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Abstract

The development of innovative food packaging technologies, particularly those capable of monitoring freshness, has become increasingly important in the food industry. This research explores the development of a pH-responsive freshness sensor using polyvinyl alcohol–red cabbage (PVA/RC) nanofibers. The nanofibers are fabricated through the electrospinning technique and meticulously analyzed via scanning electron microscopy, Fourier-transform infrared (FTIR) spectroscopy, and differential scanning calorimetry (DSC). The results underscore the fine structure of the nanofiber matrix, with an average diameter of ~68 nm. FTIR analysis substantiates the presence of anthocyanin compounds from RC within the PVA/RC nanofibers, which confirms the integration of beneficial components into the nanofiber matrix. Moreover, DSC investigations reveal the outstanding thermal properties of PVA/RC, which demonstrates the resilience of the nanofibers to higher temperatures, with a melting point of ~223 °C. Notably, the PVA/RC nanofibers with a 3:1 ratio exhibit excellent thermal stability, although the color change due to pH fluctuations shifts toward transparency. This study lays down the foundation for future exploration and the potential for a diverse array of applications and material enhancements. The findings presented herein open up new opportunities for the use of PVA/RC nanofibers in the development of freshness sensors, heralding a new era in smart food packaging technology.

Keywords: characterization, nanofiber, PVA, red cabbage

Introduction

Smart food packaging refers to packaging solutions that combine advanced technology and innovative design features to enhance the safety, quality, and overall consumer experience of food products. The associated technologies and features include sensors, indicators, QR codes, augmented reality, and other elements that provide real-time and interactive information to both consumers and producers [1–4]. This type of packaging plays a crucial role in the modernization and optimization of food product packaging to meet the demands of modern consumers and address sustainability issues.

Freshness sensors in smart food packaging are a crucial component that serves several important purposes to ensure food safety and consumer satisfaction. These objectives include the reduction of food wastage, the extension of the shelf life of food products, and an increase in consumer trust. These sensors monitor food conditions such as pH, temperature, and humidity and alert consumers or producers when these conditions deviate from safe levels [5, 6]. This helps prevent the consumption of stale or contaminated food and reduces the risk of foodborne illnesses.

The quality of food deteriorates over time. Freshness sensors help maintain and monitor food quality. These sensors provide real-time information to consumers about the quality of food products through the detection of changes in factors such as pH, which helps consumers make informed decisions before consuming or purchasing food products. The presence of freshness sensors in smart food packaging helps build trust between consumers and food producers or retailers. When consumers can rely on accurate information about the freshness of products, they are more likely to trust the brand and its commitment to quality.

Approximately one-third of the total global food production is lost or wasted each year [7]. This alarming statistic indicates that a substantial amount of food, equivalent to ~1.3 billion tons, does not reach consumption. This wastage not only has significant implications for global food security but also bears serious economic ramifications, costing the world economy close to $940 billion annually. The extent of this issue raises concerns about resource management and highlights the need for more sustainable and efficient food management practices on a global scale. Addressing this challenge is crucial for reducing economic losses and mitigating environmental impact, as up to 10% of global greenhouse gas emissions are associated with the production of food that ultimately goes to waste. This information, obtained from the United Nations Environment Programme (UNEP) in 2021, underscores the urgency of implementing strategies to minimize food waste for the benefit of both the economy and the environment [8].

Food waste is a global issue, and freshness sensors help address this problem. They enable consumers to make more informed decisions about food consumption by providing accurate information about food freshness. Consequently, consumers would waste less food, which reduces the overall waste. Freshness sensors contribute to sustainability efforts by reducing food waste and improving supply chain efficiency. Less food waste means fewer resources are wasted in food production, transportation, and disposal, which ultimately reduces the environmental impact of the food industry.

The freshness of meat is a critical factor affecting product safety and quality. Imperfections during storage or transportation can alter the pH level of meat, a key indicator of spoilage and quality decline [9, 10]. pH sensors allow manufacturers and traders to monitor these pH changes in real-time, alerting them about food conditions which enables prompt action to be taken when undesired changes are detected.

Red cabbage (RC) is a kind of cabbage rich in anthocyanins, pigments that are sensitive to color changes depending on the pH of the environment [11, 12]. This makes it a highly suitable component for use in pH sensors, where the color changes produced can serve as effective pH indicators. Polyvinyl alcohol (PVA) in the form of nanofibers is a nanomaterial with unique properties, and it is applied in various fields. The production of nanofibers from PVA is typically achieved through electrospinning, a technique that involves the application of an electric field to draw charged threads of polymer solution into ultrafine fibers [13, 14]. The combination of PVA and RC extract as a pH sensor represents a significant breakthrough in the development of sensory technology, with great potential for various applications, especially in monitoring pH levels of food in different environments and industries [15–17].

