Enhanced durability and tribological performance of polyvinyl alcohol/layered double hydroxide/tannic acid composites under repeated swelling cycles

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Abstract: In recent years, the exploration of polyvinyl alcohol (PVA) composites has garnered significant attention due to their versatility applications in aqueous environments. However, despite their promise, neat PVA exhibit limitations such as significant mechanical degradation under repeated swelling cycles. This study investigates the durability and tribological performance of polyvinyl alcohol (PVA) composites reinforced with nickel-iron layered double hydroxide (LDH) and tannic acid (TA) under repeated swelling cycles. Building on previous research that explored composite preparation and initial characterization, this research emphasizes the effects of cyclic swelling on wear resistance, friction behavior, and mechanical properties. Tribological tests were conducted to evaluate the coefficient of friction (COF) and wear rate before and after multiple swelling cycles, alongside tensile strength and strain measurements. The results revealed that the PVA/TA2/LDH2 composite, containing the highest additive content, exhibited the lowest wear rate of 11.52×10^{-5} mm³/Nm after 3 swelling cycles, demonstrating superior resistance to material degradation. Although PVA/TA2/LDH1 exhibited a slightly lower COF, its wear rate was higher due to reduced reinforcement. Compared to neat PVA, which showed a COF increase from 0.45 to 0.53, the PVA/LDH/TA composites retained their tribological stability, with only a marginal increase in COF and wear rate. Similarly, tensile strength of PVA/TA2/LDH2 decreased by only 11% after 3 cycles (from 33.3 MPa to 30 MPa), while neat PVA experienced a 25.5% reduction (from 30 MPa to 22.5 MPa). These findings highlight the potential of PVA/LDH/TA composites for applications in aqueous environments, offering significantly enhanced long-term performance and reliability.

Keywords: polyvinyl alchohol; LDH; tannic acid; swelling test

1. Introduction

The growing concern for environmentally sustainable materials has brought significant interest in developing biodegradable polymers as alternatives to traditional synthetic polymers [1], [2], [3], [4], [5]. The excessive use of non-degradable materials has led to severe ecological issues, including plastic pollution and the accumulation of hazardous waste in ecosystems [6]. To address these challenges, researchers and industries have focused on creating materials that can decompose naturally, reducing their environmental impact [7], [8]. The importance of biodegradable polymers lies in their ability to bridge the gap between technological advancement and environmental responsibility [9], [10], [11]. These materials not only address pressing ecological concerns but also open new pathways for sustainable innovation in industries reliant on polymers. From reducing

landfill waste to enabling eco-friendly product designs, biodegradable polymers contribute significantly to achieving long-term environmental goals [12].

Biodegradable polymers offer a promising solution, as they not only alleviate waste management problems but also align with global efforts to promote sustainable development and reduce dependency on fossil-based resources [13]. Furthermore, these materials align with the principles of a circular economy, driving their adoption in diverse fields such as biomedical applications, packaging, and lubrication systems [14]. Among these biodegradable materials, composites have gained attention due to their ability to combine the properties of a polymer matrix with the enhancements provided by various additives or reinforcements. This strategy not only improves performance but also broadens the utility of biodegradable materials in areas previously dominated by synthetic polymers.

Despite their potential, biodegradable polymers often exhibit inherent limitations, such as low mechanical strength, thermal instability, and suboptimal tribological properties, which restrict their use in demanding environments [15], [16]. To overcome these challenges, researchers have focused on incorporating functional reinforcements into polymer matrices [17], [18], [19], [20]. Additives such as nanomaterials, polyphenols, and inorganic fillers can enhance mechanical properties, thermal stability, and tribological performance, making polymer composites a versatile solution for diverse applications [21], [22].

Polyvinyl alcohol (PVA) has gained significant attention in recent years as a versatile polymer matrix for developing advanced composites due to its excellent film-forming ability, biocompatibility, and mechanical properties [23]. However, its hydrophilic nature and moderate tribological performance limit its applications in humid or aqueous environments, where water absorption can lead to swelling, plasticization, and deterioration of mechanical and tribological properties [24], [25]. To address these challenges, reinforcement with functional fillers such as layered double hydroxides (LDHs) and tannic acid (TA) has emerged as a promising strategy.

