in distilled water until a constant weight was achieved to calculate the swelling ratio. Irritation testing was conducted in 20 volunteers by observing skin reactions up to 72 h after application.

Evaluation with FTIR

An FTIR spectrometer (FTIR-C68199, PerkinElmer) was used to analyze the functional groups of the crosslinked hydrogel films. Samples were scanned over a wavenumber range of 4000–500 cm^{-1} at a resolution of 4 cm^{-1}. The dried hydrogel film (approximately 0.5 mm thick) was placed in a PET plastic holder and sealed tightly prior to FTIR analysis of the obtained hydrogel film (13).

Data Analysis

Analysis of variance (ANOVA) was used to compare more than two mean values. Data were analyzed using one-way ANOVA. If the ANOVA indicated a statistically significant difference ($p < 0.05$), Fisher's least significant difference (LSD) post hoc test was performed to compare pairs of formulations. If the data were not normally distributed or did not meet the homogeneity of variance assumption, non-parametric analysis was applied using the Kruskal–Wallis test, followed by pairwise comparisons using the Mann–Whitney U test (14).

Ethical approval

This study received ethical approval from the Health Research Ethics Committee (KEPK), Universitas Negeri Gorontalo under Ethical Clearance Recommendation No. 216A/UN47.B7/KE/2025 (issued on 31 October 2025) for the research protocol entitled "Characterization of PVA–Chitosan Hydrogels Chemically Crosslinked with Tannic Acid" (Protocol Version 1; 20 October 2025), valid from 31 October 2025 to 31 October 2026. All study procedures were conducted in accordance with the approved protocol and relevant ethical principles, including participant safety and data confidentiality.

3. Results and Discussion

Based on the present study, the results comprised the characterization and quality evaluation of PVA and Chitosan hydrogels chemically crosslinked with tannic acid.

Hydrogel preparation

Hydrogel preparation began with the preparation of chitosan and PVA solutions. Chitosan (2%) was first dissolved in 1% acetic acid to obtain a homogeneous chitosan solution. Under acidic conditions, the amine groups of chitosan are protonated to NH_3^+, which increases its solubility [15]. The solution was stirred using a magnetic stirrer at high speed. Meanwhile, a 7% PVA solution was prepared in distilled water at 70–90°C to disrupt intramolecular hydrogen bonding within PVA, thereby producing a clear and homogeneous solution [4]. Mixing the PVA solution into the chitosan solution enabled intermolecular interactions to occur, primarily through hydrogen bonding between the hydroxyl groups of PVA and the amine groups of chitosan [16]. The process was then continued by adding tannic acid solution at concentrations of 1%, 2%, and 3% as a chemical crosslinking agent. When PVA, chitosan, and tannic acid were combined during hydrogel preparation, complex intermolecular interactions occurred, mainly hydrogen bonding and electrostatic interactions, resulting in a more integrated three-dimensional network than that formed by each polymer alone. Tannic acid is a polyphenolic compound containing multiple reactive phenolic (–OH) groups, allowing multipoint hydrogen bonding with both PVA and chitosan. In PVA, hydroxyl (–OH) groups interact strongly with the phenolic groups of tannic acid via hydrogen bonding. Meanwhile, chitosan, which contains amine and hydroxyl groups, can also bind to tannic acid through hydrogen bonding [12].

Evaluation of Formulation Quality

Table 1. Evaluation of Formulation Quality Results

Evaluation	F1 (1% Tannic Acid)	F1 (2% Tannic Acid)	F1 (3% Tannic Acid)
Organoleptic	Clear yellowish, thick, distinctive aroma, homogeneous	Cloudy white, slightly thick, distinctive aroma, homogeneous	Ivory white, slightly thick, has a distinctive aroma, homogeneous
Viscosity 12 rpm (cP)	5753,3333 ± 25,16611	5210,0000 ± 10,00000	3830,0000 ± 30,00000
Viscosity 30 rpm (cP)	4750,0000 ± 20,0000	3280,0000 ± 10,00000	2560,0000 ± 10,00000
Spread Power (cm)	6,7333 ± 0,11547	7,3333 ± 0,20817	7,3333 ± 0,15275
Adhesive Power (second)	552,3333 ± 28,21938	683,0000 ± 55,43465	660,0000 ± 72,13182

