

Araştırma Makalesi / Research Article

BIO-BASED ALGINATE/LIGNIN/CHITOSAN HYBRID HYDROGEL COMPOSITES INCORPORATING ZNO FOR MULTIFUNCTIONAL PLA NONWOVEN FABRICS

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ABSTRACT: The growing demand for sustainable and bio-based materials has accelerated the development of multifunctional hydrogel systems potential for biomedical applications. In this study, hydrogel–textile composites based on alginate, lignin, chitosan, and zinc oxide (ZnO) were synthesized via a sol–gel approach at two alginate concentrations (0.5% and 1%) and applied as functional coatings onto polylactic acid (PLA) nonwoven fabrics. The coated fabrics were systematically characterized by FTIR spectroscopy, SEM/EDS, and tensile testing, while air permeability, water absorption, and antibacterial performance against *Escherichia coli* and *Staphylococcus aureus* were evaluated. The hydrogel coatings enhanced the surface characteristics and antibacterial performance of the PLA nonwoven substrates while maintaining their mechanical integrity. FTIR analysis suggested possible intermolecular interactions among the biopolymer components, particularly in the high-alginate SA/L/CS/ZnO (Y3) formulation. SEM observations indicated the formation of a hydrogel coating on the examined fabric regions, while EDS analysis qualitatively confirmed the presence of ZnO. The coated fabrics exhibited antibacterial efficiencies exceeding 98% together with improved tensile properties. Increasing sodium alginate content also enhanced water absorption, although a reduction in air permeability was observed. The developed bio-based hydrogel–textile composites exhibited promising mechanical, antibacterial, and fluid-management properties, supporting their potential for further investigation in sustainable medical textile applications.

Keywords: PLA nonwoven fabric, Hydrogel, Hybrid composites, Zinc oxide nanoparticles

ÇOK FONKSİYONLU PLA NONWOVEN KUMAŞLAR İÇİN ZNO İÇEREN BİYO-BAZLI ALJİNAT/LİGNİN/KİTOSAN HİBRİT HİDROJEL KOMPOZİTLER

ÖZ: Sürdürülebilir ve biyo-bazlı malzemelere olan artan talep, biyomedikal uygulamalar için çok fonksiyonlu hidrojel sistemlerinin geliştirilmesini hızlandırmıştır. Bu çalışmada, aljinat, lignin, kitosan ve çinko oksit (ZnO) esaslı hidrojel–tekstil kompozitleri, iki farklı aljinat konsantrasyonunda (%0,5 ve %1) sol–jel yöntemi ile sentezlenmiş ve polilaktik asit (PLA) nonwoven kumaşlar üzerine fonksiyonel kaplama olarak uygulanmıştır. Kaplanmış kumaşlar FTIR spektroskopisi, SEM/EDS ve çekme testleri ile sistematik olarak karakterize edilmiş; ayrıca hava geçirgenliği, su emme kapasitesi ve *Escherichia coli* ile *Staphylococcus aureus* bakterilerine karşı antibakteriyel performansları değerlendirilmiştir. Hidrojel kaplamalar, PLA dokusuz yüzeylerin mekanik bütünlüğünü korurken yüzey özelliklerini ve antibakteriyel performansını geliştirmiştir. FTIR analizi, özellikle yüksek aljinat içeriğine sahip SA/L/CS/ZnO (Y3) formülasyonunda biyopolimer bileşenleri arasında olası moleküller arası etkileşimlere işaret etmiştir. SEM gözlemleri, incelenen kumaş bölgelerinde bir hidrojel kaplamasının oluştuğunu göstermiş, EDS analizi ise ZnO varlığını nitel olarak doğrulamıştır. Kaplanan kumaşlar, geliştirilmiş çekme özellikleri ile %98'in üzerinde antibakteriyel etkinlik sergilemiştir. Sodyum aljinat miktarındaki artış su emme kapasitesini artırırken, hava geçirgenliğinde bir azalma gözlemlenmiştir. Geliştirilen biyo-bazlı hidrojel–tekstil kompozitleri, umut verici mekanik, antibakteriyel ve sıvı yönetimi özellikleri göstermiş olup, sürdürülebilir medikal tekstil uygulamalarındaki potansiyellerinin daha ileri araştırmalarla değerlendirilmesini desteklemektedir.

Anahtar Kelimeler: PLA Nonwoven Kumaşlar, Hidrojel, Hibrit kompozitler, Çinko oksit nanopartikülleri

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1. INTRODUCTION

Wound dressings play a critical role in protecting damaged tissue and promoting the healing process. Conventional gauze dressings can absorb wound exudate; however, they often require frequent replacement, may cause pain during removal, and provide limited fluid management. An ideal wound dressing should therefore be biocompatible, non-toxic, highly absorbent, antimicrobial, and capable of maintaining a moist environment that supports drug delivery and cell proliferation. In recent years, advanced wound dressings based on synthetic, natural, and hybrid polymers have been developed in various forms, including films, foams, nanofibers, hydrogels, and other bioactive materials [1]. Among these systems, hydrogels have attracted considerable attention because of their ability to retain moisture, absorb wound exudate, protect against infection, and support tissue regeneration [2].

Hydrogels are three-dimensional cross-linked polymer networks capable of absorbing large amounts of water while maintaining structural integrity. They can be prepared from natural polymers such as chitosan (CS), alginate (AL), and collagen, as well as synthetic polymers including poly(acrylic acid) (PAA), polyethylene glycol (PEG), and polyvinyl alcohol (PVA) [2]. Depending on the crosslinking mechanism, hydrogels can be classified as physically crosslinked systems, formed through interactions such as freeze–thaw cycles, hydrogen bonding, or ionic interactions, or chemically crosslinked networks produced via covalent bonding through grafting, polymerization, irradiation, enzymatic reactions, photo-crosslinking, or polyelectrolyte complexation [3, 4]. Natural polymers such as CS and AL are particularly valuable in wound care. They mimic the extracellular matrix (ECM), support controlled drug release, absorb exudate, and allow gas exchange, enhancing healing and patient comfort [5, 6]. AL, a marine-derived polysaccharide obtained from brown seaweed, is widely used in biomedical materials due to its biocompatibility, biodegradability, and ability to form hydrogels through ionic crosslinking with divalent cations [7]. CS, produced by the deacetylation of chitin, is a cationic polysaccharide known for its biodegradability, biocompatibility, and inherent antimicrobial activity. Under acidic wound conditions, protonation of its $-NH_2$ groups enhances antibacterial activity, making CS particularly suitable for biomedical applications [8]. The combination of AL and CS has attracted considerable attention because these oppositely charged polymers form polyelectrolyte complexes that enhance the structural stability and functional performance of hydrogel systems. These interactions improve water retention, oxygen permeability, and drug-delivery capacity, thereby supporting the wound-healing process [9]. Sodium alginate (SA), one of the most widely used AL derivatives, is frequently employed in wound dressings due to its extracellular matrix (ECM)-like structure, moisture-retention ability, and biodegradability. However, SA alone may promote bacterial proliferation, which limits its effectiveness; therefore, it is often combined with antibacterial polymers such as CS, a linear

polysaccharide recognized as generally safe (GRAS) by the U.S. Food and Drug Administration (FDA) [10, 11].

Recent studies have focused on the development of next-generation hydrogels with smart, responsive, and multifunctional properties. These materials are designed not only to ensure biocompatibility but also to provide appropriate mechanical strength, flexibility, and gas permeability for effective wound management [4]. To further improve their performance, composite hydrogels incorporating bioactive or reinforcing components have been widely investigated. Among these, lignin has recently gained significant attention as a sustainable and multifunctional biopolymer. Lignin is a major structural component of plant cell walls and the second most abundant natural biopolymer after cellulose, accounting for approximately 15–35% of lignocellulosic biomass [12–13]. Lignin is a three-dimensional aromatic biopolymer containing phenolic and aliphatic hydroxyl groups that enable strong intermolecular interactions with polysaccharides such as AL and CS. In lignocellulosic-based hydrogels, lignin can act as a reinforcing component by forming hydrogen bonds and rigid microdomains within the polymer network, thereby improving mechanical strength. Owing to its aromatic structure and diverse functional groups, lignin and its nanoscale derivatives, such as lignin nanoparticles (LNPs), have been investigated for antioxidant systems and biomedical applications [12]. Lignin shows strong compatibility with polysaccharide matrices such as AL, enabling the formation of reinforced AL-based materials through hydrogen bonding interactions [14–17]. The integration of CS into lignin-containing systems can further enhance antibacterial performance against both Gram-positive bacteria (e.g., *Bacillus subtilis* and *Staphylococcus aureus*) and Gram-negative bacteria (e.g., *Pseudomonas aeruginosa* and *Escherichia coli*) [13]. Additionally, the functionality of polysaccharide-based hydrogels can be further improved through the incorporation of metal or metal-oxide nanoparticles such as Ag, Au, ZnO, or CuO [18]. Among these, ZnO nanoparticles are particularly attractive because they are FDA-approved, non-toxic, and capable of providing antibacterial and ultraviolet (UV) protective effects [11]. Numerous studies have reported that AL/CS polyelectrolyte complex hydrogels exhibit improved mechanical strength and antibacterial performance [3,9–11], while lignin incorporation enhances antioxidant activity and structural stability [12–17]. In textile-related applications, AL/CS-based coatings have been shown to improve moisture management and biocompatibility, while ZnO nanoparticles provide enhanced antibacterial activity and multifunctional surface properties [19–21]. Therefore, the synergistic integration of lignin, CS, and ZnO nanoparticles within hybrid hydrogel composite hydrogels represents a promising strategy for developing sustainable multifunctional textile materials with enhanced mechanical strength and bioactivity.

Several recent studies have investigated hydrogel-based composite systems for biomedical and functional textile applications. Zhang et al. [10] fabricated an antibacterial

composite hydrogel composed of aldehyde-modified SA, CS oligosaccharide (COS), and ZnO nanoparticles via spontaneous Schiff-base formation. The resulting hydrogel promoted cell proliferation and accelerated wound closure in both in vitro and in vivo models. Similarly, Mohandas et al. [9] prepared porous AL/ZnO hydrogels using a freeze-drying technique to produce bandage-like scaffolds with high porosity (60–70%) and tunable swelling behavior, demonstrating strong antibacterial activity. Ahmad et al. [22] developed a biodegradable nonwoven composite derived from cotton waste coated with an AL/CS hydrogel, highlighting the feasibility of integrating hydrogel systems with textile substrates to provide moisture control and biodegradable functionality. In another study, Türkoğlu et al. [23] produced textile-hydrogel hybrid dressings using SA/CS hydrogels crosslinked with poly(ethylene glycol) diglycidyl ether (PEGDGE) and applied them onto viscose and Tencel nonwoven substrates. Bakil et al. [24] fabricated SA-based ZnO nanoparticle films via solvent casting, demonstrating improved antibacterial activity and surface properties. More recently, Hajishirki et al. [25] developed a CS/carbon dots/SA/ZnO double-layer sponge hydrogel, which exhibited enhanced antibacterial, antioxidant, and hemostatic properties. Biodegradable textiles such as polylactic acid (PLA) often suffer from low hydrophilicity and limited intrinsic functionality, which restricts their performance in biomedical applications. Therefore, surface modification strategies are required to enhance moisture management, antibacterial activity, and overall functionality [26]. Higher SA concentrations may lead to crack formation within the hydrogel structure [27–29]. Natural polymer-based hydrogels, particularly those combining the cationic polysaccharide CS and anionic alginate, provide a biocompatible and hydrophilic three-dimensional network through ionic crosslinking via electrostatic interactions between NH_3^+ groups of CS and COO^- groups of SA [30]. The incorporation of ZnO nanoparticles into such hydrogel systems has also been successfully demonstrated, where ZnO particles were effectively embedded and stabilized within the polymer network without compromising biocompatibility, while simultaneously providing antibacterial and photocatalytic functionality [31]. For adhesion onto textile substrates such as PLA nonwoven fabrics, the natural adhesive characteristics of CS can play a significant role, since protonated amine groups ($-\text{NH}_3^+$) are capable of forming electrostatic and hydrogen-bond interactions with negatively charged or polar surface groups [30]. Furthermore, designing hybrid hydrogels with interpenetrating polymer networks, such as CS-SA systems combined with phenolic or partially hydrophobic components like lignin, can enhance network cohesion, mechanical stability, and compatibility with both hydrophilic and hydrophobic domains [30, 32, 33]. In PLA-based systems, ester carbonyl groups may additionally interact with polar functional groups of SA, lignin, and ZnO through dipole-dipole interactions, contributing to the stabilization and adhesion of the hydrogel coating on the PLA surface [34, 35]. Therefore, hybrid hydrogel composites can provide stable and adhesive multifunctional coatings on PLA

nonwoven fabrics through a combination of hydrogen bonding, dipole-dipole interactions between PLA ester groups and hydrogel functional groups, as well as possible ion-dipole interactions involving ZnO and protonated CS groups [36].