Nickel-iron layered double hydroxide (LDH), a type of LDH, is an inorganic material known for its exceptional mechanical strength, high thermal stability, and ability to enhance the structural integrity of polymer matrices [26], [27]. LDHs have also been reported to improve wear resistance and reduce friction in polymer composites, making them attractive for tribological applications [28]. LDHs are synthesized through various methods, each offering unique advantages depending on the desired properties and applications. Common synthesis techniques include co-precipitation, where metal salts are precipitated in an alkaline medium; hydrothermal synthesis, which involves high-pressure and high-temperature conditions to improve crystallinity; and sol-gel methods, known for producing uniform and high-purity materials [29].

Meanwhile, tannic acid, a naturally occurring polyphenol, offers multifunctional properties, including antioxidant activity, excellent adhesion to polymer matrices, and enhanced hydrophobicity through hydrogen bonding and cross-linking [30]. The synergistic effect of LDH and TA in PVA-based composites can potentially mitigate the drawbacks associated with swelling while improving the composite's overall durability and functionality. Figure 1 demonstrated the effective distribution of LDH within the PVA matrix from the previous work, where LDH's high surface area and layered structure aid in evenly distributing mechanical loads and increasing water contact angle. These findings serve as the foundation for the current study, where the long-term performance of such composites under conditions simulating real-world applications, such as repeated exposure to moisture or swelling cycles, remains largely unexplored [31].

This study aims to investigate the effects of repeated swelling cycles on the tribological performance and mechanical properties of PVA/LDH/TA composites. It is hypothesized that the addition of

LDH and TA can significantly enhance the durability and tribological performance of the composites by reducing water-induced degradation. The study evaluates changes in the coefficient of friction, wear scar morphology, and tensile strength after multiple swelling cycles. These findings will provide valuable insights into the potential of PVA/LDH/TA composites for applications in humid environments, such as biomedical devices, coatings, and lubrication systems.

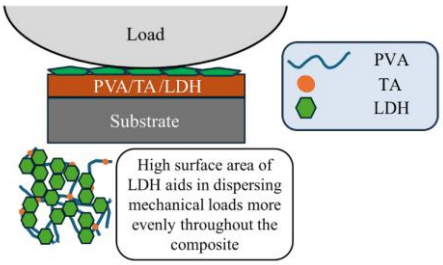


Figure 1. Schematic representation of the distribution of TA and LDH within the PVA matrix and its role in enhancing mechanical and tribological properties. Reproduced under the terms of the CC-BY Creative Commons Attribution 4.0 International license (https://creativecommons.org/licenses/by/4.0) [31]. Copyright [2024], The Authors, published by IOP Publishing.

2. Material and methods

2.1 Material

Polyvinyl alcohol (PVA) powder (molecular weight 146,000–186,000, 99+% hydrolyzed) was purchased from Sigma Aldrich. Nickel (II) nitrate hexahydrate ($Ni(NO_3)_2{\cdot}6H_2O$) and urea (NH_2CONH_2) were supplied by Katayama Chemical Co., Ltd. Iron (III) nitrate nonahydrate and tannic acid were purchased from Daejung Chemicals & Metals Co., Ltd. Ammonium fluoride (NH_4F) was provided by Showa Denko K.K. All chemicals were used as supplied, without additional purification.

2.2 Sample preparation

The composition of each sample is summarized in Table 1. The values are expressed in weight percent. The composite samples were prepared using a three-step method adapted from the previous work [31]. First, PVA was dissolved in distilled water at 100°C under continuous stirring until a homogeneous solution was obtained. Next, TA and LDH were sequentially added to the PVA solution, and the mixture was stirred to ensure proper dispersion of the additives. Finally, the prepared solution was subjected to ultrasonication using a probe sonicator to enhance the uniform distribution of LDH and tannic acid within the PVA matrix. The resulting solution was cast into molds and left to dry at controlled conditions to form composite films with consistent thickness and quality.