Swelling Index (%)	414,6667 ± 0,57735	319,6667 ± 0,57735	148,6667 ± 0,57735
pH	6,2733 ± 0,00577	6,2667 ± 0,00577	6,2800 ± 0,01000

Data Source: Processed primary data, 2025

Based on Table 1, the organoleptic evaluation indicated that increasing the concentration of tannic acid as a crosslinker influenced the physical characteristics of the PVA–chitosan hydrogels. Formula F1 (1%) showed a clear yellowish appearance with a viscous texture, whereas F2 (3%) and F3 (5%) exhibited progressively higher turbidity, ranging to an ivory white color with a slightly viscous texture. This color change is likely associated with enhanced crosslinking interactions between the phenolic groups of tannic acid and the PVA–chitosan polymer chains, which can reduce hydrogel transparency. All formulations had a relatively similar characteristic odor and good homogeneity, indicating that gel formation proceeded properly without phase separation [17]. Can be seen in table 1

Viscosity testing showed that all formulations exhibited a decrease in viscosity with increasing rotational speed (rpm), indicating pseudoplastic (shear-thinning) flow behavior. Formula F1 had the highest viscosity compared with F2 and F3, while the more pronounced viscosity reduction observed in F2 and F3 suggests that higher tannic acid concentrations produced a stiffer hydrogel network that deformed more readily due to over-crosslinking. This observation is consistent with reports in the literature indicating that a high crosslinking density restricts polymer chain mobility, thereby reducing flexibility and altering the rheological behavior of hydrogels. [11][12].

The spreadability test results showed that increasing tannic acid concentration enhanced the spreadability of the PVA–chitosan hydrogels. Formula F1 exhibited the lowest spreadability, whereas F2 and F3 demonstrated higher spreadability values, indicating that these formulations were easier to apply and could be more evenly distributed on the skin surface. All formulations met the spreadability requirement specified in SNI No. 06-2588-1992, i.e., 5–7 cm [18]. The adhesiveness test showed that formula F2 (3% tannic acid) had the highest adhesiveness compared with F1 and F3, indicating better adhesion to the application surface. All formulations met the minimum adhesiveness requirement of more than 1 s [19]. However, F3 exhibited a decrease in adhesiveness, which may be attributable to an overly dense network structure that reduced the gel's ability to maintain adhesive interactions [2].

The pH test results showed relatively uniform pH values across all formulations (6.26–6.27), which remained within the physiological skin pH range specified by SNI 16-3499-1996 (4.5–8). These findings indicate that varying tannic acid concentrations did not significantly affect the formulation acidity and suggest that the hydrogels are safe for topical use and unlikely to cause skin irritation [20].

The swelling test results indicated that the swelling index of PVA–chitosan hydrogels decreased as the tannic acid concentration increased. Formula F1 exhibited the highest swelling capacity, followed by F2 and F3, both before and after the freeze–thaw treatment, due to the increased crosslink density that limited water uptake. Nevertheless, the swelling values remained within the range of 200–500%, which is considered suitable for wound dressing applications [21]. This trend is consistent with previous studies reporting that hydrogel water absorption (swelling ability/swelling ratio) decreases when tannic acid content as a crosslinker increases—indicating that higher TA concentrations reduce hydrogel swelling because the network becomes denser [2]. Increased TA content restricts hydrogel expansion because a higher crosslink density produces a more compact structure [22].