Publications on the combination of nanofiber PVA and RC extract for pH sensors are scarce. This scarcity highlights an unexplored area in scientific literature, which presents a unique opportunity for further investigation and contribution to the field. An exploration of the potential advantages and applications of this novel combination could yield valuable insights into the development of innovative pH sensing technologies. In this combination, nanofiber PVA is used as a substrate or matrix. The material has excellent mechanical and chemical properties to support the structure of a delicate sensor. This allows the sensor to provide fast and accurate responses to pH changes.

Wu *et al.* (2022) blended PVA with glycerol as a plasticizer; this required several meticulous preparation steps, including vacuum degassing, casting, and drying to fabricate film-based sensors. Upon gentle agitation, the films exhibited a propensity to fracture within minutes [17]. In the present study, we employed the electrospinning technique to develop nanofibers from PVA and RC extract, a method more straightforward than previously reported methods. Notably, this technique obviated the need for supplementary chemical additives, such as plasticizers, to facilitate the formation of PVA nanofibers. Furthermore, we explored the unique properties of the developed nanofibers, such as the thermal properties, morphology, and stability. We also explored the potential application of these fibers in pH sensing, specifically to monitor the freshness of meat. The unique attributes of the nanofibers offer a promising avenue for the development of innovative sensing technologies with potential applications in the smart food packaging industry.

Methods

Materials. RC (Brassica oleracea) was procured from a local marketplace in Bogor, Indonesia. PVA with a molecular weight of ~60,000 was purchased from Sigma-Aldrich, USA. It was used as the polymer matrix for the nanofiber fabrication. Sodium hydroxide (NaOH) was purchased from Merck KgaA, Darmstadt, Germany, and hydrochloric acid (HCl) was procured from Mallinckrodt Chemicals, USA.

RC extraction. Cabbage leaves (~19 grams) were meticulously cleaned, finely chopped, and subjected to a hot water extraction process. The RC was soaked in 25 ml of boiling water for 1 hour, followed by filtration and subsequent centrifugation at 1500 rpm for 5 min to obtain a concentrated RC extract. The RC extraction method followed a previous study with some modifications [17].

Nanofiber synthesis via electrospinning. The hybrid nanofiber PVA/RC was fabricated via the electrospinning technique which is similar to that of a previous study except for some modifications [18]. The PVA solution, with a concentration of 0.1 mg/ml in distilled water, was prepared before mixing with the RC solution. The concentration of RC extract in the mixture was adjusted to 0 (PVA), 1/16 (PVA/RC 5:1), and 1/4 (PVA/RC 3:1), respectively. The PVA/RC solution was poured into a syringe and ejected with a syringe pump at a pumping rate of 0.5 ml/h. An electrospinning setup equipped with a high-voltage power supply capable of generating up to 30 kV was used for the electrospinning process. A flat collector covered with aluminum foil served as the grounded target for collecting the electrospun nanofibers.

Nano-PSA analysis. The particle size distribution of the RC suspension was analyzed with the use of a laser diffraction Microtrac MRB Nanotrac Wave II across a dynamic range of 0.3 nm to 10 µm. Before analysis, the sample was well shaken to ensure homogeneity, and a refractive index of 1.33 was selected for water.

Ultraviolet–visible analysis. The pH levels of the RC extract were modified within the range of 1 to 14 through the addition of either 1 M HCl or 1 M NaOH. Subsequently, a Shimadzu ultraviolet–visible (UV–vis) spectrophotometer (UV-1700) was used to obtain the UV–vis spectra of these suspensions, under various pH conditions, within the wavelength range of 400 to 800 nm. Images of the samples were taken to correspond with their respective UV–vis spectra.

Fourier-transform infrared analysis. In this analysis, the Fourier-transform infrared (FTIR) technique was used to investigate the functional groups and molecular constituents present in the nanofiber samples. The presence of PVA and RC extract components, as well as their interactions, can be identified through characteristic infrared absorption peaks. Therefore, an Agilent Technologies Cary 630 FTIR was used to record the FTIR spectra of the RC extract, PVA, and PVA/RC solution and nanofiber within the range of 600 to 4000 cm^{-1}.

Scanning electron microscopy analysis. Scanning electron microscopy (SEM) is a powerful technique for visualizing and characterizing the morphological features and structural properties of nanofibers. It enables high-resolution imaging of the nanofiber surfaces to reveal their topography, size, shape, and distribution. The analysis can reveal any structural irregularities present, such as beads, voids, or aggregations within the nanofiber matrix. The JEOL JSM-6510 LA SEM, operating at an acceleration voltage of 10 kV, was used to analyze the microstructures of the PVA and PVA/RC nanofibers. The average nanofiber diameter was determined from SEM images using image analysis software (ImageJ, version 1.49). A minimum of 100 random points were selected from each SEM image of the respective sample to calculate the average diameter [19].

