



Properties and printability of polylactic acid films : An effect of polyglutamate coated natto as a bio-plasticizer

Natrata THITINARDWONG¹, Thira CHAVARNAKUL^{2,3}, and Kawee SRIKULKIT^{4,*}

¹ Department of Technopreneurship and Innovation Management Program, Graduate School, Chulalongkorn University, Bangkok 10330, Thailand

² Department of Commerce, Chulalongkorn Business School, Chulalongkorn University, Bangkok 10330, Thailand

³ Transportation Institute, Chulalongkorn University, Bangkok 10330, Thailand

⁴ Department of Materials Science, Faculty of Science, Chulalongkorn University, Bangkok 10330, Thailand

*Corresponding author e-mail: kawee.s@chula.ac.th

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Abstract

Polylactic acid (PLA) film, a biodegradable alternative to petroleum-based plastics, has gained significant attention for sustainable packaging applications. However, its limited screen-printability remains a barrier to broader commercial use. This study aims to enhance the screen-printability of PLA film by incorporating a polyglutamate coated natto (designated as Nat-M). PLA/Nat-M films having 1%, 3%, and 5% Nat-M were prepared and evaluated for their key performance indicators, including ink adhesion (ASTM D3359), tone reproduction accuracy, and printing resolution. The screen-printing resolution was optimized between 400 DPI and 500 DPI (dots per inch) to achieve optimal tone reproduction and edge sharpness. Additionally, ink abrasion resistance and surface wettability were assessed by water contact angle (WCA) measurements. The results indicated that all PLA/Nat-M films exhibited improved screen-printing characteristics when compared to pristine PLA. These findings suggest that PLA/modified natto, particularly at 3% and 5% Nat-M loadings, provided a viable and environmentally friendly solution for improving the printability of biodegradable label.

1. Introduction

Polylactic acid (PLA) is a compostable, bio-based polymer that has gained widespread use in sustainable packaging [1]. PLA is now among the most important biodegradable polymers made on an industrial scale, with widespread use in a variety of packaging sectors due to environmental benefits. PLA is particularly attractive due to its distinctive physicochemical properties, such as high optical transparency and excellent tensile strength. Nevertheless, films fabricated from PLA are inherently brittle and prone to fracture. This limitation arises from its relatively high glass transition temperature ($T_g \sim 60^\circ\text{C}$), which exceeds ambient conditions and restricts polymer chain mobility arising from dipole-dipole interaction among polymer chains. Therefore, the addition of nucleating agents and plasticizers is necessary to overcome these limitations [2]. A common approach involves the incorporation of plasticizers, such as polyethylene glycol (PEG) [3], polypropylene glycol (PPG) [5], and polyester diol (PED) [6], which have been shown to improve film flexibility while maintaining optical clarity. According to the study polyethylene glycol (PEG) has been identified as an effective plasticizer for incorporation into PLA, primarily due to its non-toxicity and stable semicrystalline nature [3]. Nevertheless, PLA/PEG blends are often constrained by phase separation phenomena [4,5]. This issue arises from the high polarity of PEG, which enables rapid moisture absorption and accelerates crystallization compared with PLA. Consequently, PEG tends to crystallize preferentially and segregate from the PLA matrix, thereby undermining the structural homogeneity

and long-term stability of the blends. Polypropylene glycol (PPG) has been proposed as an alternative plasticizer for PLA, offering a low glass transition temperature, amorphous structure, and better miscibility. Studies have shown that PPG improves PLA's flexibility and elongation at break, although its effect on enhancing crystallization is weaker than PEG. Additionally, phase separation can occur at higher PPG concentrations, particularly with higher molecular weight PPG, highlighting the limitations of current plasticization strategies [7]. Blending PLA with polyester diols (PED) enhances flexibility and reduces brittleness by lowering the glass transition temperature, but limited miscibility with certain PED types can lead to phase separation, reducing mechanical strength and overall performance [6]. The incorporation of bio-based plasticizers into PLA has been demonstrated to effectively reduce the glass transition temperature (T_g), increase crystallinity, and significantly improve mechanical properties, including toughness and impact strength, without causing phase separation [8]. Moreover, effective bio-based plasticizers can enhance printability by improving ink absorption, thereby expanding the potential applications of PLA in flexible and printed packaging materials.