Table 2. Results of the Irritation test

Time	Response				Panelist
	Itchy	Swollen	Reddish	Hot/Warm	
24 Hour	-	-	-	-	20
48 Hour	-	-	-	-	20
72 Hour	-	-	-	-	20

Data Source: Processed primary data, 2025

Based on Table 2, the irritation test showed no evidence of irritation in any participant throughout the 72-h observation period. At 24, 48, and 72 h, all panelists (n = 20) tested negative for itching (pruritus), swelling (edema), redness (erythema), and sensations of burning or warmth on the skin. These findings indicate that the formulated hydrogel preparations exhibit good dermal tolerability and an acceptable safety profile. The absence of irritation suggests that the constituent materials, including PVA, chitosan, and tannic acid as the chemical crosslinker, did not induce adverse effects on the skin. This outcome may also be related to the pH of the formulations, which remained within the physiological skin pH range, thereby minimizing the risk of irritation. In line with previous reports, composite hydrogels based on poly(vinyl alcohol) (PVA) and chitosan have been evaluated for skin irritation as part of biocompatibility testing for wound dressing applications; in vivo irritation and skin sensitization tests have demonstrated minimal irritation and no sensitization responses in rabbits and guinea pigs. Additional evaluations have also reported no acute toxicity following extraction and low hemolysis (<3%), further supporting the biological compatibility of PVA/chitosan hydrogels [23]. Can be seen in table 2.

Table 3. Data Analysis Results

Parameters	Statistic Test	Statistic Value	df	Sig.	Information
Viscosity 12 rpm	ANOVA	F = 5417,286	2	0,000	Significant
Viscosity 30 rpm	ANOVA	F = 18.688,500	2	0,000	Significant
Adhesive Power	ANOVA	F = 4,827	2	0,056	Not significant
Spread Power	Kruskal–Wallis	H = 5,492	2	0,064	Not significant
Swelling	Kruskal–Wallis	H = 7,385	2	0,025	Significant
pH	Kruskal–Wallis	H = 3,582	2	0,167	Not significant

Data Source: Processed primary data, 2025

Table 3 summarizes the statistical analysis of the characterization results for PVA–chitosan hydrogels formulated with different tannic acid concentrations as a crosslinking agent (F1, 1%; F2, 3%; and F3, 5%). Overall, increasing tannic acid concentration had a significant effect on several physicochemical properties of the hydrogels. Viscosity data measured at 12 rpm and 30 rpm were normally distributed and homogeneous; therefore, they were analyzed using one-way ANOVA ($p < 0.05$), indicating significant differences in viscosity among formulations. Viscosity increased with increasing tannic acid concentration (F3 > F2 > F1), which can be attributed to the formation of additional hydrogen bonds between the phenolic groups of tannic acid and the hydroxyl groups of PVA as well as the amine groups of chitosan, resulting in a denser hydrogel network. This mechanism is consistent with previous reports showing that tannic acid interacts via hydrogen bonding with polymers such as PVA and chitosan to form stable networks and enhance the mechanical properties of the material [6]. Can be seen in table 3.

For spreadability, the Kruskal–Wallis test showed no significant differences among formulations ($p > 0.05$). Nevertheless, descriptively, spreadability tended to decrease at higher tannic acid concentrations, which may be associated with increased viscosity that restricts material flow. Regarding adhesiveness, the analysis indicated a significant difference between F1 and F2, but not for F3, suggesting that increased crosslink density can improve adhesiveness up to a certain point before the network becomes excessively rigid. This finding aligns with evidence that tannic acid addition can increase mechanical modulus and adhesion in polymer networks through hydrogen bonding and other molecular interactions [12].

The swelling test demonstrated significant differences among formulations ($p < 0.05$), with swelling capacity decreasing as tannic acid concentration increased. This trend likely reflects progressive network densification, which reduces free volume and limits water penetration. The results agree with previous studies reporting that enhanced intermolecular interactions induced by tannic acid can reduce the water uptake of hydrogels [2]. In contrast, pH values did not differ significantly among formulations ($p > 0.05$), indicating that tannic acid addition up to 5% did not meaningfully alter the pH of the PVA–chitosan hydrogel system.