Differential scanning calorimetry analysis. A SETLINE differential scanning calorimetry (DSC) instrument was used to conduct the DSC analysis. The nanofiber samples were encapsulated in aluminum pans with lids, to ensure a hermetic seal that prevented moisture absorption and contamination during the analysis. The DSC analysis was performed under a controlled argon atmosphere to prevent oxidation and ensure a stable baseline. The temperature and heating rate were set according to the specific requirements of the analysis, within a range of 30 °C–250 °C. A typical heating rate of 20 °C/min was adopted to observe thermal transitions. The data obtained were interpreted to understand the influence of the RC extract on the thermal behavior of the PVA nanofibers.

Freshness sensor application. In the freshness sensor evaluation, PVA, PVA:RC 5:1, and PVA:RC 3:1 nanofibers were placed on the surface of fresh meat with a pH of ~5.8. Subsequently, a BYK Gardner colorimeter was used to observe color changes in the sensors, and the L, a, and b parameters were measured. The delta E (ΔE) formula was employed to determine the color changes[20]. ΔE is calculated as follows:

$$\Delta E = \sqrt{\Delta L^2 + \Delta a^2 + \Delta b^2} \quad (1)$$

ΔL is the difference in the L parameter (lightness) between the initial and final measurements, Δa is the difference in the *a* parameter (green to red spectrum) between the initial and final measurements, and Δb is the difference in the *b* parameter (blue to yellow spectrum) between the initial and final measurements. Formula (1) was used to quantify and determine the extent of color change that had occurred in the sensor; the obtained numerical value (ΔE) represents the color change.

Results and Discussion

PSA and UV–vis analysis of RC extract. The RC extract analysis revealed several important findings. First, the measurement of particle size via dynamic light scattering (Figure 1) represents the population of particle sizes. The % channel indicates the population of particles in each size range, while %pass represents the cumulative percentage of particle size distribution. In this curve, the %pass was 70% at a particle size of 400 nm, which meant that 70% of the particles were under 400 nm. As the curve rises exponentially, the larger the particles, the higher the %pass. This implies that large particles were not present in significant amounts, or that

almost all particles were of relatively small sizes. The RC extract used in this research had an average particle size (D_{av}) of 342 nm, which is still in the nanometer range. D_{av} refers to the average diameter, commonly described as the diameter of the cumulative distribution curve. D_{av} is also known as D_{50}, or the particle size with a cumulative distribution of 50%. This result indicated that the extracted components from RC, anthocyanins, existed as nanoparticles in the solution. These nanoparticles have many applications owing to their small size and potential for enhanced solubility. In contrast, other anthocyanin sources such as blueberry and jaboticaba were reported to have average particle sizes of 87.2 µm and 104.9 µm, respectively [21].

UV–vis spectrum analysis via UV–vis spectrophotometry confirmed the color changes observed in the solutions (Figure 2). The variations in wavelength and absorbance values corresponded with the different colors produced in the solutions. This provided quantitative data to complement the visual color changes and confirmed the ability of anthocyanins to respond to changes in pH. The shifting and changes in the intensity of absorbance peaks provided valuable insights into the electronic transitions that occurred within the anthocyanin molecules as a function of pH.

Furthermore, a previous study reported an experiment involving the variation in pH levels from 1 to 14 [22]. The solutions exhibited a wide range of color changes; this indicated that the anthocyanins from RC went through structural changes based on the pH of the immediate environment (Figure 3). The changes in color were attributed to the transformation of anthocyanin molecules into different chemical forms, such as flavylium cations and quinoidal bases, according to the pH [11]. This demonstrated the sensitivity of anthocyanins to changes in the immediate environment and the potential for use as natural pH indicators.

SEM analysis. The SEM results provided significant insights into the morphological characteristics of the synthesized nanofibers (Figure 4). The results revealed distinct differences in the diameter of various nanofiber samples; this highlighted the impact of the PVA/RC ratio on nanofiber morphology. The pure PVA nanofiber had an average diameter of ~215 nm, which was slightly smaller than the 260 nm diameter reported in the study by Fatahian *et al*. (2021) [23].

The PVA/RC nanofibers with a 5:1 ratio had a significantly smaller average diameter (~96 nm). This reduction in diameter demonstrated that the addition of the RC extract led to finer nanofiber formation. However, bead formation was observed in this sample, and the beads observed had an average diameter of 665 nm. The presence of beads in the nanofiber structure could have implications on the mechanical and functional properties. A study conducted by Hashmi *et al*. demonstrated that the active ingredient (Momordica charantia extract) used did not integrate into the nanofiber structure. Instead, it was encapsulated and formed bead-like structures [24].