Poly(γ -glutamic acid) (γ -PGA) [9,10] produced by *Bacillus subtilis* is the major biopolymer component of natto, a traditional fermented soybean food. Its unique biochemical properties, including biodegradability, water solubility, and biocompatibility, make γ -PGA an attractive candidate for applications in food, biomedical, and pharmaceutical industries [11]. γ -PGA is chiefly responsible for the characteristic stickiness, viscosity, and hydrophilicity of natto [11,12].

Owing to these properties, natto represents a promising natural source of γ -PGA for use in PLA-based composites. The incorporation of natto-derived γ -PGA has the potential to improve the elasticity and biodegradability of PLA films. Nevertheless, the intrinsic hydrophilicity of γ -PGA and the hydrophobic nature of PLA may lead to interfacial incompatibility, highlighting the need for modification of coated γ -PGA to achieve compatibility with PLA matrix.

Packagings are typically printed prior to use. Plastic films like low-density polyethylene (LDPE) and polyethylene terephthalate (PET) have low surface energy and are commonly printed with solvent-based flexographic inks [13,14]. In instance, PLA film's higher surface energy than petroleum-based plastics limits its printability due to poor solvent-based ink adhesion. Therefore, acrylic-based inks are more suited for PLA printing because of the positive interaction between acrylic ester and PLA ester groups, which improves ink adherence. As a result, adding biopolyester-based plasticizers could improve the printability of acrylic ink and the flexibility of PLA films.

In this study, the modified natto (γ -polyglutamate coated natto or Nat-M) was used as a natural bio-plasticizer, to modify the surface properties and enhance the printability of PLA films, aiming at improving the screen printability of PLA. PLA/Nat-M films having three loadings of 1 wt%, 3 wt%, and 5 wt% Nat-M were prepared. The mechanical and thermal properties of the PLA/Nat-M films were evaluated. The printability of PLA/Nat-M films utilizing acrylic-based inks was examined using a screen printing machine. The key qualities including ink adhesion ASTM D3359-17 (2017), color tone accuracy, and ink abrasion resistance were tested.

2. Materials and experimental

2.1 Materials and processing

Nat-M (γ -polyglutamate coated natto which is currently under consideration in a peer-reviewed journal) was synthesized in Professor Kawee Srikulkit's laboratory. Polylactic acid (PLA) pellets (grade 2003D) were supplied by NatureWorks LLC (USA). This grade exhibits a melt flow index (MFI) of $6 \text{ g} \cdot 10 \text{ min}^{-1}$ at 210°C and a density of $1.24 \text{ g} \cdot \text{cm}^{-3}$, both measured in accordance with ASTM D1238. The glass transition temperature (T_g) lies between 55°C and 60°C . PLA 2003D is a versatile biopolymer commonly used for extrusion and thermoforming applications, particularly in food-packaging products. Commercial acrylic inks were kindly provided by Chaiyoboon Brothers Co., Ltd.

2.2 Preparation of PLA/Nat-M masterbatch

PLA and Nat-M powder were blended at a weight ratio of 60:40 and vacuum-dried at 55°C for 24 h to remove moisture. The dried mixture was then melt-compounded using a twin-screw extruder (Prism TSE 16TC, Thermo Electron Corporation, Germany) equipped with a 16 mm diameter screw and an L/D ratio of 25. The extrusion temperature profile was set as follows: feeding zone at 165°C , zones 2, 3, and 4 each maintained at 170°C . The screw speed was controlled at 30 rpm throughout the process. The extruded strands were subsequently pelletized to produce masterbatch chips suitable for film casting.