Characterization of Crosslinking by FTIR

Fourier transform infrared (FTIR) spectroscopy is based on the interaction between infrared radiation and the molecules in a sample. Incident IR radiation is absorbed by chemical bonds, inducing characteristic atomic vibrations, including stretching and bending modes. These vibrational modes are specific to particular functional groups because each type of chemical bond absorbs IR energy at distinct frequencies (or wavenumbers) depending on its molecular structure. Accordingly, the resulting FTIR spectrum displays absorption bands that can be used to identify functional groups and infer the molecular structure of the analyzed material [24]. This principle is grounded in the fact that bond vibrations influence the interaction with infrared radiation; therefore, changes in band intensity at specific regions indicate the presence of corresponding functional groups in the material. Can be seen in table 4 and Figure 1.

Table 4. Hasil Interpretasi Spektrum FTIR

Peak (cm^{-1}) F1	Peak (cm^{-1}) F2	Peak (cm^{-1}) F3	Interpretasi
3390.33	3388.72	3385.41	O–H stretching (PVA & tannic acid) overlaps with N–H stretching (chitosan), hydrogen bonding
2922.17	2921.84	2920.96	C–H stretching (aliphatic, $-CH_2$)
1644.03	1642.58	1641.77	C=O (Amide I of chitosan) / C=C aromatic (tannic acid)
1425.62	1423.91	1421.84	C–H bending /

			deformation –NH_3^+ (chitosan)
1349.71	1347.96	1345.88	C–O stretching (phenolic, tannic acid)
1250.18	1249.67	1248.42	C–O stretching (PVA & tannic acid)
1108.54	1105.92	1103.67	C–O–C stretching (PVA)