To the best of our knowledge, no previous study has investigated lignin/CS/ZnO-containing hybrid hydrogel composites for the development of multifunctional medical textile materials using PLA nonwoven fabrics as the substrate. In this study, multifunctional SA/L/CS/ZnO hybrid hydrogel coatings were developed and applied onto PLA spunlace nonwoven fabrics via a sol-gel approach. The novelty of the work lies in the synergistic integration of these components within a hybrid hydrogel network to simultaneously enhance moisture management, antibacterial activity, and mechanical performance of biodegradable textile materials. The coated fabrics were characterized by tensile strength analysis, antibacterial testing, air permeability measurements, FTIR spectroscopy, and SEM imaging to evaluate coating morphology and hydrogel distribution.

2. MATERIAL AND METHOD

2.1 Material

In this study, PLA spunlace nonwoven fabric (Mogul Tekstil, Türkiye) with a weight of 50 g/m² was used. Composite hybrid hydrogels were prepared on the PLA fabric using SA (alginic acid sodium salt from brown algae, Merck, Germany; ~200 cP at 1% w/v in water at 20 °C; ~70% guluronic acid, ~30% mannuronic acid; MW ~200,000 g/mol), calcium chloride ($\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$, 99%, Kimyalab, Türkiye), alkali lignin ($25 \pm 5 \mu\text{m}$, Merck, Germany), ZnO nanopowder (<100 nm, Merck, Germany), low molecular weight CS (50–190 kDa, 75–85% deacetylation, Cat. No. 448869), and glacial acetic acid (99%, Kimyalab, Türkiye). Sodium chloride (NaCl, Kimyalab, Türkiye) and calcium chloride dihydrate ($\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$, Kimyalab, Türkiye) were also used to prepare solutions for evaluating the wound exudate absorption of PLA nonwoven and hybrid hydrogel composites.

2.2 Method

2.2.1 Preparation of Hybrid Hydrogel Composites and Their Applications on PLA Nonwoven Fabric

The hybrid hydrogel composite fabrics were prepared as illustrated in Figure 1. Higher SA concentrations can cause cracks in the hydrogel [27–29]; therefore, two concentrations of alginic acid sodium salt (0.5% and 1%) were dissolved in deionized water and stirred at 750 rpm for 24 h to achieve homogeneous solutions. Lignin, a natural polymer with poor water solubility, was modified using CHPTMAC, containing a positively charged quaternary ammonium group $[\text{NR}_4]^+$, to produce quaternized alkali lignin (QAL) as reported in a previous study [26]. CS (3 g) was dissolved in 100 mL of 1 M acetic acid under continuous stirring for 4–5 h. AL solutions (0.5% and 1%, 100 mL) were then added separately to the CS solution and stirred for 4 h to obtain a uniform blend. Subsequently, 100 mL of 1% quaternized alkali lignin (QAL) solution and 100 mL of a 2% ZnO dispersion were incorporated

into the mixture. The ZnO dispersion was prepared by dispersing ZnO powder in deionized water under mechanical stirring at 750 rpm for 2 h at room temperature. After adding the QAL solution and ZnO dispersion, the mixture was mechanically stirred at 600 rpm for 3 h to obtain the hybrid hydrogel composite. A 0.25 M CaCl_2 solution was prepared as the gelation medium and stirred at 750 rpm for 20 min. PLA spunlace nonwoven fabrics (5×5 cm) were soaked in the SA/L/CS/ZnO hybrid hydrogel for 20 min at room temperature to allow full absorption. The wet pick-up of the impregnated fabrics was not quantitatively determined in this study. After immersion in the SA/L/CS/ZnO hybrid hydrogel for 20 min, the samples were allowed to drain naturally for approximately 5–10 s to remove excess hydrogel before being

transferred to the CaCl_2 gelation bath. Therefore, the dry hydrogel add-on reported in this study reflects the amount of coating retained after drying rather than the amount of hydrogel absorbed immediately after impregnation. The fabrics were then immersed in the CaCl_2 solution for 20 min to trigger gelation, during which Ca^{2+} ions diffused into the system, forming the hybrid hydrogel composite. Finally, the treated fabrics (Figure 1c) were air-dried at room temperature for 24 h and subsequently thermally treated at 50°C for 1 h to remove residual moisture and stabilize the hydrogel coating. A schematic representation of the hybrid hydrogel composites on PLA nonwoven fabrics is shown in Figure 1 a). The structure of hybrid hydrogel composites on PLA nonwoven fabrics is shown in Figure 1b).

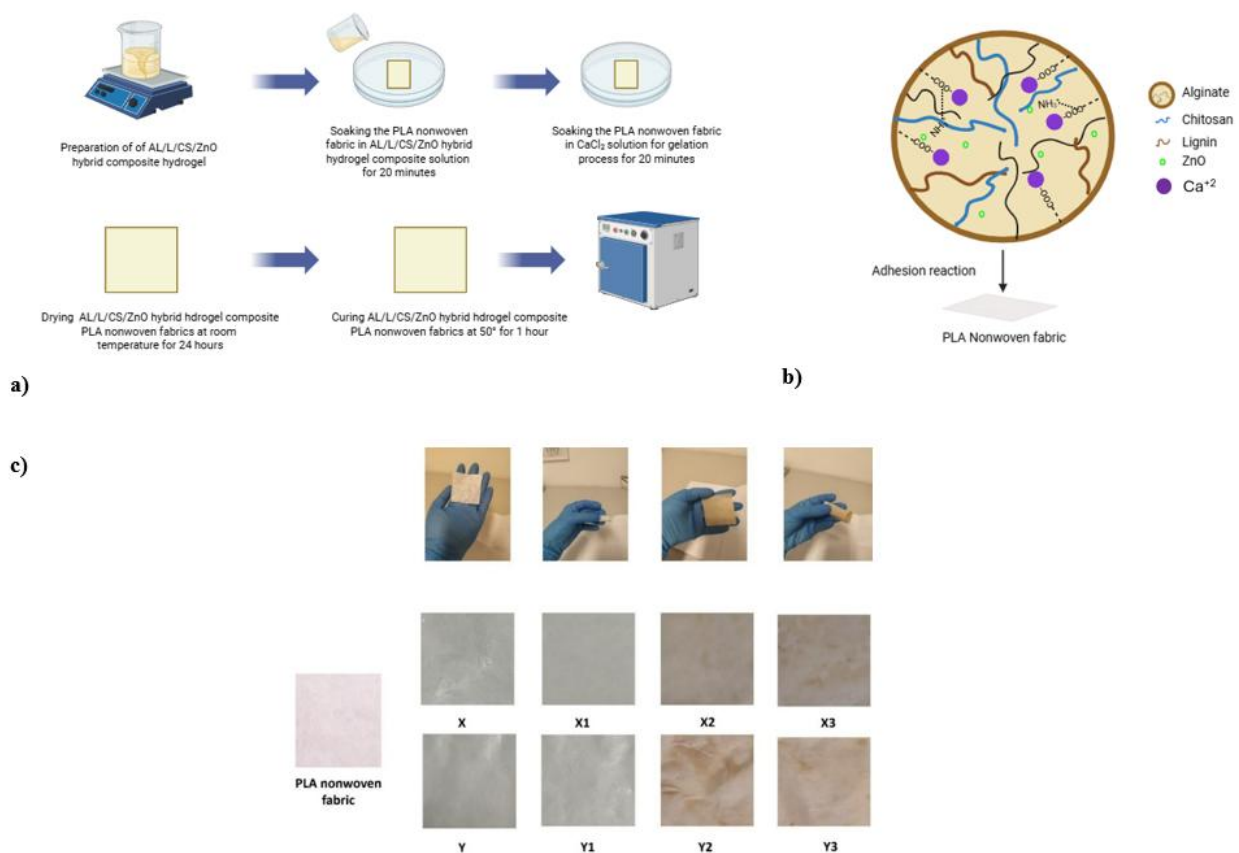


Figure 1. a) Schematic illustration of the preparation process, b) structure of hybrid hydrogel composites on PLA nonwoven fabrics, c) Photographic images of untreated PLA fabric, and hybrid hydrogel composite fabric samples

Table 1 AL, lignin, CS, and ZnO components used for the preparation of hybrid hydrogel composites on PLA nonwoven fabric samples

Sample Code	SA (%)	L (%)	CS (%)	ZnO (%)
X (SA)	0.5	-	-	-
X1 (SA / ZnO)	0.5	-	-	2
X2 (SA / L / ZnO)	0.5	1	-	2
X3 (SA / L / CS / ZnO)	0.5	1	2	2
Y (SA)	1	-	-	-
Y1 (SA / ZnO)	1	-	-	2
Y2 (SA / L / ZnO)	1	1	-	2
Y3 (SA / L / CS / ZnO)	1	1	2	2

2.2.2 Fourier Transform - Infrared (FT-IR) Spectroscopy

Hybrid hydrogel composite fabric samples were examined by Fourier transform infrared (FTIR) spectroscopy in transmission mode using an FT/IR-6X type A FT-IR spectrometer (Jasco, Tokyo, Japan) over the wavenumber range of 4000–400 cm^{-1} with a spectral resolution of 4 cm^{-1} .

2.2.3 Scanning Electron Microscopy (SEM)

The surface morphology of untreated PLA and hybrid hydrogel-coated fabric samples (1 × 1 cm) was examined using a field-emission scanning electron microscope (FE-SEM, Hitachi Regulus 8230) equipped with an energy-dispersive X-ray spectroscopy (EDS) detector. SEM imaging was performed at an accelerating voltage of 10.0 kV with a working distance of 8.7 mm. Prior to analysis, the samples were sputter-coated with a thin layer of gold using a Leica EM ACE600 coating system at a deposition rate of 0–10 nm/min.

2.2.4 Tensile Strength Measurements

Tensile strength measurements of untreated PLA and hybrid hydrogel composite fabric samples (20 cm × 5 cm) were conducted utilizing a universal tensile testing machine (Shimadzu AGS-X, Japan) according to the standard of TS 2008 EN ISO 1421:2000. All measurements were performed using five replicates (n=5), and the results are reported as mean ± standard deviation. The measurements were made with a testing speed of 100 mm/min, pre-tension maintained at 2 N, and a distance of 10 cm between the jaws. Tensile measurements were carried out along the machine direction (MD) of the PLA nonwoven fabrics, as all samples were prepared from the same nonwoven substrate and evaluated under identical testing conditions to enable a consistent comparison of the effect of the hydrogel coatings. Statistical analyses were performed using Minitab statistical software. One-way ANOVA followed by Tukey's HSD post hoc test was used to evaluate differences in tensile strength and elongation at break among the formulations. Results are presented as mean ± SD (n=5), and statistical significance was accepted at $p < 0.05$.