2.3 Preparation of PLA/Nat-M film casting

The PLA/Nat-M masterbatch composites was physically mixed with polylactic acid (PLA) to prepare feeding pellets containing 1 wt%, 3 wt%, and 5 wt% of Nat-M. Before processing, the blended pellets were vacuum-dried at 55°C for 24 h to remove moisture. The composite films were produced using a cast film and sheet extruder. During the extrusion process, the temperature of the feeding zone was maintained at 170°C , while the die zone temperature was set to 220°C . The screw speed was controlled within the range of 10 rpm to 15 rpm to ensure uniform mixing and stable film formation. The final films had a thickness ranging from 0.1 mm to 0.2 mm and a width of approximately 200 mm. The extruded films were collected and stored under controlled conditions for subsequent characterization.

2.4 Hand screen printing

Hand screen printing was carried out on PLA and PLA/Nat-M films containing 1%, 3%, and 5% Nat-M to evaluate printability. Samples measuring $5 \text{ cm} \times 15 \text{ cm}$ were prepared for the tests. The acrylic-based ink was used as the printing medium. The films were placed flat on the printing surface, and the ink was applied through a fine 200 mesh screen using a squeegee to transfer the design onto the film surface. Printing was carried out under ambient laboratory conditions. After printing, the samples were dried and cured at 70°C for 5 min.

2.5 Mechanical properties

The tensile behavior of PLA/Nat-M composites was investigated using a universal testing machine (Instron 5566, USA) under ambient conditions, in accordance with the requirements of ASTM D638 and ASTM D882. Test specimens, cut into rectangles measuring $20 \text{ mm} \times 150 \text{ mm}$ with a thickness between $100 \mu\text{m}$ and $200 \mu\text{m}$, were pulled at a constant crosshead speed of $5 \text{ mm} \cdot \text{min}^{-1}$ using a 5 kN load cell. The tensile modulus, ultimate tensile strength (σ_b), and elongation at break (ϵ_b) were obtained from the corresponding stress-strain curves, and all data represent the mean of a minimum of ten replicates.

2.6 Thermal properties by differential scanning calorimetry (DSC)

Differential scanning calorimetry (DSC, DSC 1, Mettler Toledo, Switzerland) was employed to obtain thermograms. Approximately three representative specimens, each weighing $2 \text{ mg} \pm 0.5 \text{ mg}$, were analyzed over a temperature range of 0°C to 200°C at a heating rate of $10^\circ\text{C} \cdot \text{min}^{-1}$ under a nitrogen atmosphere. The degree of crystallinity (χ_c) was calculated according to:

$$\chi_c = \frac{(\Delta H_m - \Delta H_{cc})}{\Delta H_m^0 \times \phi} \times 100$$

In this equation, ϕ represents the mass fraction of the polymer within the blend, ΔH_m is the melting enthalpy, ΔH_{cc} denotes the enthalpy of cold crystallization, and ΔH_m^0 corresponds to the melting enthalpy of fully crystalline PLA, taken as $93 \text{ J} \cdot \text{g}^{-1}$.

2.7 Water absorption properties

Following the procedure outlined in ASTM D570-22 (2022), the specimens were initially dried in an oven at 65°C for a minimum of 24 h. After drying, each sample was conditioned and its initial weight was recorded (W_c). The specimens were then immersed in distilled water for intervals ranging from 1.8 min to 24 h. At each designated time point, the samples were taken out, gently wiped with tissue paper or a soft cloth to remove any surface water, and weighed immediately (W_w). The water absorption (W_f) was calculated according to:

$$W_f (\%) = \frac{W_w - W_c}{W_c} \times 100$$

W_c is the dry (conditioned) weight of the specimen, W_w is the weight after immersion at a specific time, and W_f is the percentage of water uptake.