Table 1. Composition of composite coating films

Sample	Concentration (wt%)			
	DI Water	PVA	LDH	TA
PVA	90	10	0	0
PVA/TA0.5/LDH1	88.5	10	0.5	1
PVA/TA1/LDH1	88	10	1	1
PVA/TA1/LDH2	87	10	1	2
PVA/TA2/LDH2	86	10	2	2

2.3 Sweliing, tensile and tribological test

The prepared samples of the composite films were subjected to a series of tests to evaluate their mechanical and tribological properties after exposure to water. Initially, the samples were immersed in distilled water for 24, 48, and 72 hours at room temperature to simulate swelling conditions. During the immersion period, the swelling ratio of the samples was periodically measured. Specifically, after 12 hours, the samples were removed from the water, gently wiped to eliminate surface water, and weighed to determine the swollen weight. The swelling ratio was calculated using the equation (1).

$$\text{Swelling Ratio } (\%) = [(W_s - W_d)/W_d] \times 100\% \quad (1)$$

where W_s is the swollen weight and W_d is the dry weight of the sample.

The samples were then returned to the distilled water to continue swelling. After each 24-hour immersion period, the samples were removed, gently wiped to eliminate surface water, and dried in an oven at 60°C for 24 hours to ensure consistent moisture removal. This process of immersion and drying was repeated for each cycle, with one cycle comprising 24 hours of immersion followed by 24 hours of drying. After completing the designated number of cycles (1, 2, or 3 cycles), the samples were subjected to tensile testing using a universal testing machine (AGS-X, SHIMADZU, Japan) at a speed of 12 mm/min.

Subsequently, the tribological properties of the samples were evaluated using a ball-on-disk tribometer (POD-FM406-10NT, Fu Li Fong Precision Machine, Kaohsiung, Taiwan). The testing parameters, including normal load, sliding speed, and test duration, were consistent with those used in the previous work [31]. Figure 2 shows the schematic diagram of the methodology for this study.

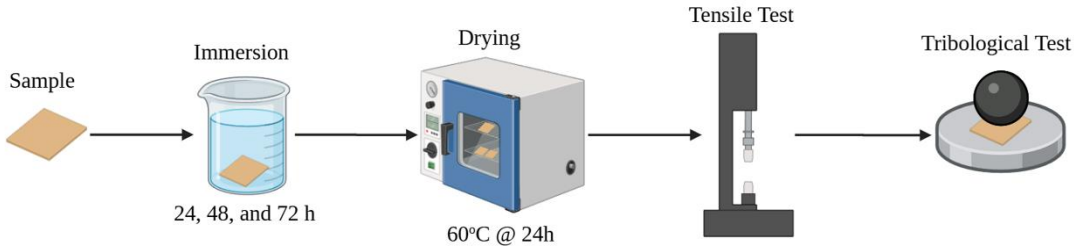


Figure 2. Schematic diagram of the immersion methodology

3. Results and discussion

3.1 Swelling test

Figure 3 shows the swelling ratio of PVA/LDH/TA composites as a function of time. The neat PVA sample exhibited the highest swelling ratio, reaching approximately 110 g/g after 72 hours of immersion. This high-water uptake is due to the hydrophilic nature of PVA, which contains numerous hydroxyl (-OH) groups capable of forming hydrogen bonds with water molecules [32]. However, the incorporation of tannic acid (TA) and LDH significantly reduced the swelling ratio. For the composites containing TA and LDH, the swelling ratio consistently decreased with increasing TA content and LDH loading. For instance, PVA/TA0.5/LDH1 showed a swelling ratio of 80 g/g at 72 hours, a reduction of ~27% compared to neat PVA. Further reductions were observed in PVA/TA1/LDH1 and PVA/TA2/LDH1, with swelling ratios of 70 g/g and 60 g/g, respectively. Composites with higher LDH content (LDH2) exhibited even lower swelling ratios,

with PVA/TA1/LDH2 and PVA/TA2/LDH2 reaching 50 g/g and 40 g/g, respectively, representing reductions of ~55% and ~64% compared to neat PVA.