2.2.5 Air Permeability Measurements

Air permeability measurements were conducted on both untreated PLA nonwovens and hybrid hydrogel composite PLA nonwovens using an air permeability tester (Textest/FX 3300, Switzerland), in accordance with the TS 391 EN ISO 9237 standard. Three replicate measurements were performed for each sample, and the mean values were reported. Air permeability measurements were carried out on dry samples under ambient laboratory conditions. This property was defined as the amount of air, in liters (L), that passes through one square meter of fabric per second under a pressure equivalent to a 10 mm water column. During the test, a pressure increase of 100 Pa was applied to each sample, and the corresponding values were recorded in $\text{l/m}^2/\text{s}$. The air permeability ratio (R) was then determined using Equation (1) [26]. In this context, “qv” refers to the average vertical airflow volume passing

through the test area, measured in liters per second (L/s), while “A” indicates the size of the test area, which is 20 cm^2 .

$$R = qv/A \quad (1)$$

2.2.6 Fluid Absorbency Capacity

The wound exudate absorption capacity of both neat PLA nonwoven and hybrid hydrogel composite fabrics was evaluated in accordance with EN 13726-1:2002 standard. All measurements were performed in triplicate (n=3), and the results are reported as mean ± standard deviation. A simulated wound exudate solution was prepared by dissolving 2.298 g of sodium chloride (NaCl) and 0.368 g of calcium chloride dihydrate ($\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$) in 1 L of water. The solution was then heated to 37 °C to mimic the physiological temperature, ensuring it matched the ionic composition of actual wound exudate [29]. To evaluate the liquid absorption capacity of the hydrogels, the specimens were accurately cut into 5 cm × 5 cm squares. Each dry sample was first weighed and recorded as (W_i) before being individually transferred into petri dishes. The pre-prepared simulated wound exudate solution was applied to the samples, which were subsequently incubated at 37 °C for 30 minutes to promote equilibrium absorption under standardized conditions. Upon completion of the incubation period, each sample was suspended for 30 seconds to allow for the removal of unbound excess fluid. The final step involved measuring the weight of the specimens (W_f), after which the fluid absorbency (%) of the hydrogels was determined using the following Equation (2):

$$\text{Fluid Absorbency (\%)} = (W_f - W_i) / W_i \times 100 \quad (2)$$

2.2.7 Antibacterial activity analysis

The antibacterial activity of hybrid hydrogel composite PLA nonwoven fabrics was evaluated quantitatively according to the AATCC 100 standard (Antibacterial Finishes on Textile Materials). Antibacterial measurements were performed using five independent replicates, and the results are presented as mean values. Cultures of *Staphylococcus aureus* (ATCC 6538) and *Escherichia coli* (ATCC 25922) were prepared and inoculated onto sterile 1 × 1 cm^2 fabric specimens placed in sterile capped tubes to ensure sufficient contact between the bacterial suspension and the fabric surface. Following incubation under controlled conditions, the bacterial suspensions were serially diluted using phosphate buffer, plated onto Mueller–Hinton agar, and incubated at 35 °C for 24 h. After incubation, surviving bacterial colonies were counted, and the percentage reduction in viable bacterial counts was calculated using the following Equation (3):

$$\text{Antibacterial efficiency (\%)} = \frac{R_f - R_s}{R_f} \times 100 \quad (3)$$

2.2.8 Degradation Process of Degradable Hybrid Hydrogel Composites

The degradation behavior of hybrid hydrogel composite fabric samples was evaluated at room temperature under alkaline (0.5 M NaOH) and acidic (HCl solution adjusted to pH 5) conditions.

Samples were cut into dimensions of 2×2 cm and placed in Petri dishes. Approximately 20–30 mL of the respective solutions was added to ensure complete immersion. The samples were maintained under static conditions and monitored daily. Changes in physical appearance and structural integrity were recorded throughout the degradation period.

3. RESULTS AND DISCUSSION

3.1 FT-IR Analysis Results

To investigate the physical interactions between the PLA fabric and the hybrid hydrogel composite, which may influence the stretching and bending vibrations of molecular bonds, FT-IR spectra of the PLA nonwoven fabric samples were recorded. FT-IR spectra of the neat PLA fabric are given in Figure 2. FT-IR spectra of X and Y samples, containing 0.5% and 1% SA concentrations, are shown in Figure 3. FT-IR spectra of the coated PLA nonwoven fabrics exhibited characteristic absorption bands associated with both PLA and hydrogel constituents. The characteristic PLA bands were observed at approximately 1750 cm^{-1} (C=O stretching) and 1080 cm^{-1} (C–O–C stretching), consistent with previous studies [37]. With the incorporation of SA, a broad O–H stretching band appeared around 3400 cm^{-1} , along with asymmetric carboxylate (COO^-) bands near $\sim 1600 \text{ cm}^{-1}$, indicating the presence of hydroxyl and carboxyl functional groups. An increase in SA content from 0.5% (X samples) to 1.0%

(Y samples) resulted in more pronounced band intensities, which may be associated with increased hydrophilicity of the coated fabrics [38–41]. The presence of ZnO was supported by Zn–O stretching vibrations in the $500\text{--}600 \text{ cm}^{-1}$ region and a broad Zn–OH band within $3000\text{--}3700 \text{ cm}^{-1}$ (Figure 3). In addition, slight shifts in the COO^- bands (e.g., from $\sim 1618 \text{ cm}^{-1}$ to $\sim 1605\text{--}1595 \text{ cm}^{-1}$, depending on formulation) suggest possible coordination interactions between Zn^{2+} ions and carboxylate groups within the hydrogel network. The formation of the SA–ZnO composite hydrogel on the PLA surface was further supported by changes observed in the carbonyl region after crosslinking. Moreover, slight variations in the intensity of the characteristic PLA bands can be attributed to the presence of the hydrogel coating, which may partially attenuate the PLA signals depending on the amount of deposited hydrogel [42–45]. Furthermore, the presence of Ca^{2+} ions used for alginate crosslinking may also influence the COO^- band positions. According to the “egg-box” model, Ca^{2+} ions interact with the carboxylate groups of alginate, forming ionic coordination between adjacent chains, which can modify the local chemical environment and lead to shifts in FTIR band positions [46, 47]. FTIR spectra of lignin-containing samples exhibited additional bands at approximately $\sim 1524 \text{ cm}^{-1}$ (aromatic C=C stretching) and $\sim 1192 \text{ cm}^{-1}$ (phenolic C–O stretching), which are consistent with the characteristic vibrations of lignin reported in the literature and therefore support its successful incorporation into the coating [48–49].

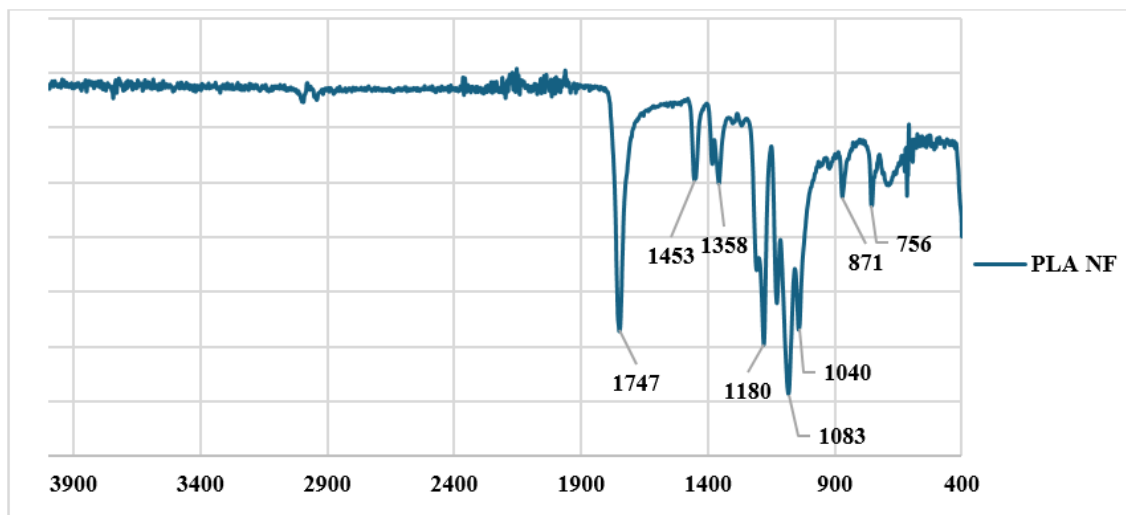


Figure 2. FTIR spectra of untreated PLA nonwoven fabric

Table 2. FTIR peak assignment and shifts

Band	Assignment	PLA (cm^{-1})	Composite (cm^{-1})	Shift	Comment	References
$\sim 3421 \rightarrow 3325$	O–H stretching	3421	3335	–96	Possible H-bonding	[19, 20]
~ 1750	C=O stretching	1747	1751	–4	Weak interaction	[36]
~ 1600	COO^- asymmetric	1618	1605	–13	Zn^{2+} / Ca^{2+} coordination	[45, 46]
500–600	Zn–O	-	604–665	–	ZnO presence	[42, 44]
~ 1550	Amide II (CS)	-	1545	–	CS incorporation	[20]

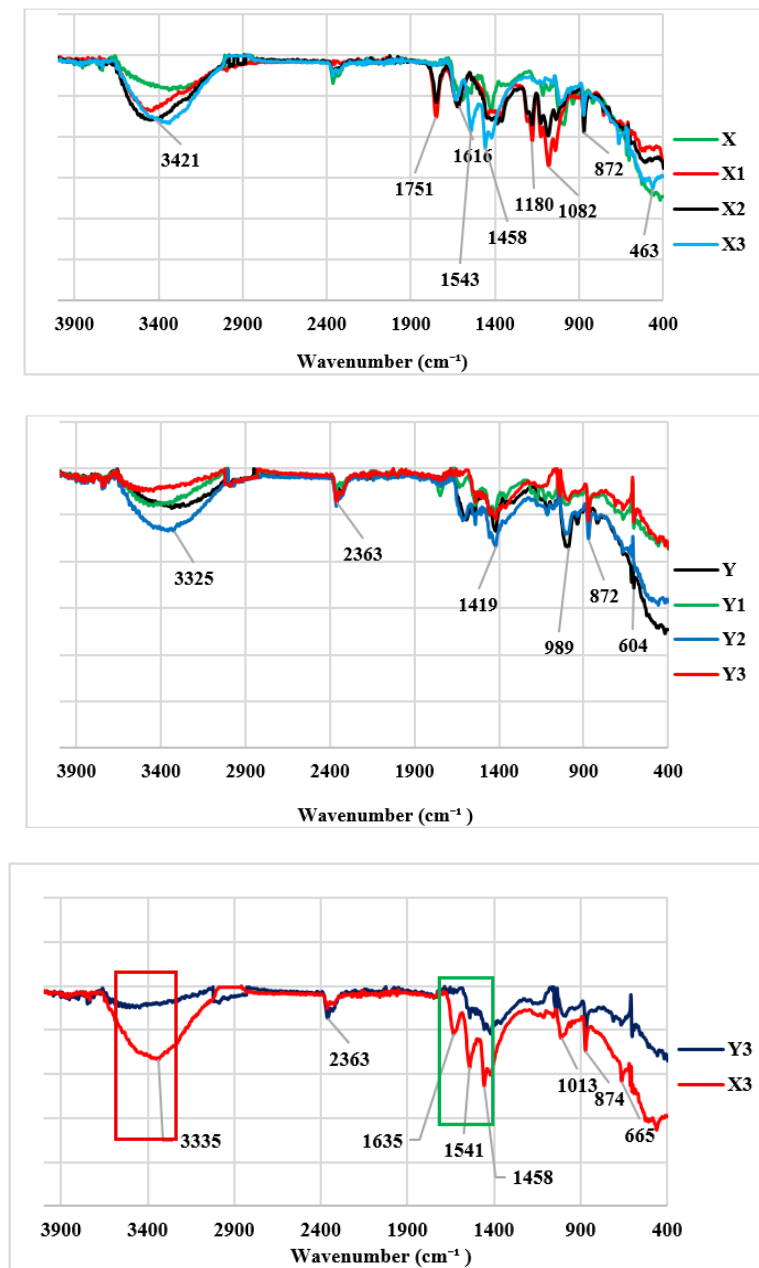


Figure 3. FTIR spectra of untreated PLA and PLA nonwoven conductive hydrogel composite fabric samples