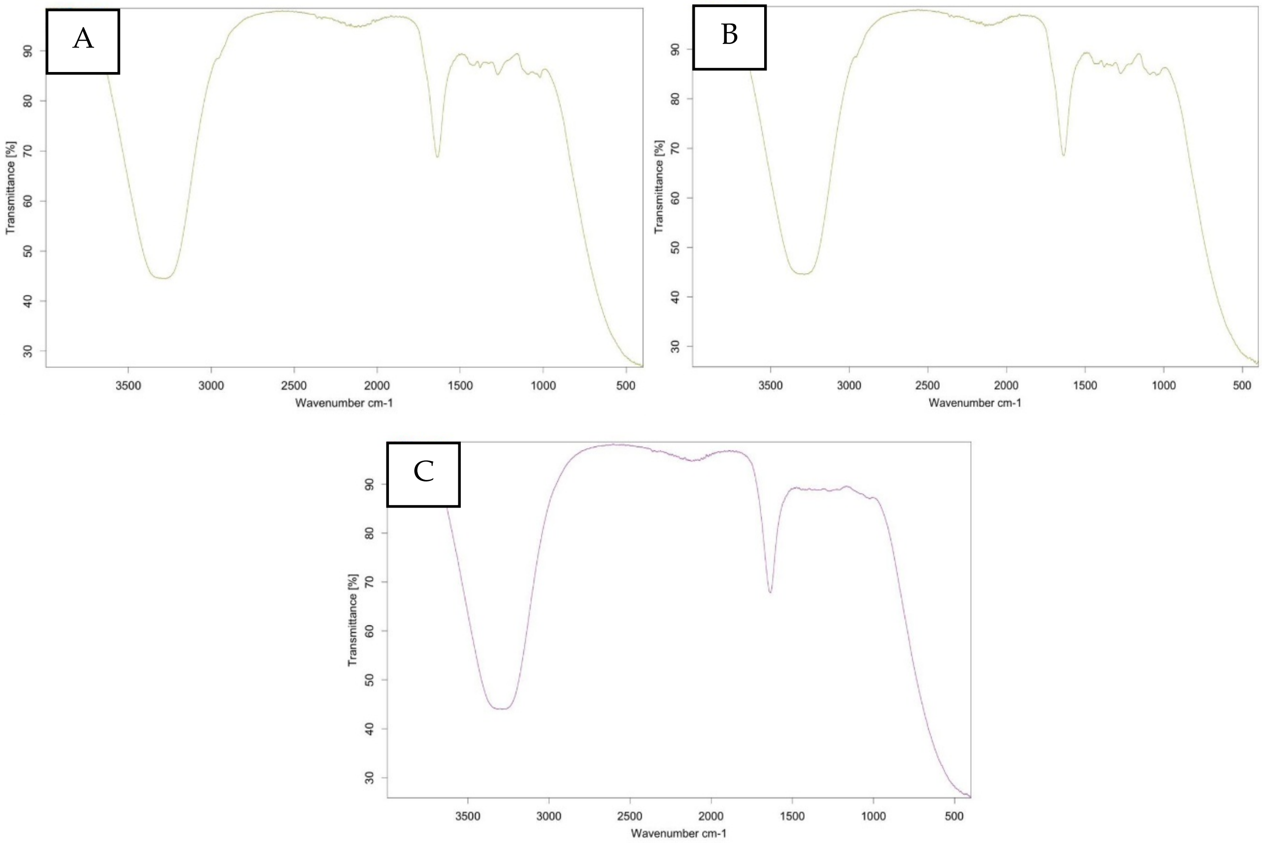

Figure 1. Spectrum of FTIR. A) Formula 1 (F1), B) Formula 2 (F2), and C) Formula 3 (F3)

Spektrum FTIR dari tiga formulasi hidrogel PVA–kitosan yang masing-masing The tannic acid-treated samples (A, B, and C in Figure 1), summarized in Table 4, exhibit characteristic changes in the absorption bands consistent with increased interactions between functional groups within the polymer network.

The broad absorption band appearing around 3390–3385 cm⁻¹ in all samples is attributed to the O–H stretching vibrations originating from the hydroxyl groups of PVA and the phenolic groups of tannic acid. This region also overlaps with the N–H stretching of chitosan, so the N–H band is not observed as a separate peak. This broadening of the O–H band indicates strong hydrogen bonding between the –OH groups of PVA, the phenolic –OH groups of tannic acid, and the –NH_2 groups of chitosan. These results align with reports that in the PVA–chitosan–tannic acid system, the broad O–H band often masks the N–H band due to intense intermolecular hydrogen bonding [6][25] .

The absorption peak in the 2922–2921 cm^{-1} region was consistently observed in all samples and was attributed to the aliphatic C–H stretching vibration ($-CH_2$) of the PVA chain. The presence of this peak indicates that the basic structure of PVA was maintained during the hydrogel formation process. This finding is consistent with studies reporting that the aliphatic C–H bands in PVA remained even when PVA interacted with chitosan and other binding agents [26].

The absorption in the 1644–1641 cm^{-1} region is associated with the C=O stretching vibration (Amide I) of chitosan which can overlap with the aromatic C=C vibration of tannic acid. No significant peak shift or emergence of new bands was observed in this region, indicating that the interactions occurring are physical in nature, not the formation of new covalent bonds. This result is consistent with reports stating that small shifts or changes in the intensity of the band around 1640 cm^{-1} in tannic acid-based hydrogels are indicative of hydrogen bonding interactions [11].

The absorption band in the 1425–1421 cm^{-1} region is associated with C–H bending and/or $-NH_3^+$ deformation of chitosan. The presence of this band indicates that some of the amine groups of chitosan are in the protonated form, which contributes to the formation of the hydrogel network through electrostatic interactions and hydrogen bonds. This phenomenon was also reported by researchers who stated that protonation of the amine groups of chitosan is common in polyphenol-based hydrogel systems [27].

Furthermore, the absorption band in the 1349–1345 cm^{-1} region is associated with the phenolic C–O stretching vibration of tannic acid, while the band at 1250–1248 cm^{-1} is associated with the C–O stretching vibration of PVA and tannic acid. The presence of these bands confirms the role of tannic acid as a physical crosslinker that interacts with PVA and chitosan through hydrogen bonds. This finding is in accordance with the results of research stating that tannic acid is able to form a hydrogel network through non-covalent interactions without changing the chemical structure of the main polymer [11].

The absorption peak in the 1108–1103 cm^{-1} region is associated with the C–O–C stretching vibration of the PVA structure. The presence of this band in all samples indicates that the primary structure of PVA remains stable after the hydrogel formulation process. A similar finding was reported by researchers who stated that the C–O–C band of PVA remained detectable in PVA–chitosan-based hydrogels [26].