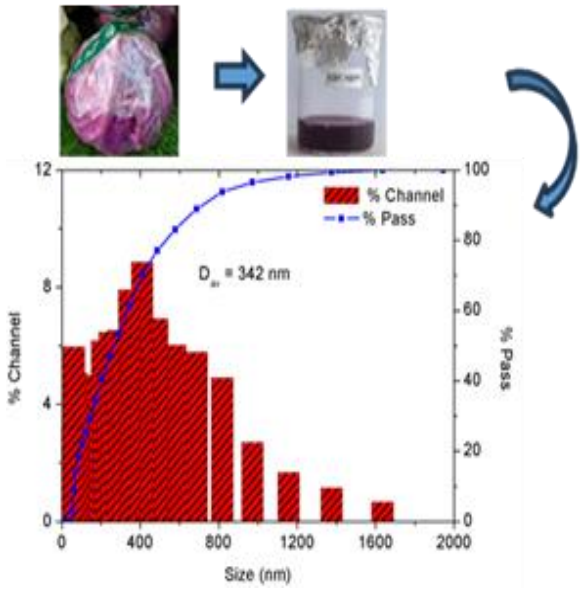


Figure 1. Particle Size Measurement of RC Extract

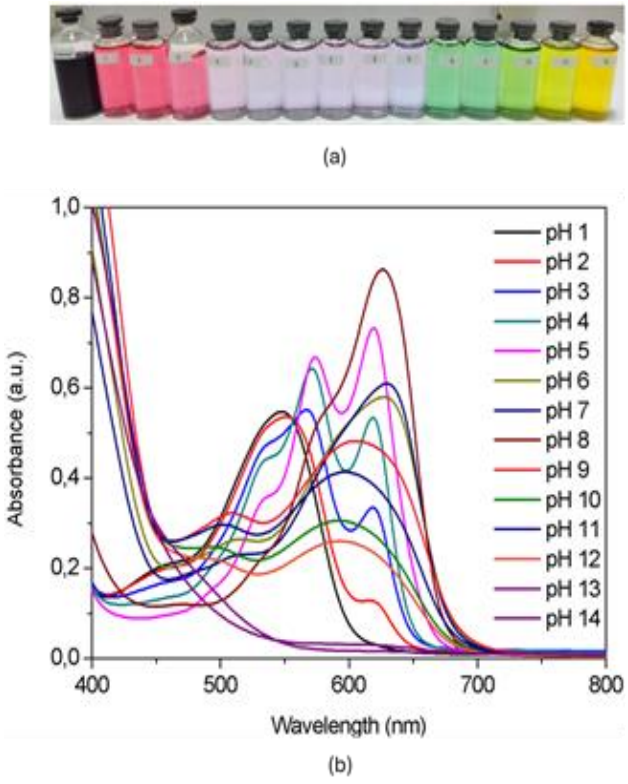


Figure 2. pH-Responsive Test of RC Extract (a) and Absorbance Spectra (b)

The PVA/RC nanofibers with a 3:1 ratio exhibited the smallest average diameter (~68 nm) among the tested samples. This reduction in diameter, compared with both the pure PVA and the 5:1 PVA/RC nanofibers, demonstrated the influence of the RC extract in the formation of finer nanofibers. The smaller diameter implied that this nanofiber sample had a higher surface area, which could result in increased sensitivity when used as a pH sensor. Similar to the 5:1 PVA/RC sample, bead formation was observed in this sample, though the average bead diameter was smaller (~449 nm). The increase in the amount of RC extract correlated with the increase in bead formation.

FTIR. FTIR analysis provided valuable insights into the chemical composition and the functional groups present in the samples (Figure 5). The peaks observed at ~3268 cm^{-1}, 2955 cm^{-1}, and 1083 cm^{-1} (Figure 5a) offered critical insights into the molecular composition of the sample solutions. These peaks corresponded to stretching vibrations of specific functional groups within the sample solution. The peak at 3268 cm^{-1} corresponded to the stretching of hydroxyl groups (-OH); this indicated the presence of compounds with hydroxyl functional groups, which are commonly found in phenolic compounds.

Additionally, the peak at 2955 cm^{-1} corresponded to the stretching vibrations of carbon-hydrogen bonds (-CH) in PVA. This indicated the presence of organic compounds containing the C-H bond. This finding aligned with peaks observed in previous studies [25]. Furthermore, the peak at 1083 cm^{-1} indicated stretching vibrations of the carbon–oxygen (C–O) bond. This bond is prevalent in various organic compounds and is commonly associated with the presence of carbonyl or ether groups [26].