CS-containing samples (X3, Y3) exhibited amide II bands (~ 1550 cm^{-1}), attributed to N–H bending and C–N stretching vibrations. These were accompanied by broader OH/NH stretching bands ($3200\text{--}3400$ cm^{-1}), particularly in Y3. The observed band broadening and slight shifts in the COO^- region suggest possible hydrogen bonding and electrostatic interactions between COO^- groups of SA and NH_3^+ groups of CS [50–52]. Compared to X3 (0.5% SA), Y3 (1.0% SA) showed more pronounced OH/NH band broadening and stronger amide II absorption, which may be attributed to the increased availability of carboxylate groups, which may indicate enhanced intermolecular interactions. These findings suggest the potential of the developed coating for applications where improved surface functionality and

hydrophilicity are desirable. For X3 (0.5% SA), CS induces OH/NH broadening with moderate Amide II intensity, whereas Y3 (1.0% SA) shows more pronounced OH/NH broadening, stronger Amide II absorption, and a marked COO^- shift due to ionic interactions. The red rectangle area ($3000\text{--}3400$ cm^{-1}) represents the OH/NH expansion region, and the green rectangle area represents the Amide II / COO^- region [48]. The FTIR spectra showed that Y3 exhibited higher-intensity peaks in these regions than X3 (Figure 3). Compared with untreated PLA, the O–H stretching band shifted from 3421 to 3325 cm^{-1} , which may indicate the establishment of hydrogen-bonding interactions between the hydroxyl groups of the hydrogel components and the PLA/WPU matrix [19, 20]. Nevertheless, FTIR spectroscopy

alone cannot conclusively confirm hydrogen bond formation. Furthermore, slight shifts in the C=O and C–O bands suggest intermolecular interactions among PLA, alginate, lignin/CS, and WPU. The presence of ZnO was evaluated through weak absorption bands in the 500–600 cm^{-1} region, typically associated with Zn–O stretching vibrations [42, 44]. Although this region is known to be weak and partially overlapped, the appearance and gradual intensification of bands in the 604–665 cm^{-1} range with increasing ZnO content is consistent with the successful incorporation of ZnO into the composite structure. It should be noted that overlapping bands among PLA, alginate, lignin, and CS may influence peak positions and intensities. Therefore, peak assignments (Table 2) were made cautiously and supported by comparative analysis across formulations rather than relying on single-band identification. As a result, the observed spectral changes, including band shifts, peak broadening, and intensity variations, are consistent with the successful incorporation of alginate, ZnO, lignin, and CS into the PLA-based coating and suggest the presence of possible intermolecular interactions, such as hydrogen bonding, ionic interactions, and metal–carboxylate coordination. However, FTIR spectroscopy alone cannot conclusively verify these interactions because of overlapping absorption bands and the complex nature of the composite system. Therefore, the peak assignments and proposed interactions are based on comparative spectral analysis and supported by previously published literature rather than direct spectroscopic evidence.

3.2 SEM Analysis Results

SEM images of untreated PLA fabric and hybrid hydrogel composite PLA nonwoven fabric samples are shown in Figures 4–7. EDS results of the sample of Y1, Y2 and Y3 were given in Figure 8. SEM image of untreated PLA fabric with x50 and x200k magnification rates was given in Figure 4.

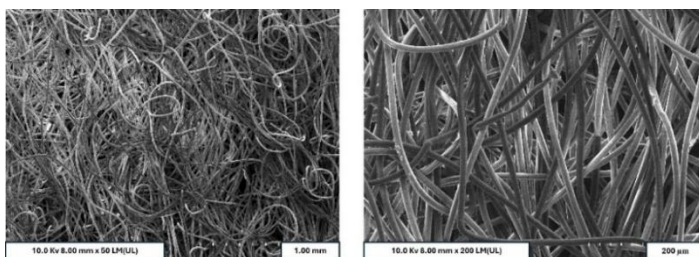


Figure 4. SEM images of untreated PLA fabric with x50 and x200 magnifications

Figure 4 shows that the untreated PLA spunlace nonwoven fabric exhibited smooth fibers with distinct gaps, forming a typical porous structure of loosely packed fibers. No coating or particles were observed, indicating preserved fiber morphology and high air permeability. After coating, a continuous layer formed on the fiber surfaces, confirming the successful deposition of the alginate-based WPU coating (Figures 4–7). At higher magnification, ZnO particles appeared as localized agglomerated clusters in some regions, while partial particle dispersion was observed in others; however, a fully homogeneous distribution

could not be clearly confirmed from the SEM images. The thin SA layer partially covered the fibers while largely preserving the underlying fibrous structure. Figure 5 shows that lignin addition provided a more matte appearance. Compared to Y1, the gaps between the fibers were partially reduced, and coating continuity improved. ZnO particles were still visible on the fiber surface; however, lignin altered the surface topography, reducing the clarity of particle boundaries. The coating layer appeared more compact, indicating improved surface coverage. At lower alginate concentrations (0.5%), the coating layer remained relatively thin and partially covered the fibers, leaving some fiber boundaries visible. This incomplete coverage suggests limited film-forming ability, which may explain the reduction in tensile strength observed in these samples. In contrast, increasing the alginate concentration to 1% resulted in a more uniform and continuous coating layer. The fibers appeared more interconnected, indicating enhanced inter-fiber bonding due to improved film formation. This more homogeneous structure likely contributed to the higher tensile strength values observed in the Y-series samples (Figures 5–7).

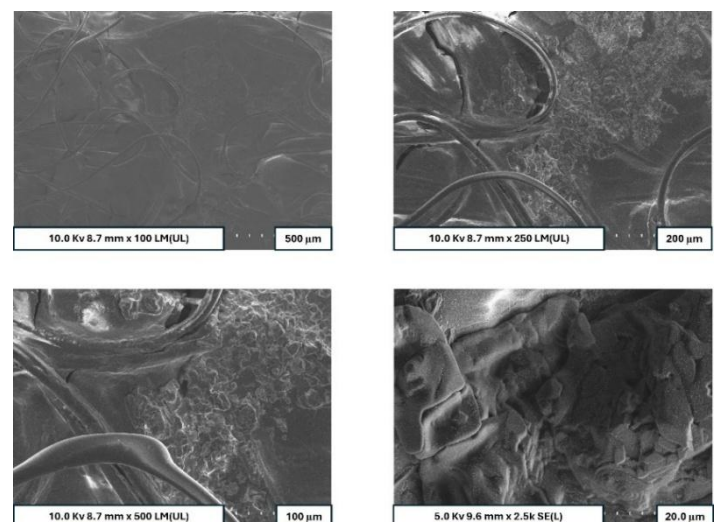


Figure 5. SEM images of Y1 sample with x100, x250, x500 and x2.50k magnifications

Figure 7 demonstrates that the incorporation of CS further enhanced coating continuity and film formation. At high magnification, the fibers were almost completely covered by the coating layer, while ZnO particles appeared to be embedded within the polymer matrix, although localized particle agglomerates were observed in some regions. These localized agglomerates did not appear to induce coating cracking, delamination, or discontinuities, suggesting that they did not adversely affect the overall integrity of the coating at the applied ZnO loading. The coating surface became rougher and denser, with minimal visible voids or structural defects. The CS–SA matrix likely formed a three-dimensional crosslinked network, contributing to stronger fiber bonding and improved coating integrity. In addition, lignin incorporation may generate irregular grain-like surface features and micro-protrusions within the

composite layer [53, 54]. The Y3 (SA/L/CS/ZnO) sample exhibited a more compact and continuous coating morphology together with ZnO particle deposition, which may enhance antibacterial performance (Table 6). EDS analysis (Figure 8) was performed at different regions of the coated fabric surfaces to evaluate elemental distribution. The EDS analysis performed in this study was qualitative rather than quantitative. Although Zn was detected in all analyzed regions, the spectra cannot quantitatively verify the spatial homogeneity of ZnO distribution across the entire fabric surface. Zn signals were consistently detected in the coating layers of Y1 (SA/ZnO), Y2 (SA/L/ZnO), and Y3 (SA/L/CS/ZnO), confirming the successful incorporation of ZnO into the hydrogel matrix. In addition, Ca signals detected in the spectra are attributed to the Ca^{2+} ions used during the hydrogel crosslinking process. The presence of Ca may also suggest residual CaCl_2 salts remaining within the hydrogel matrix after crosslinking. The coating morphology showed that the fibers were largely covered by a continuous film layer. The CS-SA matrix appeared to form a three-dimensional crosslinked structure [46], while lignin incorporation contributed to irregular flake-like or suture-like surface features. ZnO nanoparticles were observed as granular bright spots either on the fiber surface or partially embedded within the coating network.

The coating exhibited relatively continuous morphology with similar features in the examined regions; however, complete surface homogeneity cannot be confirmed solely by SEM observations. Owing to the denser coating structure, a reduction in porosity may have occurred, which could potentially influence breathability [55]. These results indicate that alginate concentration plays a dominant role in determining coating morphology and mechanical performance, rather than ZnO content under constant loading conditions. To evaluate coating

homogeneity, SEM images were obtained from multiple randomly selected regions across the fabric surface at different magnifications. The consistency of fiber coverage, coating continuity, and the absence of large uncoated areas or severe structural defects were used as qualitative indicators of homogeneity. Although a full surface mapping analysis was not performed, the similar morphological features observed across different regions suggest that the coating was relatively uniformly distributed. The coating mechanism is supported by the complementary information obtained from FTIR, SEM, and EDS analyses. FTIR spectra suggest possible intermolecular interactions among SA, CS, and lignin, as evidenced by the broadening of the OH/NH stretching region together with slight changes in the COO^- and amide II bands. Although these spectral changes are consistent with hydrogen bonding and electrostatic interactions reported in the literature, they do not constitute direct proof of these interactions. SEM observations confirm the formation of a continuous hydrogel coating on the PLA fibers and reveal the incorporation of ZnO particles within the coating layer, although localized particle agglomerates were observed in some regions. EDS analysis verified the presence of Zn and Ca in the coated samples, supporting the incorporation of ZnO and the presence of Ca^{2+} associated with the alginate crosslinking process. Collectively, the FTIR, SEM, and EDS results provide complementary evidence supporting the successful deposition of a Ca^{2+} -crosslinked SA/L/CS hydrogel coating containing ZnO particles onto the PLA nonwoven substrate.

3.3 Tensile Analysis Results

The tensile strength (TS) and elongation at break (E) of untreated PLA and hybrid hydrogel composite PLA nonwoven fabric samples were measured to evaluate the mechanical performance of various hydrogel formulations (Table 3).