The decrease in swelling ratio can be attributed to two primary mechanisms. Firstly, TA forms strong hydrogen bonds with the hydroxyl groups in PVA, reducing the availability of free -OH groups to interact with water molecules, thereby increasing the hydrophobicity of the network [33]. Secondly, LDH contributes to the formation of a denser, layered structure that acts as a physical barrier to water diffusion [34], [35]. This effect is more pronounced with higher LDH content, as seen in the LDH2 samples, where the swelling resistance was significantly improved. The swelling behavior also revealed that all samples exhibited rapid water absorption within the first 12 hours, followed by a gradual leveling-off as equilibrium was reached. The presence of TA and LDH slowed the initial swelling rate, indicating improved resistance to water absorption even during the early stages of immersion.

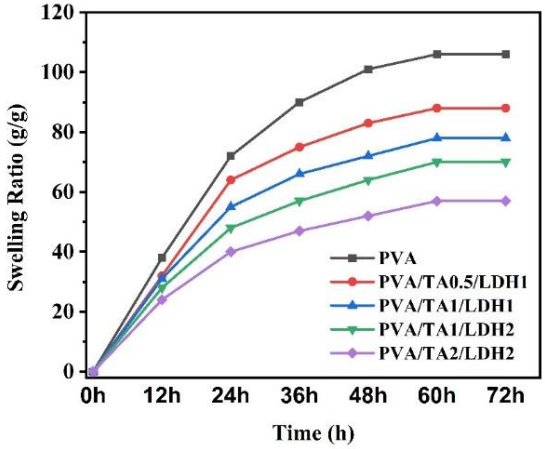


Figure 3. Swelling ratio of PVA/LDH/TA composites as a function of time

3.2 Tensile test

Figure 4 shows the tensile strength and tensile strain percentage change of PVA/LDH/TA composites under different swelling cycles. The neat PVA sample exhibited the most significant reduction in tensile strength after three swelling cycles, decreasing from 30 MPa to approximately 22.5 MPa, corresponding to a 25% loss. This substantial reduction underscores the hydrophilic nature of PVA and its susceptibility to mechanical degradation upon water absorption. In contrast, the addition of TA and LDH significantly improved the composites' ability to retain tensile strength, with performance enhancement depending on the TA content and LDH loading. For instance, the PVA/TA0.5/LDH1 composite showed a moderate decrease in tensile strength, from 30 MPa (no swelling) to 25 MPa (after three cycles), representing a 16% reduction. Composites with higher TA and LDH content, such as PVA/TA1/LDH1 and PVA/TA2/LDH1, demonstrated further improvements, with tensile strength reductions of only 13.8% and 14.2%, respectively, after three cycles. Notably, samples with higher LDH content (PVA/TA2/LDH2) showed the most resilience, retaining over 90% of their initial tensile strength.

The improved mechanical stability can be attributed to the synergistic effects of TA and LDH. Tannic acid forms strong hydrogen bonds with PVA, enhancing the polymer network's density and crosslinking, which reduces the susceptibility to water-induced weakening [36], [37]. Furthermore, the layered structure of LDH acts as a reinforcing agent, providing mechanical stability and resistance to deformation during swelling and drying [38]. The neat PVA sample exhibited the largest increase in tensile strain, reaching nearly 50% after three cycles, indicating significant plasticization and softening due to water absorption. In contrast, composites with TA and LDH

exhibited much lower strain increases. For example, PVA/TA1/LDH2 and PVA/TA2/LDH2 maintained tensile strain changes below 20%, demonstrating their ability to resist plastic deformation and maintain structural integrity.