For the RC extract solution, a peak was observed at 1636 cm^{-1}. This peak is associated with the stretching vibrations of carbon–carbon double bonds (C=C) found in aromatic compounds [11]. This indicated the presence of phenolic compounds, which are characterized by aromatic structures and are known for their significant antioxidant properties.

The FTIR spectra of the nanofiber structures (Figure 5b) revealed peaks that indicated a gradual reduction in the intensity of hydroxyl groups with an increase in the concentration of RC extract added. This suggests a reduction in the amount of hydroxyl groups present in the PVA owing to the addition of RC extract and evaporation at the electrospinning stage. This reduction in the amount of hydroxyl groups is significant for pH sensor applications, as PVA nanofibers become less hydrophilic and more resistant to damage when interacting with hydrogen ions in water. Additionally, the peaks associated with -CH bonds and C–O bonds showed a slight shift at wavenumbers 2923 cm^{-1} and 1088 cm^{-1}, respectively.

DSC. The DSC results provided significant insights into the thermal properties of the PVA/RC nanofibers (Figure 6). First, on exposure to heat, an endothermic peak was observed at ~120 °C. This indicated the evaporation of water (Remiš *et al.*, 2021). This is a common occurrence in polymer materials, as any residual moisture or solvents are released when heated.

A distinct melting event occurred at a melting point (T_m) within the range of 219 °C–224 °C. Notably, as the concentration of RC extract increased, the melting point decreased. This indicated that the presence of the RC extract in the nanofibers had a plasticizing effect, which reduced the energy required for melting. A similar observation was made by Remiš *et al.*, where the melting point of PVA was ~218 °C and slightly shifted to a lower temperature with the addition of nanodiamonds [27].

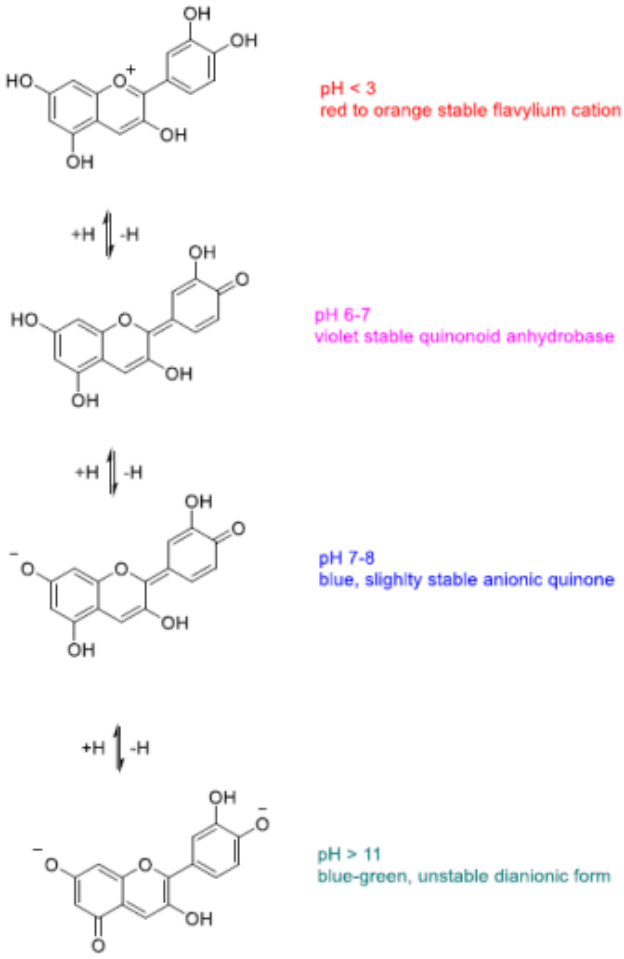


Figure 3. Changes in the Chemical Structure of the Anthocyanin Molecule as the pH Shifts from Acidic to Neutral to Basic [22]

After the melting event, the DSC curve indicated a crystallization process at a crystallization temperature (T_c) within the range of 182 °C–194 °C. Similar DSC results were obtained in another study on the melting and crystallization of PVA samples [28]. This illustrates the propensity of the nanofibers to reorganize and form crystalline structures upon cooling. However, a higher concentration of the RC extract led to a reduction of the crystallization temperature. This implies that a higher RC extract content could reduce the tendency to form crystalline structures upon cooling.

However, the DSC analysis also demonstrated that the addition of the RC extract only had a minor influence on the melting point and crystallization temperature of PVA. This indicates that the RC extract had a limited impact on the thermal properties of PVA. This finding demonstrates the thermal stability of PVA even in the presence of the RC extract. The minimal change in the melting point and crystallization temperature implies that the overall thermal characteristics of PVA were largely unaffected by the addition of the RC extract. Such stability is advantageous for applications where the preservation of the original thermal properties of PVA is crucial.