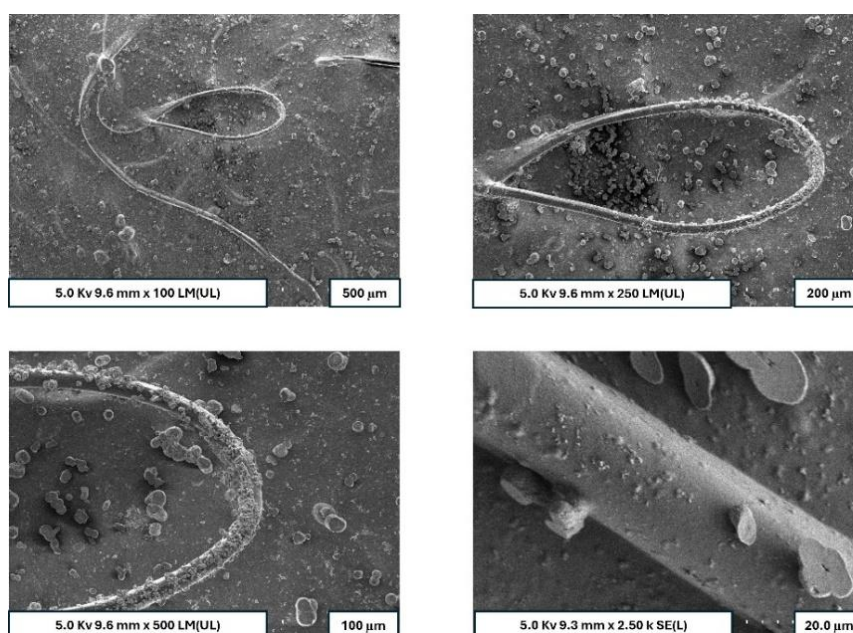


Figure 6. SEM images of Y2 sample with x100, x250, x500 and x2.50k magnifications

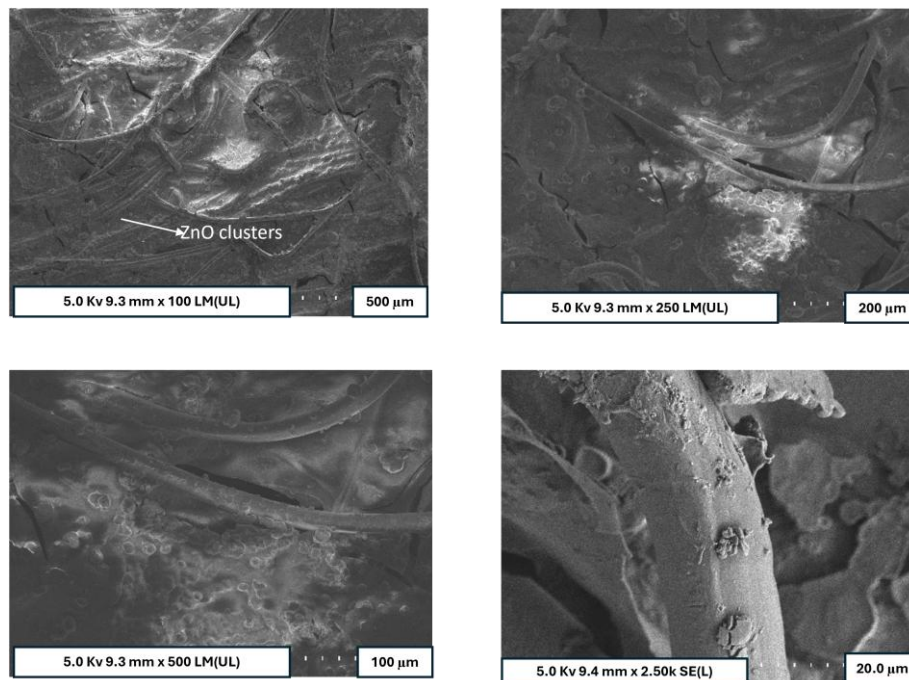


Figure 7. SEM images of Y3 sample with x100, x250, x500 and x2.50k magnifications

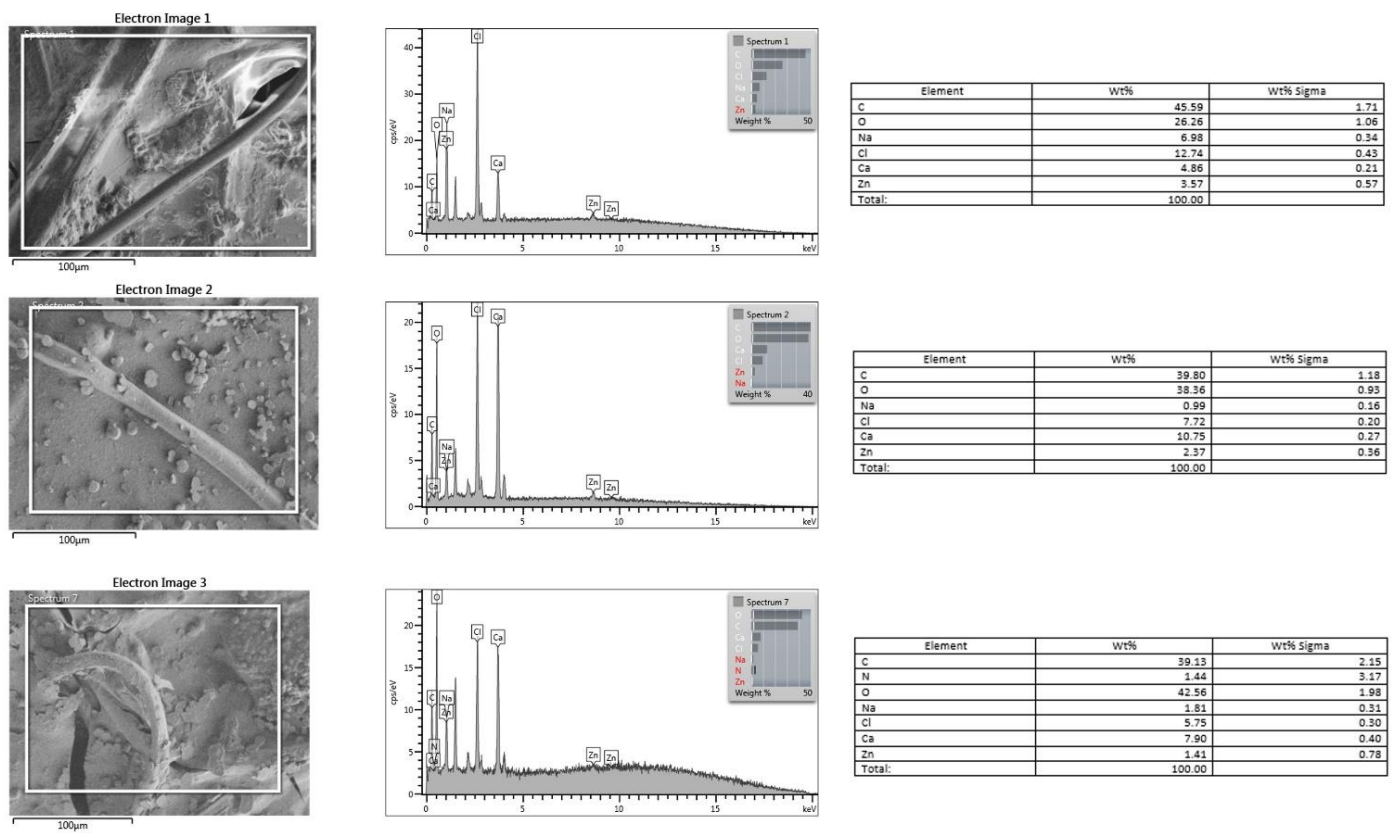


Figure 8. EDS results of fabric samples prepared with Y1, Y2 and Y3 hybrid hydrogel composite solutions

Table 3. Tensile strength and elongation at break of untreated PLA fabric, and hybrid hydrogel composite fabric samples (TS: Tensile strength, E: Elongation at break), ($\pm$ SD: standard deviation, CV (%): Coefficient of variation)

Sample Code	TS (N, mean $\pm$ SD)	% CV	E (% , mean $\pm$ SD)	% CV
PLA fabric	77 $\pm$ 1.31 ^{bcd}	1.7	31.5 $\pm$ 2.46 ^a	7.8
X	80 $\pm$ 3.44 ^a	4.3	26.0 $\pm$ 1.51 ^a	5.8
X1	69 $\pm$ 2.35 ^d	3.4	30.5 $\pm$ 1.80 ^a	5.9
X2	74 $\pm$ 1.92 ^{cd}	2.6	29.0 $\pm$ 2.06 ^a	7.1
X3	75 $\pm$ 3.08 ^{cd}	4.1	29.0 $\pm$ 2.73 ^a	9.4
Y	85 $\pm$ 3.91 ^a	4.6	27.0 $\pm$ 2.24 ^a	8.3
Y1	77 $\pm$ 3.31 ^c	4.3	30.0 $\pm$ 0.69 ^a	2.3
Y2	86 $\pm$ 5.07 ^{ab}	5.9	28.0 $\pm$ 2.32 ^a	8.3
Y3	77 $\pm$ 4.16 ^c	5.4	27.0 $\pm$ 2.46 ^a	9.1

*Values are presented as mean $\pm$ standard deviation (n = 5). Different superscript letters within the same column indicate statistically significant differences according to one-way ANOVA followed by Tukey's HSD test ($p < 0.05$).

Pure PLA fabric exhibited a tensile strength of 77 N [26]. The coated samples retained approximately 90–112% of the tensile strength of untreated PLA fabric, indicating that the hydrogel coatings generally preserved the mechanical integrity of the nonwoven structure. In the samples coded as X, containing 0.5% SA, TS values ranged from 69 to 80 N, indicating that hydrogel composites at this concentration did not adversely affect the mechanical integrity of the fabric. In the samples coded as Y series with 1.0% AL, TS values were slightly higher in some formulations, reaching up to 86 N for Y2, while most samples remained in the 77–85 N range. One-way ANOVA demonstrated that tensile strength differed significantly among the formulations ($p < 0.001$). Tukey's HSD test further revealed that Y2 exhibited higher tensile strength than X1, X2, and X3, whereas no significant difference was observed between Y2 and the untreated PLA fabric or sample X. This suggests that increasing SA content to 1.0% may provide a slight reinforcement effect without weakening the fabric structure in some formulations. Similar trends have been reported for AL hydrogel fibers, where increasing AL concentration from 0.5% to 1.0% (w/v) increased tensile tenacity from 2.1 to 8.16 cN/Tex due to enhanced crosslinking density [40]. These findings are consistent with previous reports suggesting that higher SA content may contribute to improved mechanical performance through increased network formation. The untreated PLA fabric demonstrated an elongation at break of 31.5 %. Coating with X-series hydrogels (0.5% SA) led to a moderate decrease for sample X (26.0%), while variants X1–X3 retained elongation values close to untreated PLA (29.0–30.5%). In the Y-series with higher SA content (1.0%), elongation at break ranged from 27.0% to 30.0%, slightly lower than that of untreated PLA. These results suggest that increasing SA concentration causes a minor reduction in flexibility; however, the overall extensibility remains acceptable for practical applications. Similar trends have been observed in the literature, for example, increasing SA concentration in SA hydrogel fibers decreases elongation at break due to enhanced cross-linking and resultant rigid structure formation [42]. This supports the notion that higher SA content slightly diminishes flexibility, as reflected in the results of hybrid hydrogel composite fabrics (Table 3). Although

the hydrogel coating formed a continuous layer on the fiber surface, only a slight reduction in elongation at break was observed compared with the untreated PLA fabric. This suggests that the coating did not substantially restrict fiber mobility within the nonwoven structure. The coated fabrics, therefore, retained a relatively high degree of flexibility, which is advantageous for applications requiring both mechanical integrity and conformability. The slightly lower elongation observed for the Y-series samples is likely associated with the higher SA content and the resulting denser hydrogel network, which may moderately increase coating stiffness while maintaining acceptable flexibility. SEM observations showed that the hydrogel coating covered the fiber surfaces without completely filling the inter-fiber spaces. This morphology may explain why the coated fabrics maintained considerable flexibility despite the presence of the hydrogel layer.