2.8 Ink abrasion test

The abrasion resistance of the printed films was evaluated according to ASTM D5264–98 (2017) using a Sutherland Rub Tester (Sutherland 2000™, USA). Prior to testing, all specimens were conditioned at 23°C ± 1°C and 50% ± 2% RH for 24 h. Printed samples were cut into strips measuring 50 mm × 100 mm and mounted on the tester platform, while a standard receptor sheet was attached to a 2 lb test weight. The printed surfaces were rubbed under a constant load for 100 rub cycles. Each condition was tested in five replicates to ensure reproducibility and reliability of the results. After the test, the abraded areas were visually examined for ink removal, smearing, or color transfer. The extent of abrasion was recorded as the percentage of ink removal, providing a comparative evaluation of the ink durability and surface resistance of the pristine PLA and PLA/Nat-M films under mechanical rubbing.

2.9 Soil burial test

The biodegradation of the samples was assessed using a soil burial test under outdoor conditions in accordance with ASTM D5338-15 (2015). Precisely weighed specimens were buried individually in holes approximately 20 cm deep for periods of 90 d (3 months) and 210 d (7 months), respectively. The soil pH was maintained at 5 to 6 by adding a citric acid–water solution. Weight loss was recorded at 90 d and 210 d, and morphological changes of the samples were examined using scanning electron microscopy (SEM). The surface morphology using a scanning electron microscope (model JSM-6480LV, JEOL Ltd., Japan) operated at an accelerating voltage of 15 kV. For each sample type, three representative specimens with dimensions of 1 cm² were prepared, gold-coated via sputter deposition,

and subsequently dried in an oven at 60°C for 24 h prior to SEM observation.

3. Results and discussion

3.1 Thermal properties of PLA/Nat-M films

Typically, PLA film exhibits poor stabilities including thermal and aging properties, leading to weak and fragile film. The effect of Nat-M on PLA film's thermal properties was evaluated by DSC analysis and the DSC thermograms are shown in Figure 1. The T_g and T_m values of PLA film are measured at 60.82°C and 148.50°C, respectively, where the glass transition state is high but the melting temperature is low when compared to petroleum plastics. Moreover, cold crystallization temperature (T_{cc}) is observed in the region of 110°C to 120°C, implying that PLA poorly crystallized during film extrusion. This is why typical PLA film is prone to losing its stability, such as tear strength, fragility, and aging. For PLA/Nat-M films, the addition of Nat-M results in a significant reduction of T_g of PLA. The thermal properties of PLA and PLA/Nat-M composites were investigated using DSC. The glass transition temperature (T_g) and melting temperature (T_m) of the samples are presented in Table 1. The addition of Nat-M resulted in a significant reduction in T_g , where it falls to 46.08°C, 45.54°C, and 44.52°C for PLA filled with 1 wt%, 3 wt%, and 5 wt% Nat-M, respectively.

The plasticization effect of polyglutamate-coated natto is found in a similar manner to the study of previous reports concerning bio-plasticizers [15] such as maleinized Brazil nut seed oil/maleinized hemp seed oil [16], and epoxidized Brazil nut oil [17]. Similarly, Nat-M reduced the glass transition temperature and increased chain mobility, while maintaining or moderately improving tensile strength and modulus. Thermal analysis indicated that melting temperatures significantly rise, and % crystallinity (X_c %) was enhanced (from 7.5% to 8.5% to 12.2%), contributing to improved mechanical properties. Furthermore, the incorporation of Nat-M improved film processability and printability, suggesting its potential as a bio-based plasticizer alternative to conventional bioplasticizers for PLA processing aid.