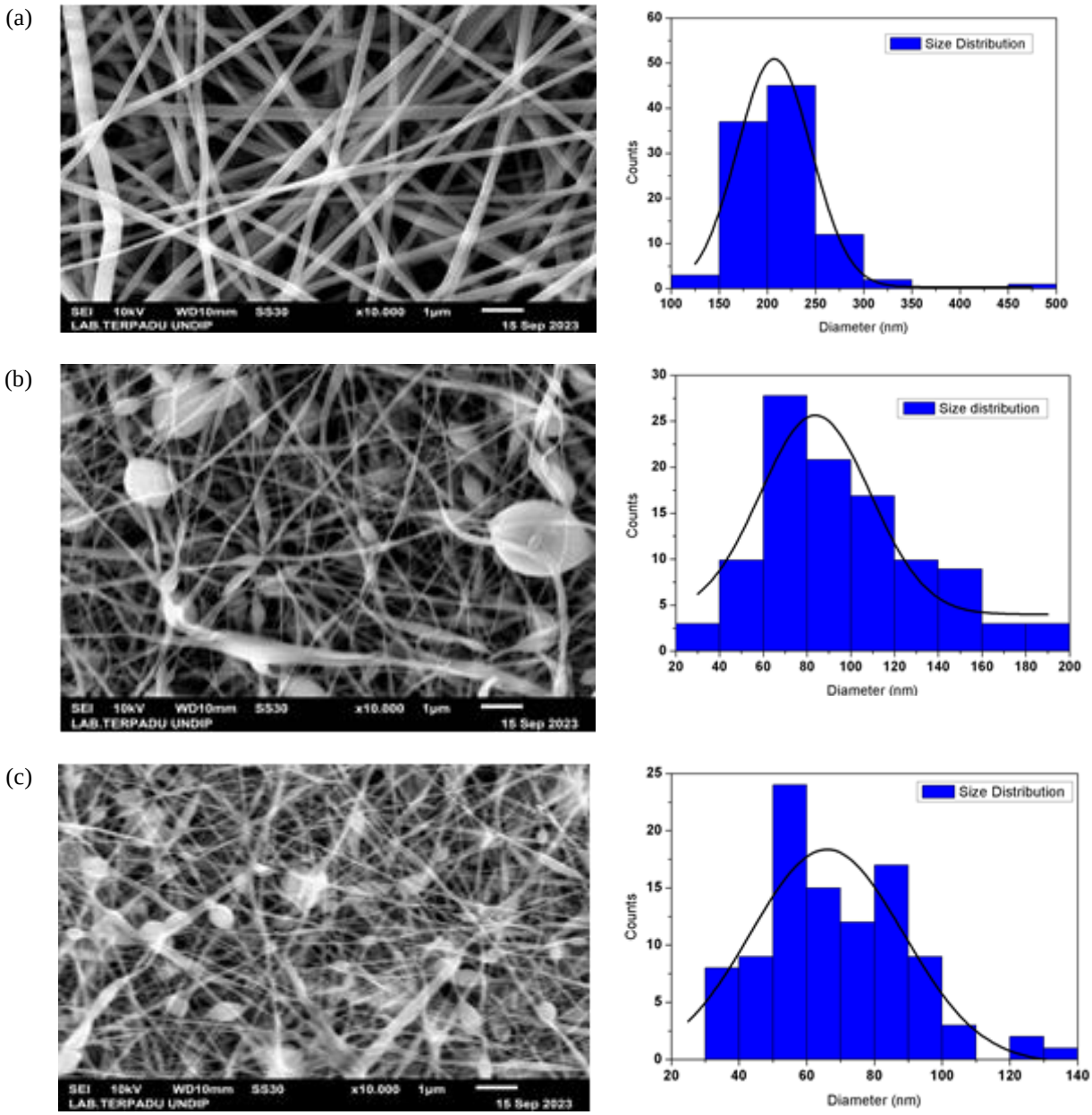
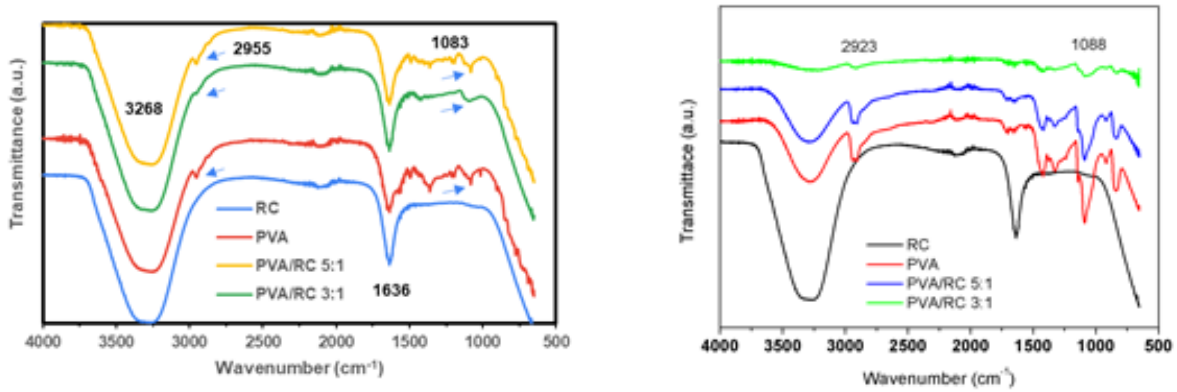