In Y2 (SA/L/ZnO), lignin appeared to contribute to a more uniform coating morphology, which may have contributed to the observed tensile behavior. The incorporation of lignin and ZnO affected the tensile properties of the hybrid hydrogel composites. Among the tested formulations, Y2 (SA/L/ZnO) exhibited the highest tensile strength (86 N). This behavior may be associated with the combined presence of lignin and ZnO together with the higher SA concentration, which likely promoted the formation of a more continuous coating layer, as suggested by the SEM observations. Similar strengthening effects of lignin-containing hydrogels have been reported in previous studies, where lignin acts as a rigid domain or reinforcing filler within polymer matrices [2, 16, 37]. In sample Y3, the TS (77 N) was lower than that of Y2 (Table 3). Although Y3 exhibited a relatively dense coating morphology, its tensile strength was slightly lower than that of Y2. This difference may be associated with localized coating heterogeneity and the presence of possible local ZnO particle agglomeration, as suggested by some SEM images. Although statistically significant differences in tensile strength were identified, the absolute differences among several formulations remained relatively modest (approximately 10–17 N) [13].

The addition of CS (X3 and Y3) slightly reduced TS compared to lignin/ZnO-only samples but maintained elongation values

comparable to untreated PLA. Although ZnO incorporation may contribute to reinforcement through nanoparticle–polymer interactions, the reduced TS observed for Y3 may be associated with non-uniform coating morphology and irregular particle dispersion within the hydrogel matrix. Coating morphology can influence failure behavior by affecting stress distribution during tensile loading. More homogeneous coating structures may provide improved stress transfer and reduced stress concentration points, whereas local agglomeration and structural heterogeneity may promote earlier failure under mechanical stress. However, the slightly lower TS observed for Y3 indicates that excessive additive content or non-uniform dispersion can weaken intermolecular interactions and reduce mechanical integrity. In addition, the incorporation of ZnO nanoparticles can enhance tensile strength through NP–polymer interactions and load transfer within the hydrogel network, as reported for CS-based nanocomposites [25, 51]. Furthermore, ionically cross-linked hydrogels can exhibit enhanced mechanical stability due to the formation of physical cross-linking networks. Cross-linking agents such as Ca^{2+} and Zn^{2+} improve the tensile strength (TS) of CS/SA hydrogels [3]. Similarly, embedding ZnO nanoparticles in CS matrices generally increases TS and Young's modulus through strong matrix–nanoparticle interactions and NP reinforcement [56, 57]. The incorporation of ZnO, lignin, and CS into the SA matrix generally preserved the TS and elongation properties of the PLA fabrics. Although one-way ANOVA indicated an overall difference in elongation at break, Tukey's HSD test revealed no statistically significant pairwise differences among the formulations. Therefore, the observed variations in elongation

should be interpreted as descriptive trends rather than statistically confirmed differences. These results indicate that hydrogel composition can be adjusted to achieve a balance between strength and flexibility while retaining the mechanical integrity of the PLA fabric. The Y-series (1.0% SA) exhibited a reinforcement trend comparable to that observed in recent PVA–PLA and PVA-cotton hydrogel composite systems [58, 59], showing increased tensile strength while maintaining acceptable flexibility. As summarized in Table 4, the tensile strength values obtained in this study fall within the range reported for various hydrogel-coated or composite textile systems.

testing conditions reported in the literature. Nevertheless, these findings suggest that the hydrogel composition can be tailored to balance tensile strength and flexibility, allowing the coated PLA nonwoven fabrics to maintain their mechanical integrity while preserving sufficient flexibility for potential biomedical and textile applications. While hydrogel add-on may have partially influenced the tensile behavior, the lack of a direct correlation between add-on percentage and tensile strength suggests that compositional interactions within the hydrogel matrix also played a role in the mechanical performance. Although statistically significant differences were detected for tensile strength, the overall mechanical performance of all hybrid hydrogel composite fabrics remained comparable to that of untreated PLA, indicating that the hydrogel coatings preserved the mechanical integrity of the nonwoven substrate while allowing modest formulation-dependent variations (Table 3).

Table 4. Summary of the tensile strength performances of this study and previous studies

Matrix/Filler	Substrate / Fabric Type	Method	Tensile Strength (TS)	Elongation at Break (EB)	Key Findings / Mechanism	References
-	PLA	Spunlace nonwoven technology, fabric weight: 50 g/m ²	77 N	31.5%	Baseline mechanical performance	This work – untreated PLA
SA/L/CS/ZnO hydrogel	PLA nonwoven	Sol-gel + ionic crosslinking (Ca^{2+})	69–80 N	26–30.5 %	Slight TS decrease vs pure PLA; flexibility mostly retained	This work – X-series (0.5% SA)
SA/L/CS/ZnO hydrogel	PLA nonwoven	Sol-gel + ionic crosslinking (Ca^{2+})	77–86 N (Y2 = 86 N)	27–30 %	Higher SA gives mild reinforcement; Y3 drops due to lignin dispersion	This work – Y-series (1.0% SA)
Alginate hydrogel fibers (1.0 → 1.5 %)	-	Wet spinning + CaCl_2 crosslinking	Tenacity ↑ 2.1→8.16 cN/Tex	EB ↓ with higher AL	Higher alginate increases crosslink density; TS↑, flexibility↓	[30]
Chitosan + nano ZnO composite films	-	Solution casting	TS ↑ with ZnO	EB variable	ZnO–polymer matrix interactions reinforce structure	[60]
PVA hydrogel	PLA mesh	In situ polymerization + freeze-drying	TS ↑ by ≈ 25 % vs hydrogel alone	EB retained	PLA mesh improves tensile resistance; tissue scaffold potential	[58]
PVA-based hydrogel	Cotton fabric	In situ gelation + fabric embedding	Breaking load ↑~1.8×	EB maintained	Fabric reinforcement provides stretchability and strength; textile–hydrogel synergy validated	[59]

3.4 Air permeability results

Table 5 presents the air permeability performance of untreated PLA fabric and hybrid hydrogel composite PLA fabrics.

In wound dressings, controlled air permeability is crucial to maintain a moist healing environment while allowing adequate oxygen transfer to the wound bed. Excessively high permeability can cause dehydration, whereas very low permeability may risk anaerobic bacterial growth and delayed healing [61, 62]. Neat PLA fabric exhibited extremely high air permeability ($1287 \text{ l/m}^2\cdot\text{sec}$) [26, 63], higher than the relatively low permeability ranges commonly reported for moisture-retentive wound dressing materials [64]. The incorporation of hybrid hydrogel composites resulted in a substantial reduction in air permeability, suggesting partial pore blockage and modification of air transport pathways within the fabric structure by the hydrogel layer. The substantial reduction in air permeability, together with SEM observations, suggests partial restriction of air transport pathways after hydrogel coating. ZnO-containing composites (X1, Y1) showed substantial permeability reduction (29.6 and $2.73 \text{ l/m}^2\cdot\text{sec}$, respectively), consistent with ZnO's fine particulate dispersion filling micro voids, as reported in hydrogel–nanoparticle dressings [65]. Lignin-containing composites (X2, Y2) showed lower permeability (6.22 and $1.58 \text{ l/m}^2\cdot\text{sec}$). Lignin's ability to reinforce the hydrogel matrix and reduce porosity may contribute to this effect [66]. L/CS/ZnO composites (X3, Y3) had the lowest permeability in the X series ($22.9 \text{ l/m}^2\cdot\text{sec}$) and moderately low in the Y series ($6.41 \text{ l/m}^2\cdot\text{sec}$). CS's film-forming ability and strong intermolecular bonding likely further restricted open pore pathways, contributing to reduced air permeability and potentially enhancing moisture retention [67]. The permeability behavior appeared to be qualitatively consistent with the SEM observations, where samples exhibiting more continuous coating coverage generally showed lower air permeability, suggesting partial closure of the interconnected pore network. The reduced permeability suggests restricted air transport behavior that may help limit excessive moisture evaporation while still potentially allowing a certain degree of gas exchange through the dressing structure. However, air permeability and moisture vapor transmission are related but distinct transport properties, and air permeability alone does not directly define moisture vapor transmission behavior. Particularly, the Y-series (1.58 – $6.41 \text{ l/m}^2\cdot\text{sec}$) exhibited lower air permeability, which may contribute

to increased moisture retention. Among the investigated formulations, the Y-series, particularly Y1 and Y3, exhibited a combination of high absorbency and low air permeability, which may be advantageous for applications requiring enhanced fluid uptake and moisture retention. These properties align with the moisture balance and oxygenation requirements for chronic and exudative wound care [61]. The relatively high CV values observed for some coated samples may be related to the heterogeneous pore structure of nonwoven fabrics and local variations in hydrogel coating coverage. Since air permeability is highly sensitive to small changes in open pore pathways, small absolute differences may result in high CV values, especially at low permeability levels. The air permeability results obtained in this study are consistent with those reported in previous literature. The neat PLA fabric exhibited high air permeability ($1287 \text{ l/m}^2\cdot\text{s}$) comparable to highly open structures such as PET-based dressings [67] and commercial Suprasorb-A ($\sim 2400 \text{ l/m}^2\cdot\text{s}$), which are considered excessively breathable for maintaining a moist wound environment. Such high permeability may promote increased moisture evaporation, which could influence the moisture retention characteristics of the material. After incorporation of the hybrid hydrogel composites, the air permeability decreased markedly to 1.58 – $29.6 \text{ l/m}^2\cdot\text{s}$ depending on the formulation. These values are comparable to those reported for hydrogel or nanofiber-based wound dressing materials (e.g., PVA/NaAlg nanofibers $\approx 2 \text{ l/m}^2\cdot\text{s}$) [68], indicating the formation of a more controlled porous structure that may help limit excessive moisture evaporation while still allowing gas diffusion. In particular, the Y-series samples (1.58 – $6.41 \text{ l/m}^2\cdot\text{s}$) exhibited relatively low permeability values comparable to hydrogel-based dressing structures reported to support a moist and oxygen-permeable environment beneficial for wound healing [61, 67, 69]. Although increased hydrogel add-on generally contributed to reduced air permeability, the permeability behavior also appeared to be influenced by coating morphology and hydrogel composition. Although variations in permeability values were observed among the formulations, the results should be interpreted as indicative trends in gas transport behavior. Overall, the hybrid hydrogel composites, particularly Y1 and Y3, demonstrated reduced air permeability and enhanced fluid absorbency, indicating promising moisture management characteristics. However, their suitability for wound dressing applications requires further biological and functional evaluation.