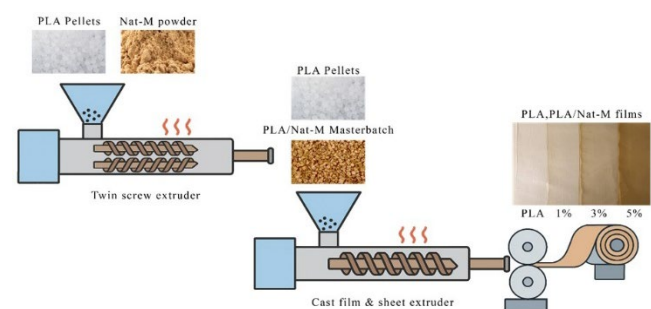


Figure 1. Preparation of PLA/Nat-M masterbatch and PLA/Nat-M films.

Table 1. Thermal characteristics of the samples evaluated as determined by DSC.

Sample	T_g [°C]	T_m [°C]
(a) PLA	60.82	148.50
(b) PLA /1 wt% Nat-M film	46.08	157.65
(c) PLA/3 wt% Nat-M film	45.54	158.14
(d) PLA/5 wt% Nat-M film	44.52	159.50

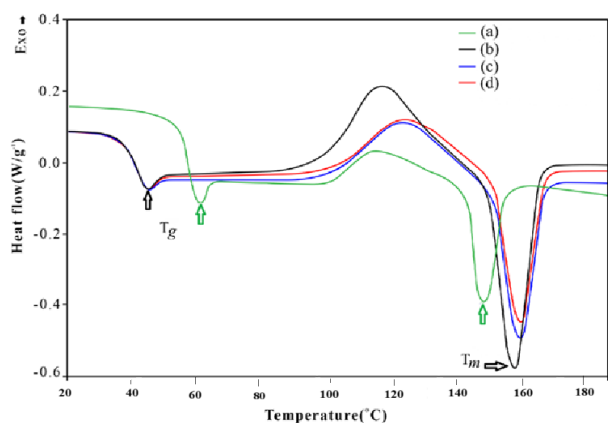


Figure 2. DSC thermograms (a) PLA film, (b) PLA/1 wt% Nat-M film, (c) PLA/3 wt% Nat-M film, and (d) PLA/5 wt% Nat-M film.

The addition of Nat-M means that PLA/Nat-M films are more flexible than pristine film. With Nat-M addition, T_m values (Table 1) increased from 148.50°C in pristine PLA to 157.65°C, 158.14°C, and 159.50°C for 1 wt%, 3 wt%, and 5 wt% Nat-M, respectively. The increase in T_m implies that PLA/Nat-M films exhibit higher thermal stability when compared to pristine PLA. However, the poor crystallization behavior during film casting still exists.

3.2 Mechanical properties

The effect of Nat-M on the mechanical properties of PLA was evaluated. 10 representatives were tested. The mechanical properties of PLA and PLA/Nat-M are shown in Figure 3. Pristine PLA film

serves as a standard, with tensile strength of 14.25 MPa and modulus of 1115 MPa. The addition of Nat-M improves mechanical properties, with the maximum improvement occurring at 1% Nat-M, which increased tensile strength to 24.18 MPa. Tensile strength results at 3% and 5% Nat-M decline dramatically to 18.05 MPa and 16.34 MPa, respectively, but remain higher than that of pristine PLA film.

Similarly, Nat-M has been shown to improve Young's modulus values. PLA film has a Young's modulus of 1115.69 MPa. The addition of 1% Nat-M leads to a significant increase in stiffness, with Young's Modulus climbing to 1628.86 MPa, the highest value among all the samples and showing a notable improvement over pristine PLA film. However, increasing the concentration of Nat-M to 3% and 5% results in a drop in Young's Modulus to 1255.80 MPa and 1206.60 MPa, respectively, while these values remained higher than pristine PLA film. Based on this finding, Nat-M can be offered as a new reinforcing material for polylactic acid. However, its performance is reliant on loading content, and loading above the optimum level will result in a drop in performance owing to agglomeration.