Figure 4. Morphologies of (a) PVA, (b) PVA/RC 5:1, (c) and PVA/RC 3:1 Nanofibers



(a) (b)

Figure 5. FTIR Spectra of RC, PVA, and PVA/RC in (a) Liquid (b) and Fiber Form

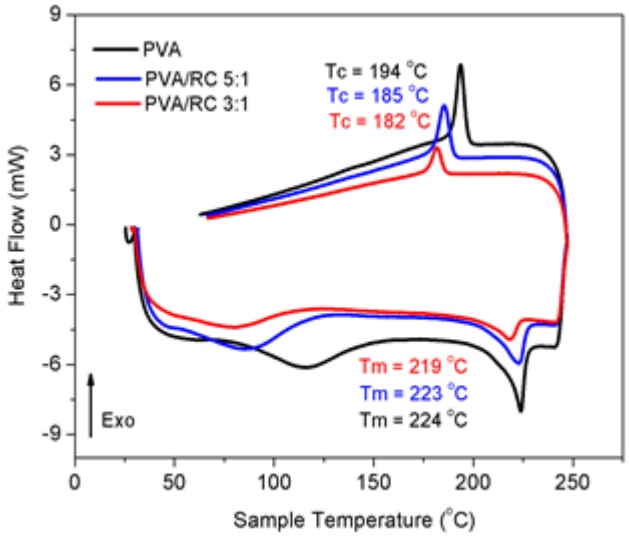


Figure 6. DSC curves of PVA, PVA/RC 5:1, and PVA/RC 3:1

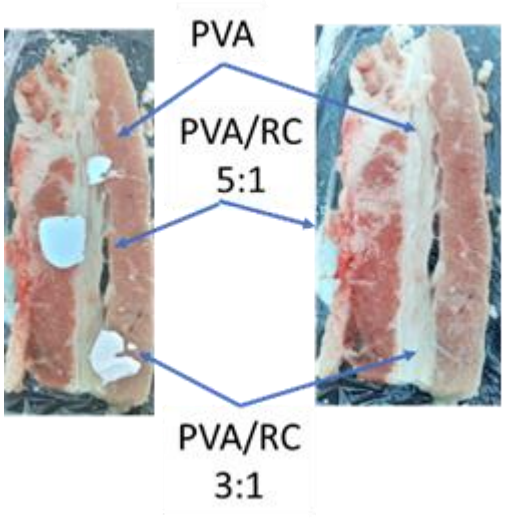


Figure 7. pH Sensor Application on Fresh Meat Surface

Table 1. Color Parameters of Samples Before and After Application

Sample	L	a	b	ΔE
Before application				
PVA	97.20	1.36	1.05	-
PVA/RC 5:1	92.83	1.24	−1.59	5.11
PVA/RC 3:1	93.04	2.04	−0.98	4.68
After application				
PVA	NA	NA	NA	NA
PVA/RC 5:1	57.64	−0.67	5.65	39.90
PVA/RC 3:1	53.88	2.83	1.67	43.30

Table 2. Comparative Study of pH-Responsive Materials

Study	Composition	Fabrication Method	Product	Key Findings
This study	PVA/RC	Electrospinning	Nanofiber	• Nanofiber diameter was 68 nm. • pH-responsive (natural dye) • Thermal stability up to 223 °C
[30]	ι-carrageenan/RC and ι-carrageenan/butterfly pea	Chemical mixing	Film	• Edible material • pH-responsive (natural dye) • Low thermal stability
[31]	Nylon 6 polymer/synthetic dye	Electrospinning	Nanofiber	• Nanofiber diameter was in the range of 40–50 nm. • pH-responsive (synthetic dye) • Thermal stability was tested at 100 °C
[32]	Chitosan/PVA/RC/ sodium tripolyphosphate (STPP)	Solvent casting	Film	• pH-responsive (natural dye) • Additional chemicals were needed. STPP was used as the cross-linking agent. • Thermal stability test was not provided.
[17]	PVA/RC	Chemical mixing	Film	• pH-responsive (natural dye) • Easily dissociates after gently shaking • Thermal stability test was not provided.