Table 5. Air permeability values of untreated PLA fabric and hybrid hydrogel composite fabric samples, ($\pm$ SD: standard deviation, CV: Coefficient of variation)

Sample Code	PLA fabric	X	X1	X2	X3	Y	Y1	Y2	Y3
Air permeability ($\text{l/m}^2\cdot\text{sec}$)	1287	0.67	29.6	6.22	22.9	0.59	2.73	1.58	6.41
CV (%)	3.6	68.0	87.2	43.0	22.0	69.0	26.0	27.0	15.0
$\pm$ SD	46.33	0.46	25.81	2.67	5.04	0.41	0.71	0.43	0.96

3.5 Fluid Absorbency Capacity Results

Table 6 presents the fluid absorbency performance of untreated PLA and hydrogel composite PLA fabrics. The dry weight values reported for the coated samples correspond to the hydrogel-coated fabrics. Therefore, the hydrogel add-on can be determined from the difference between the dry weight of the coated samples and that of the untreated PLA fabric (Table 6). The hydrogel add-on ranged approximately from 0.3% to 25.9%, depending on the coating formulation. Variations in hydrogel add-on among the formulations may be related to differences in coating composition, hydrogel structure, and coating retention behavior on the PLA fabric surface. Although the hydrogel add-on of sample X1 was slightly lower than that of sample X, its fluid absorbency was higher. This behavior can be attributed to the presence of ZnO nanoparticles, which may increase the porosity and hydrophilicity of the alginate hydrogel network. The nanoparticles can act as spacers within the polymer matrix, facilitating water diffusion and enhancing swelling capacity. Therefore, the absorbency performance is influenced not only by the coating amount but also by the microstructure of the hydrogel composite. The absorbency behavior may also be influenced by coating morphology and local variations in hydrogel distribution across the fabric surface. Untreated PLA fabric exhibited the lowest fluid absorbency value (206.579%), indicating an inherently limited water retention capacity. The incorporation of hybrid hydrogel composites into the PLA matrix (X-series) increased absorbency markedly, with values ranging from ~307% to ~363%, reflecting the hydrophilic nature and network-forming ability of SA. Hydrogels are known to form three-dimensional hydrophilic polymer networks capable

of significant water uptake and swelling [70, 71], which is particularly relevant for biomedical applications where liquid absorption ensures moisture balance, aids necrotic tissue removal, controls exudate, relieves pain, and supports autolytic debridement, while also preventing adherence and promoting granulation and epithelialization in wound healing [72]. However, excessive cross-linking can restrict water penetration and consequently limit swelling [12]. The swelling behavior may also be influenced by the effective crosslink density of the hydrogel network, where a more compact structure could restrict water diffusion and reduce fluid uptake. The reproducibility of the coating process was confirmed by the relatively low standard deviation values obtained from repeated fluid absorbency measurements, indicating consistent hydrogel deposition on the fabric surface. The absorbency values obtained in this study are comparable to those reported for hydrogel-coated textile dressings and alginate-based wound dressing materials, where the incorporation of hydrophilic polymers enhances liquid uptake while reducing air permeability. Although direct comparison is limited by differences in substrates, coating formulations, and test methods, the observed absorbency trend agrees well with previous reports [23, 61, 67, 69]. The increase in absorbency despite the reduction in air permeability can be explained by the different transport mechanisms involved [61, 62, 67]. The hydrogel coating partially blocked the open air pathways between fibers, thereby reducing convective air flow [58]. At the same time, the hydrophilic functional groups of SA, CS, and lignin increased the affinity of the coating toward water, enabling greater water uptake and swelling. Consequently, air permeability decreased while absorbency increased [19, 20, 23, 56, 61, 67].

Table 6. Fluid absorbency values of untreated PLA fabric, and hybrid hydrogel composite fabric samples, ($\pm$ SD: standard deviation)

Weight	PLA fabric	X	X1	X2	X3	Y	Y1	Y2	Y3
Dry weight	0.1962	0.1968	0.1966	0.2088	0.2388	0.2068	0.2163	0.2181	0.2470
Add-on (%)	-	0.31	0.20	6.42	21.71	5.40	10.24	11.16	25.89
Wet weight	0.8713	0.8013	0.8980	0.9682	1.0649	0.9003	1.0617	0.9078	1.1530
Fluid absorbency (%)	206.579	307.165	381.243	363.697	345.938	335.348	390.846	316.231	366.802
$\pm$ SD	0.82	1.05	0.87	2.15	0.63	1.12	1.24	0.69	0.66

Since the absorbency values were calculated based on the total mass of the hydrogel-coated composite fabrics, differences in hydrogel add-on may partially influence direct comparison among formulations. No obvious coating detachment or structural disintegration was visually observed during the absorbency measurements under the applied immersion conditions. In sample X1, the ZnO-containing sample exhibited a relatively higher absorbency value as 381.243%. This observation is consistent with findings reported for other hydrogel systems, where ZnO nanoparticles have been suggested to influence swelling behavior through possible modifications in pore structure, hydrogel network

organization, and polymer–water interactions [11, 70]. In sample X2, lignin reduced absorbency to 363.697%, and lignin/CS effect in sample X3 further decreased it to 345.933%. Lignin's hydrophobic character restricts water penetration and retention, consistent with reports of reduced water uptake in composites containing lignin [72, 73]. This effect can be explained by the presence of free lignin in the structure, which contains many hydrophilic, non-reacted groups that facilitate water absorption and transport to the substrate surface [74]. Furthermore, CS, despite being hydrophilic, likely interacts with lignin to form a denser matrix, limiting pore accessibility and water diffusion, hence the further drop in absorbency in X3.

All Y-series samples exceeded their X-series counterparts in absorbency, underscoring the positive impact of higher SA content, likely due to expanded network structure and increased water-binding sites. Y1 exhibited the highest absorbency among the tested samples at 390.846%, suggesting a possible contribution of ZnO in enhancing swelling capacity [69]. Y2 and Y3 (with lignin and lignin/CS) showed lower absorbency than Y1, again reflecting lignin's hydrophobic and blocking influence on water uptake [72, 73]. Notably, the transition from X to Y yielded the greatest improvement for the plain SA/PLA composite (~+28.18%), suggesting that SA concentration plays a dominant role when additive hindrance effects are minimal. Separately, dry and wet weights increased from neat PLA to hydrogel-modified composites, consistent with greater solid content (dry weight) and enhanced fluid retention (wet weight). ZnO contributed to absorbency without compromising the hydrophilic matrix, whereas lignin (with or without CS) limited water uptake and thus moderated the increase in wet weight, in line with reported trends of decreased moisture uptake in lignin-enriched composites [73].

CS-based hydrogels with ZnO exhibit a formulation-dependent swelling trend. ZnO can enhance swelling in matrices with low crosslink density by increasing pore volume and hydrophilic interactions, but it may suppress swelling in highly crosslinked networks where the nanoparticles act as additional physical crosslinks. This mechanism aligns with the findings of this study, where ZnO-containing samples (X1, Y1) showed the highest absorbency (~381–391%) [75]. Lignin-containing composites display mixed swelling behavior in the literature, with reports of both enhanced and reduced water uptake. This variation depends on lignin's chemistry, dispersion, and its interactions within the hydrogel network. In this work, the lignin formulations (X2/Y2 and X3/Y3) exhibited lower absorbency than the ZnO samples, consistent with studies suggesting that lignin's hydrophobic domains or its densification effect can reduce pore accessibility unless chemically modified to increase hydrophilicity [19]. The observed pattern higher swelling with ZnO and a moderating effect from lignin/chitosan accords well with prior experimental and review studies [30]. Overall, the absorbency range observed here (~300–390%) is generally comparable to values reported in the literature for hydrogel-based wound dressing materials, although direct comparison is limited by differences in testing conditions and material composition. The absorbency and air permeability behaviors appeared to be qualitatively related, since hydrogel structures promoting fluid retention also tended to restrict air transport pathways within the composite fabric structure.

3.6 Antibacterial Activity Results

Table 7 presents the antibacterial performance of untreated PLA fabric and hybrid hydrogel composite PLA fabrics against *Escherichia coli* (ATCC 25922) and *Staphylococcus aureus* (ATCC 6538). Based on the preliminary characterization results, particularly the air permeability and structural integrity relevant to wound dressing applications, the Y1 and Y3 formulations were

selected as representative samples for antibacterial evaluation. Antibacterial activity images were also given in Figure 9.

The reference untreated PLA fabric shows no measurable antibacterial effect, with high surviving bacteria counts. In contrast, coating PLA with ZnO-containing hydrogels greatly improved antibacterial performance. Y1 (SA/ZnO hydrogel) achieved strong bacterial reduction, while Y3 (SA/L/CS/ZnO hydrogel) exhibited slightly higher antibacterial activity, suggesting a possible combined antibacterial contribution from the combined antimicrobial properties of CS, the phenolic groups in lignin, and the photocatalytic activity of ZnO nanoparticles [24]. Y3 (SA/L/CS/ZnO) formed an almost continuous fiber-encapsulating film with ZnO particles embedded in the polymer matrix, indicating improved mechanical integrity (Table 3) and greater antimicrobial activity, particularly against *E. coli*. The antibacterial behavior appeared to be qualitatively associated with coating morphology and ZnO distribution within the hydrogel layer, as suggested by the SEM/EDS observations. The antibacterial behavior may also be influenced by coating morphology and the distribution uniformity of the antibacterial components within the hydrogel layer. Inorganic nanocomposite hydrogels are promising antibacterial materials, with ZnO NPs offering a safe alternative to silver- or gold-based systems. Their strong antibacterial activity arises from multiple mechanisms, including membrane disruption, zinc ion release, and ROS generation, which are further enhanced at the nanoscale [10, 76, 77]. CS complements this effect through its cationic nature, disrupting bacterial membranes and binding intracellular DNA, while also contributing wound-healing benefits such as hemostasis and analgesia [3, 5, 48]. AL, as a biocompatible and biodegradable polymer, provides high water retention capacity. When combined with CS, polyelectrolyte complex formation occurs between the negatively charged AL and positively charged CS chains, which may partially reduce the availability of free cationic sites responsible for CS's antibacterial activity [53, 78].

Lignin further strengthens the hydrogel structure and adds antioxidant and antibacterial activity, though its proportion must be optimized to avoid neutralizing CS's cationic groups [13, 48, 56, 79, 80]. Consistent with these mechanisms, the present study demonstrated that Y1 markedly reduced bacterial counts (93.32% for *E. coli* and 99.42% for *S. aureus*), whereas Y3 achieved >98% reduction against both strains, indicating that the combined effects of ZnO, CS, and lignin may enhance antibacterial performance. Previous studies have suggested that ZnO-containing systems exhibit antibacterial activity through mechanisms including membrane interaction, Zn ion release, and reactive oxygen species generation [50, 56, 76, 81]. The calculated log reduction values further supported the strong antibacterial performance of the ZnO-containing hydrogel-coated PLA fabrics.

As a result, the combination of ZnO, CS, SA, and lignin in a single hydrogel system may collectively contribute to the observed antibacterial performance together with modified physicochemical and mechanical characteristics of the coated

PLA fabrics. The antibacterial behavior appeared to be primarily associated with the presence of ZnO, while CS and lignin may provide additional supportive contributions. In this study, untreated PLA fabric showed essentially no measurable antibacterial activity, as indicated by high surviving bacterial counts. In contrast, coating the PLA fabric with ZnO-containing hydrogel formulations improved antibacterial performance. The SA/ZnO hydrogel (Y1) resulted in a strong reduction in bacterial viability, whereas the SA/L/CS/ZnO hydrogel (Y3) exhibited the highest antibacterial efficiency. This enhanced activity can be attributed to the synergistic effect of ZnO nanoparticles, which are known to induce membrane disruption and ion release, together with the intrinsic antimicrobial activity of CS and the phenolic functionalities of lignin that further contribute to bacterial inhibition. This behavior is consistent with existing literature: for example, aerogels based on alginate/ hyaluronic acid (HA) loaded with ZnO nanoparticles showed significant antibacterial activity compared to ZnO-free controls [82] likewise, sodium alginate–

chitosan-oligosaccharide–ZnO composite hydrogels exhibited pronounced antibacterial effects against *E. coli* and *S. aureus* [10] and comprehensive studies that integrate polysaccharide–metal oxide nanoparticle systems confirm that combinations of biopolymer matrices (e.g., CS, alginate) with metallic nanoparticles (such as ZnO) achieve enhanced antimicrobial effects compared to either component alone [83]. Although the present study provides insight into the structural, physicochemical, and functional characteristics of the developed ZnO-containing hybrid hydrogel composite PLA nonwoven fabrics, further biological safety and biocompatibility evaluations remain necessary for potential biomedical-related applications. In addition, the durability of the antibacterial performance after repeated washing or aging was not evaluated in the present study. Future studies should investigate the long-term stability of the hydrogel coating and the retention of its antibacterial activity under conditions relevant to practical use.