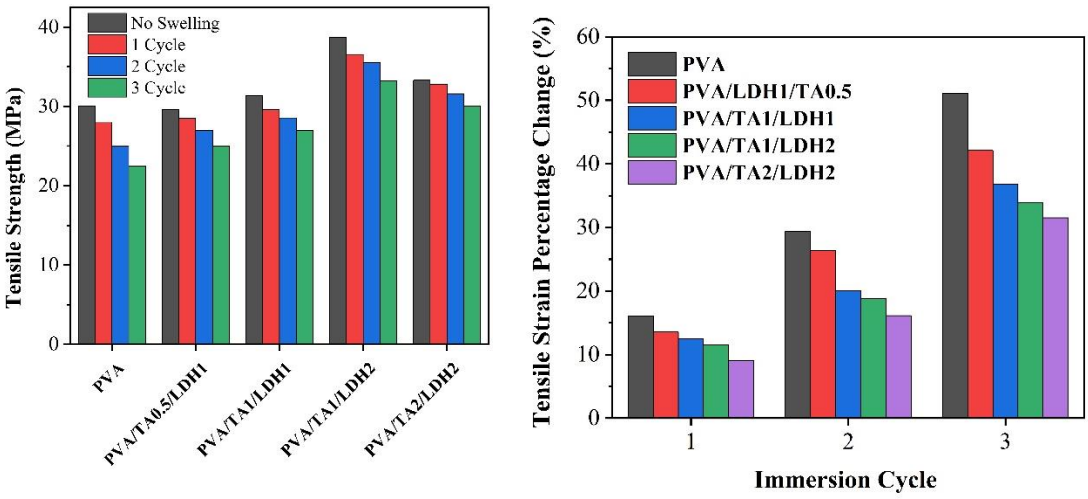


Figure 4. Tensile strength-strain curve of all samples

3.3 Tribological test

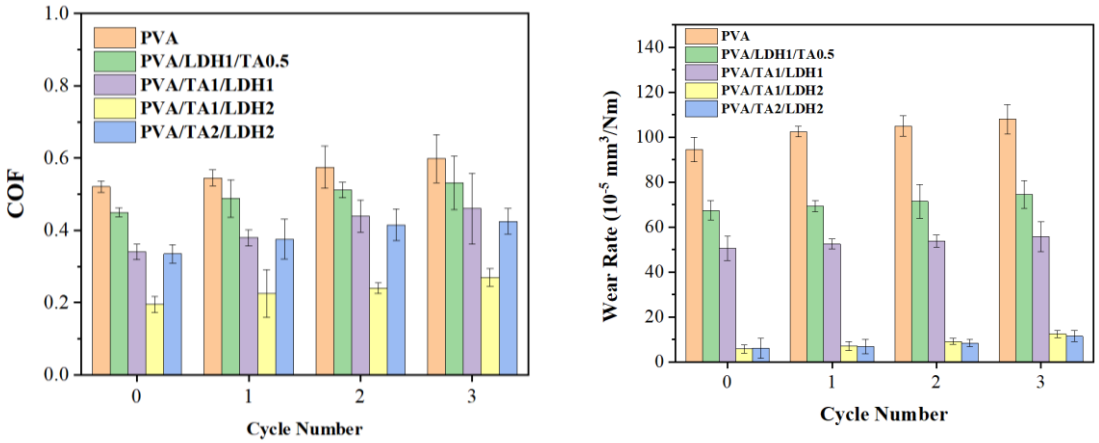
Figure 5 (a and b) presents the coefficient of friction (COF) and wear rate of PVA/ LDH/TA composites as a function of the number of swelling cycles. The neat PVA sample exhibited the highest COF and wear rate across all conditions. Before swelling (0 cycles), the COF for PVA was approximately 0.52, and the wear rate was 94.5 × 10^{-5} mm³/Nm. Both parameters increased after each swelling cycle, with the COF rising to 0.6 and the wear rate reaching 108 × 10^{-5} mm³/Nm after three cycles.

The incorporation of TA and LDH significantly reduced both COF and wear rate, demonstrating the synergistic effects of these additives. For the composite PVA/LDH1/TA0.5, the COF initially measured at 0.45 and increased to 0.53 after three cycles, while the wear rate rose from 67.5 × 10^{-5} mm³/Nm to 74.5 × 10^{-5} mm³/Nm. The composites with higher TA and LDH content, such as PVA/TA1/LDH2 and PVA/TA2/LDH2, showed superior tribological performance. For these samples, the COF remained below 0.5 even after three cycles, and the wear rate was consistently below 56 × 10^{-5} mm³/Nm. These results indicate that higher TA and LDH content effectively enhance the durability of the composites under repeated swelling conditions.