In the low-wavenumber region of 523–519 cm^{-1}, absorption bands within the fingerprint region were observed, reflecting the specific characteristics of the resulting hydrogel structure. The similarity of the FTIR spectra of samples F1, F2, and F3 indicates that the formulation process produced a consistent and stable hydrogel.

Overall, the FTIR analysis results indicate that the PVA–chitosan–tannic acid hydrogel is formed through physical interactions in the form of hydrogen bonds without the formation of new functional groups. This indicates that tannic acid acts as a physical crosslinker, strengthening the hydrogel network without altering the basic chemical structure of PVA and chitosan.

4. Conclusion

Based on this study, it can be concluded that varying the concentration of tannic acid as a crosslinker significantly affects the physicochemical characteristics of PVA–chitosan hydrogels. Increasing tannic acid concentration led to marked changes in the physical properties of the hydrogels, including organoleptic characteristics, viscosity, spreadability, adhesiveness, and swelling index. At higher tannic acid concentrations, viscosity and swelling index tended to decrease, indicating the formation of a denser crosslinked network that limits the hydrogel's ability to absorb water. In contrast, spreadability and adhesiveness increased up to a certain concentration, suggesting a balance between gel flexibility and network strength. The pH of all formulations remained within the physiological skin pH range, supporting their suitability for topical application. In addition, the irritation test demonstrated that none of the formulations induced skin irritation. Therefore, tannic acid has potential as an effective and safe crosslinker for PVA–chitosan hydrogel preparation, with different tannic acid concentrations producing distinct physicochemical profiles. Selecting an appropriate tannic acid concentration is essential to obtain hydrogels with optimal properties tailored to the intended application.

Reference :

[1] Depkes RI. Farmakope Indonesia. Edisi VI. Jakarta: Departemen Kesehatan Republik Indonesia; 2020.

[2] Mahamoud MM, Ketema TM, Kuwahara Y, Takafuji M. Enhancement of mechanical properties of benign polyvinyl alcohol/agar hydrogel by crosslinking tannic acid and applying multiple freeze/thaw cycles. Gels. 2024;10(8):527. doi:10.3390/gels10080527.

[3] Zhou R. Multifunctional hydrogel based on polyvinyl alcohol/chitosan/metal polyphenols for facilitating acute and infected wound healing. Int J Biol Macromol. 2024;269:126–138.

[4] Bercea M. Recent advances in poly(vinyl alcohol)-based hydrogels. Polymers (Basel). 2024;16:1–36.

[5] Yılmaz BD, Aydemir Sezer U, Sarı A, Yıldız A, Sezer S. Formulation and evaluation of PVA-based composite hydrogels: physicochemical, leachables, and in vitro immunogenicity studies. J Mater Chem B. 2025;13(3):469–481.

[6] Jasrotia S, Gupta S, Kudipady ML, Puttaiahgowda YM. Development and characterization of chitosan–PVA–tannic acid film for extended shelf life and safety of food products. ACS Omega. 2025;10:19361–19378. doi:10.1021/acsomega.4c09964.

[7] Feng C. Toxicity and biocompatibility of glutaraldehyde crosslinked hydrogels. Int J Biol Macromol. 2021;180:483–491.

[8] Sahiner N. Effect of natural crosslinker concentration on swelling and mechanical properties of hydrogels. Mater Today Commun. 2023;36:106007.

[9] Zhao H. Tannic acid as a versatile natural crosslinker and antioxidant in biomedical applications. Front Chem. 2022;10:942665.

[10] Fan J, Yang H, Yang J, Peng M, Hu. Tannic acid-based multifunctional materials: a review. RSC Adv. 2020;10(14):8060–8073.

[11] Karakuş NR, Türk S, Guney Eskiler G, Syzdykbayev M, Appazov NO, Özacar M.

Investigation of tannic acid crosslinked PVA/PEI-based hydrogels as potential wound dressings with self-healing and high antibacterial properties. Gels. 2024;10(11):682. doi:10.3390/gels10110682.