Table 7. Antibacterial activities of neat PLA fabric and hybrid hydrogel composite fabric samples

Samples	Initial microorganism concentration (CFU/mL)		Log reduction		Antibacterial activity (%)
	<i>E. coli</i> (ATCC 25922): 2×10^7	<i>S. aureus</i> (ATCC 6538): 4×10^7	Log reduction (<i>E. coli</i>)	Log reduction (<i>S. aureus</i>)	
Reference neat PLA fabric	3.08×10^5	1.24×10^6	-	-	-
Y1	2.06×10^4	7.02×10^3	3.17	2.25	<i>S. aureus</i> : 99.42 <i>E. coli</i> : 93.32
Y3	6.10^3	6.10^3	3.71	3.32	<i>S. aureus</i> : 99.74 <i>E. coli</i> : 98.05

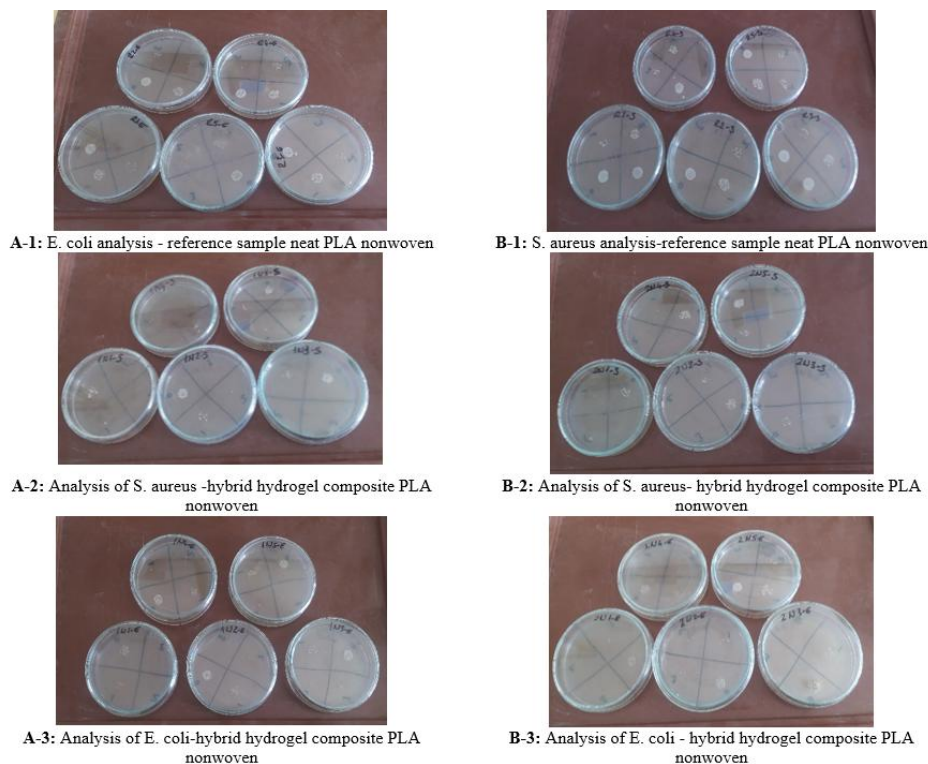


Figure 9. Representative antibacterial activity images of untreated PLA and hydrogel-coated PLA nonwoven fabric samples prepared with Y1 (A-2 and A-3) and Y3 (B-2 and B-3) hybrid hydrogel composite solution

3.7 Degradation Process of Degradable Hybrid Hydrogel Composites

This study utilized PLA fabric as a biodegradable substrate for the development of hybrid hydrogel composites. The degradation behavior of hybrid hydrogel composite fabric samples was evaluated at room temperature under alkaline (0.5 M NaOH) and acidic (HCl solution adjusted to pH 5) conditions. Visual observations indicated complete loss of structural integrity within four days in both media. Nevertheless, it should be emphasized that these degradation experiments were performed under accelerated, non-physiological conditions using 0.5 M NaOH and HCl solution adjusted to pH 5. These environments were intentionally selected to assess the relative chemical stability and degradation tendency of the developed composite system within a shortened timeframe. However, such conditions do not accurately mimic the physiological environment relevant to wound applications. Therefore, the observed degradation behavior should be interpreted as a comparative assessment of chemical stability rather than a direct indicator of *in-vivo* performance. Future studies involving physiologically relevant media (e.g., phosphate-

buffered saline (PBS) and enzymatic conditions) are necessary for a more realistic evaluation. Figure 10 presents representative photographs illustrating the degradation behavior of the hybrid hydrogel composite fabrics after immersion in alkaline (0.5 M NaOH) and acidic (pH 5 HCl) media over a four-day period. Progressive changes in the physical appearance and structural integrity of the samples were observed throughout the degradation period. Visual observations suggested more rapid structural disintegration under alkaline conditions, as evidenced by earlier fragmentation and a greater loss of structural integrity. In contrast, the samples exposed to the acidic medium retained their integrity for a longer period, indicating a comparatively slower degradation process. The observed visual disintegration is consistent with the hydrolytic degradation mechanism of PLA reported in the literature [36, 58]. The hydrogel network likely facilitates water penetration into the composite [8, 33, 56], whereas the PLA phase is expected to undergo hydrolysis of its ester bonds [36], leading to progressive loss of structural integrity. Therefore, the observed differences between acidic and alkaline conditions may be attributed to the combined influence of the hydrogel matrix and the PLA component.

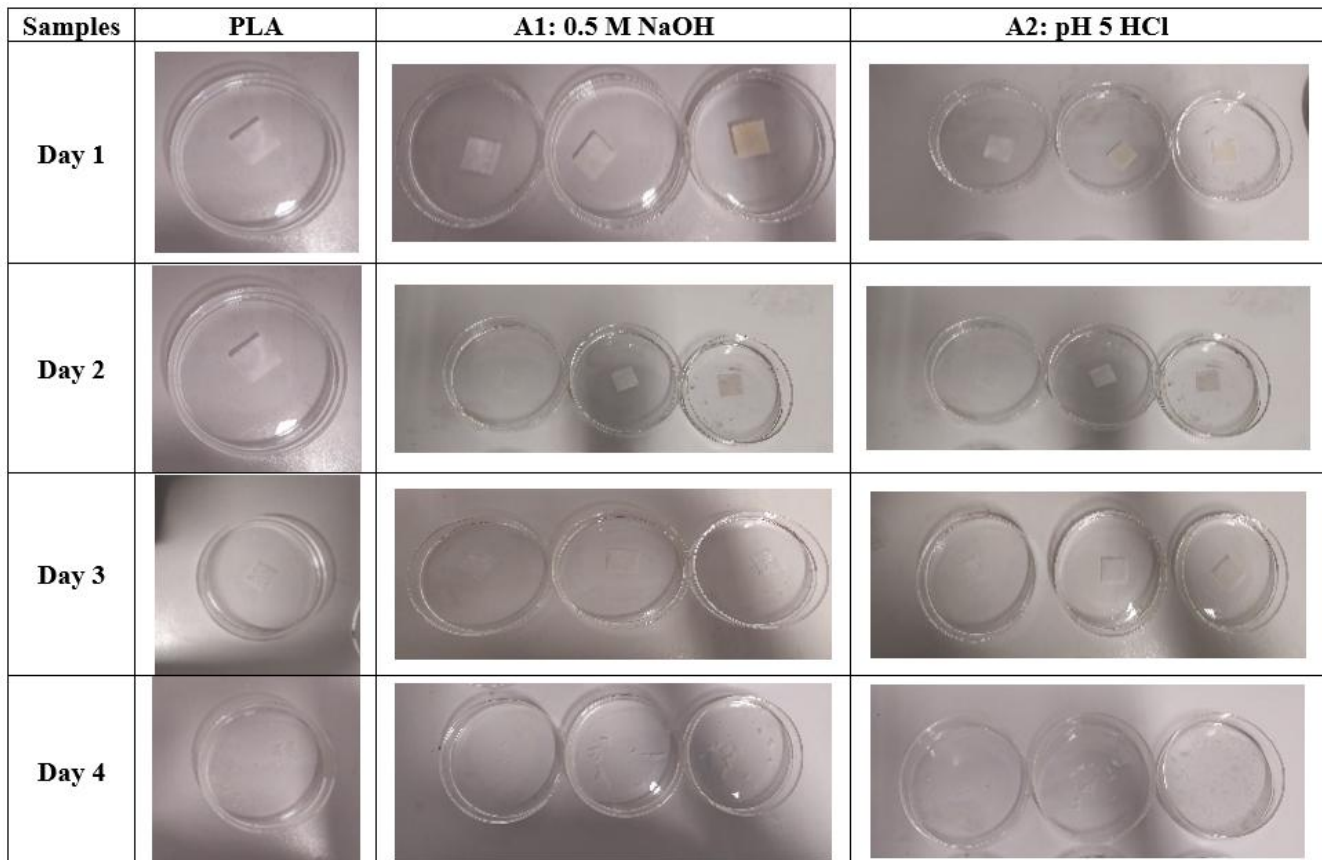


Figure 10: Photographic images of hybrid hydrogel composite fabric samples, A1 and A2; Y series samples (Y1, Y2 and Y3) immersed in 0.5 M NaOH and pH 5 HCl

4. CONCLUSION

This study demonstrated that the developed hybrid hydrogel composites can effectively modify PLA nonwoven fabrics and enhance their hydrophilicity, surface chemistry, and functional properties without severely compromising mechanical integrity. FTIR suggested the incorporation of SA, ZnO, lignin, and CS and indicated possible interactions through polyelectrolyte complex formation, particularly in Y3. SEM and EDS analyses suggested that coating composition influenced fiber morphology and coating continuity. SEM images suggested that Y3 formed a relatively dense and more continuous coating in which ZnO nanoparticles appeared to be embedded. Although coating thickness was not quantitatively measured in the present study, SEM observations indicated that increasing the SA concentration from 0.5% to 1.0% resulted in a more continuous and apparently thicker coating layer on the fiber surface. This increased coating coverage likely reduced the effective pore size and partially blocked the interconnected air transport pathways within the nonwoven structure, leading to lower air permeability values. Therefore, the observed reduction in air permeability is more likely associated with increased coating continuity and partial obstruction of interconnected pore pathways than with coating thickness alone. Tensile strength (69–86 N) and elongation at break (26–30%) were largely preserved after functionalization, indicating that the hydrogel coatings maintained the mechanical integrity of the PLA nonwoven substrate. Only modest formulation-dependent variations in tensile behavior were observed, while overall flexibility remained comparable to that of untreated PLA. Fluid absorbency was markedly improved, particularly in ZnO-containing composites, while air permeability decreased, suggesting a more controlled porous structure that may contribute to moisture management. Y1 (SA/ZnO) achieved strong bacterial reduction, and Y3 (SA/L/CS/ZnO) exhibited the highest antibacterial efficiency among the tested formulations, with more than 98% reduction against *E. coli* and *S. aureus*. The antibacterial behavior appeared to be primarily associated with the presence of ZnO, while CS and lignin may provide additional supportive contributions. While this study demonstrated promising laboratory-scale results, it remains limited to physicochemical and functional characterization under selected experimental conditions, and long-term biocompatibility, cytotoxicity, and in vivo performance have not yet been evaluated. In addition, several mechanistic interpretations regarding swelling, antibacterial behavior, and transport properties were discussed qualitatively based on experimental observations and literature findings rather than direct mechanistic verification. Further optimization of coating uniformity and large-scale processability is also needed to support potential practical application. Despite these limitations, the developed hybrid hydrogel composites exhibited several promising physicochemical and functional characteristics, including enhanced fluid absorbency, antibacterial activity, and maintained mechanical stability. Nevertheless, additional biological investigations, including cytotoxicity, biocompatibility, and in vivo wound healing studies, are required before their suitability and biomedical applicability for wound dressing applications can be established.

Artificial Intelligence Contribution Statement

No artificial intelligence (AI) tools were used in the preparation of this manuscript.

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