Among the composites, PVA/TA2/LDH2, the sample with the highest additive content, exhibited the lowest wear rate after three immersion cycles, measuring only 11.5 × 10^{-5} mm³/Nm. This remarkable wear resistance is attributed to the synergistic effects of TA and LDH at higher concentrations, which enhance the matrix's mechanical stability and form a protective barrier during tribological contact. TA strengthens the interfacial bonding within the polymer matrix and increases hydrophobicity, limiting water-induced weakening during swelling. Meanwhile, LDH contributes to reinforcement and acts as a solid lubricant, further reducing material loss.

Interestingly, PVA/TA2/LDH1, despite having a lower COF compared to PVA/TA2/LDH2, exhibited a slightly higher wear rate. This discrepancy can be explained by the differences in LDH content. The higher LDH content in PVA/TA2/LDH2 provides greater reinforcement and wear resistance due to the denser layered structure and better load-bearing capacity. However, the additional LDH layers may marginally increase the COF due to a higher frictional interface between

the contacting surfaces. In contrast, the lower LDH content in PVA/TA2/LDH1 results in reduced interfacial friction, as reflected by the slightly lower COF, but provides less reinforcement, leading to a higher wear rate compared to PVA/TA2/LDH2.



(a) (b)

Figure 5. Friction coefficient (a) and wear rate (b) of all samples on disks in ball-on-disk tribology tests after immersion

4. Conclusion

This study highlights the significant improvements in durability and tribological performance achieved by incorporating LDH and TA into PVA composites under repeated swelling cycles. The results demonstrate that the synergistic effects of TA and LDH effectively mitigate the detrimental impacts of water-induced swelling, maintaining the composites' mechanical integrity and wear resistance. Among the tested samples, PVA/TA2/LDH2, containing the highest additive content, exhibited the best overall performance, with the lowest wear rate of 11.52 × 10^{-5} mm³/Nm after three swelling cycles and only a 11% reduction in tensile strength (from 33.3 MPa to 30 MPa). This superior performance is attributed to the strong interfacial bonding provided by TA, which enhances hydrophobicity and network stability, and the reinforcing effect of LDH, which acts as a solid lubricant and improves the composite's structural durability.

The scientific justification for this research lies in addressing the critical limitations of PVA-based composites, particularly their susceptibility to water-induced degradation in humid or aqueous environments. By leveraging the complementary properties of LDH and TA, this study offers a systematic approach to improving the mechanical stability, wear resistance, and tribological performance of biodegradable materials under challenging conditions. These findings provide valuable insights into the design of advanced composites for applications where moisture resistance is crucial, such as biomedical devices, packaging materials, and lubrication systems.

Overall, this study establishes that PVA/LDH/TA composites, particularly those with higher TA and LDH content, are promising candidates for applications requiring enhanced mechanical stability, low friction, and high wear resistance in challenging environments. These findings suggest potential applications in biomedical devices, packaging materials, and lubrication systems, where repeated exposure to moisture is a critical factor. However, further research is needed to optimize additive concentrations, not by increasing them, but by identifying the most effective balance within the tested range to maximize performance while avoiding issues such as aggregation and excessive cross-linking. Additionally, future studies should focus on investigating the long-term performance under dynamic and real-world conditions, as well as evaluating their long-term environmental and

biomedical impacts of the composites, particularly through leaching tests and cytotoxicity evaluations, to fully realize their potential.

Author's declaration

Author contribution

Dieter Rahmadiawan: Conceptualization, validation, formal analysis, investigation, visualization, Writing - original draft. **Shih-Chen Shi**: Supervision, funding acquisition, writing - review & editing. Wei-Ting Zhuang: Investigation. **Eko Indrawan**: Writing - review & editing. **Yolli Fernanda**: Writing - review & editing. **Budi Syahri**: Writing - review & editing. **Irzal**: Writing - review & editing.

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Competing interest

There are no conflicts of interest in this research.

Ethical clearance

This research does not involve humansor animalsas subjects.

AI statement

The grammatical structure of this article was improved by using ChatGPT and the authors have rechecked the accuracy and correctness of the generated sentences with the topic and data of this study. The data and language use in this article have been validated and verified by an English language expert and none of the AI-generated sentences include in this article.

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