Figure 8. Schematic of the Developed Freshness Sensor Mechanism

Freshness sensor test. In this experiment, PVA, PVA/RC 5:1, and PVA/RC 3:1 nanofibers were placed on fresh meat, and color changes were observed (Figure 7). The nanofibers became transparent, and it was difficult to distinguish color changes. Therefore, a colorimeter was used to determine the color coordinates; the results are presented in Table 1.

The transformation of the nanofibers from white to transparent was due to the strong hydrophilic nature of PVA, as evidenced by the prominent hydroxyl groups identified in the FTIR analysis. These hydroxyl groups were actively bound to hydrogen ions present in water on the surface of the meat samples. However, the PVA/RC 3:1 nanofibers exhibited enhanced resilience compared with the other samples after extraction from the substrate surface and drying. This improved durability was due to the lower concentration of hydroxyl groups, which made the nanofiber sample less hydrophilic and more resistant to damage. These results highlight the potential benefits of incorporating additives, such as glutaraldehyde as a crosslinker, into the nanofiber production process. Numerous studies have demonstrated the efficacy of such additives in enhancing the mechanical and structural properties of nanofibers, providing valuable insights for optimizing the performance of the nanofibers in various applications [18, 28, 29].

Our study demonstrated superior thermal stability, finer nanofiber structure, ease of use, and the utilization of environmentally friendly materials compared with other studies (Table 2). These factors collectively highlight our study as a significant advancement in the field of pH-responsive material fabrication. The findings of our study surpass the findings of previous studies, as we achieved a remarkable thermal stability of up to 223 °C. This improved thermal stability expands the potential applications of pH-responsive materials to high-temperature environments. The production of nanofibers with diameters as small as 68 nm demonstrates precise control over fiber morphology. This level of precision is crucial for applications that require fine structures. The utilization of natural dyes for pH-responsive materials aligns with the growing demand for sustainable and environmentally friendly products. Our study contributes to reducing the carbon footprint associated with material fabrication processes by avoiding the use of synthetic dyes and other chemical additives.

The proposed mechanism of action of the developed freshness sensor (Figure 8) is based on the dynamic changes in the pH of meat during storage. These changes involve a transition from an acidic to a more alkaline environment, driven by a complex interplay of biochemical processes. The factors that contribute to these pH variations include the breakdown of glycogen, proteolysis, and the production of acidic or alkaline metabolites by microorganisms, all of which play crucial roles in meat spoilage.

The utilization of anthocyanin-containing PVA nanofibers as meat freshness sensors is based on the remarkable ability of anthocyanins to undergo detectable color changes in response to fluctuations in pH. This pH-responsive behavior makes anthocyanin-containing PVA nanofibers well-suited for real-time monitoring of meat freshness. The incorporation of anthocyanins into the nanofibers has been demonstrated to induce pronounced and visually detectable color changes in response to alterations in the pH of the meat, which enables consumers to easily distinguish the freshness indicator on the meat packaging. This mechanism is important, as it ensures consumer safety via the provision of a clear and intuitive means for the assessment of the freshness and quality of packaged meat products.

Conclusion

The synthesis of PVA/RC nanofibers via the electrospinning technique yielded favorable results. The visual color changes in the RC extract, substantiated by the shift in the UV–vis spectrum, confirmed that the RC extract could respond to changes in pH. The nanoparticle sizes of the particles suspended in the RC extract solution, as confirmed by the PSA analysis, contributed to higher sensitivity. The PVA/RC 3:1 nanofibers with an average diameter of 68 nm, as illustrated by the SEM images, demonstrated the significant potential for freshness sensor applications. The relatively small particle sizes resulted in a higher surface area, which maximized the interaction between the sensing element and the product.

FTIR analysis revealed the presence of anthocyanin, from RC, within the PVA/RC nanofibers; this demonstrated that beneficial components were incorporated into the nanofibers. Furthermore, the addition of the RC extract did not significantly alter the thermal properties of the PVA nanofibers, as confirmed by the DSC thermogram. The PVA/RC 3:1 sample was the most stable, although the change in color shifted toward transparency.

The results demonstrate the potential role of additives in maintaining and enhancing the performance of nanofibers. This study lays down the foundation for further exploration and development and provides the possibility for a wider range of applications. The findings offer exciting prospects for the utilization of PVA/RC nanofibers in freshness sensor applications and provide new opportunities for the development of smart food packaging.

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Authors' Note

The authors declare that there is no conflict of interest regarding the publication of this article. Authors confirmed that the paper was free of plagiarism.

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