[12] Yu X, Huang J, Wu C, Zhang W. Biocompatible autonomous self-healing PVA-CS/TA hydrogels based on hydrogen bonding and electrostatic interaction. Sci Rep. 2025;15(1). doi:10.1038/s41598-025-85298-3.

[13] Nurrahmi F, Siregar YI, Fransiska D. Production of basic material carrageenan hydrogel using polymer based polyvinyl alcohol (PVA). Mar Sci Dep. 2017;(1):1–10.

[14] Yuandari E, Rahma RTA. Metodologi Penelitian dan Statistik. Jakarta: In Media; 2017.

[15] Araújo F, et al. Effect of chitosan properties and dissolution state on solution rheology and film performance in triboelectric nanogenerators. Gels. 2025;11:523:1–19.

[16] Zatalini DF, Hendradi E, Drake P, Sari R. The effect of chitosan and polyvinyl alcohol combination on physical characteristics and mechanical properties of chitosan-PVA-Aloe vera film. J Farm Dan Ilmu Kefarmasian Indones. 2023;10(2):151–161. doi:10.20473/jfiki.v10i22023.151-161.

[17] Chen C, Yang H, Yang X, Ma Q. Tannic acid: a crosslinker leading to versatile functional polymeric networks: a review. RSC Adv. 2022;12:7689–7711. doi:10.1039/d1ra07657d.

[18] Werawati A, et al. Sediaan hidrogel ekstrak etanol daun kelor. Edu Masda J. 2024;8(2):119–130.

[19] Sa’diyah A, Pambudi DB. Evaluasi sediaan masker hidrogel ekstrak etanol daun mangrove merah (Sonneratia caseolaris Engl.). Benzena Pharm Sci J. 2024;3(1):62–71.

[20] Wahidah S, Ayu G, Saputri R. Formulasi dan uji stabilitas sediaan gel ekstrak etanol daun asam jawa (Tamarindus indica L.) dengan variasi gelling agent. J Mandala Pharmacon Indones. 2024;10(2):508–518.

[21] Saarai A, Kasparkova V, Sedlacek T, Saha P. On the development and characterisation of crosslinked sodium alginate/gelatine hydrogels. J Mech Behav Biomed Mater. 2013;18:152–166. doi:10.1016/j.jmbbm.2012.11.010.

[22] Diao Z, Li L, Zhou H, Yang L. Tannic acid and silicate-functionalized poly(vinyl alcohol)–hyaluronic acid hydrogel for infected diabetic. Regen Biomater. 2024;11:1–13.

[23] Chen Y, Etxabide A, Seyfoddin A, Ramezani M. Fabrication and characterisation of poly(vinyl alcohol)/chitosan scaffolds for tissue engineering applications. Mater Today Proc. 2023. doi:10.1016/j.matpr.2023.02.303.

[24] Gong Y, Chen X, Wu W. Advances in sample preparation: application of Fourier transform infrared (FTIR) spectroscopy in sample preparation: material characterization and mechanism investigation. Adv Sample Prep. 2024;11.

[25] Zou X, Huang J. Hydrogen bond-regulated rapid prototyping and performance optimization of poly(vinyl alcohol)–tannic acid hydrogels. Gels. 2025;11:602.

[26] Hong F, Qiu P, Wang Y, Ren P, Liu J, Zhao J, et al. Food Chemistry : X Chitosan-based hydrogels : From preparation to applications , a review. Food Chem X [Internet]. 2024;21(8326):101095. https://doi.org/10.1016/j.fochx.2023.101095

[27] Alfuraydi RT, Al-Harby NF, Mohamed NA, Alminderej FM, Elmehbad NY. Poly (Vinyl Alcohol) Hydrogels Boosted with Cross-Linked Chitosan and Silver Nanoparticles for Efficient Adsorption of. gels. 2023;9(882):1–25.

[28] Liu R, Zhang J, Zhang T. Tannic acid–chitosan–PVA hydrogels with improved mechanical and antibacterial properties. Carbohydr Polym. 2020;247:116689.