3.3 Printing

3.3.1 Product label printing

To assess the printability of packaging labels, PLA film and PLA/Nat-M films comprising 1 wt%, 3 wt%, and 5 wt% of Nat-M were screen-printed, as shown in Figure 4. In the figure, (a) represents screen printing on the PLA film, (b) represents screen printing on PLA/1 wt% Nat-M film, (c) represents screen printing on PLA/3 wt% of Nat-M film, and (d) represents screen printing on PLA/5 wt% of Nat-M film.

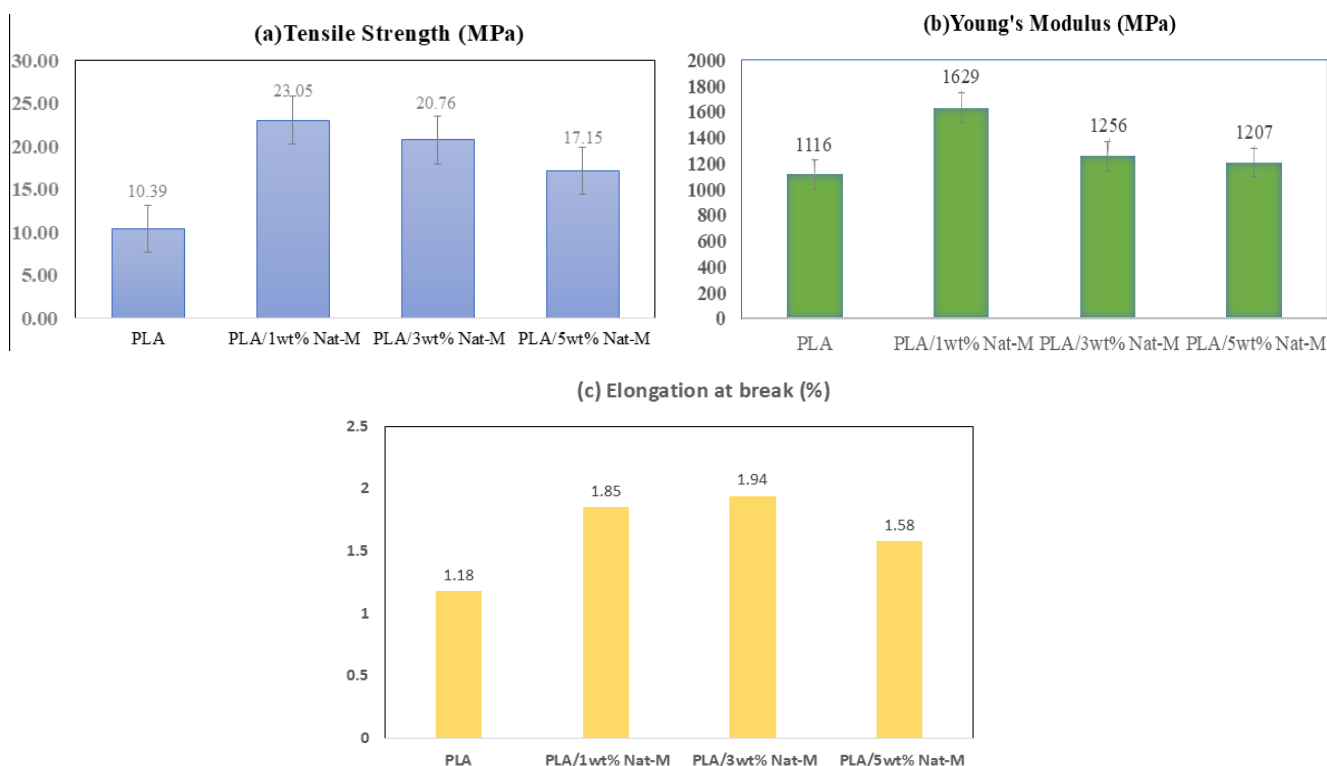


Figure 3. (a) Tensile strengths of PLA film and PLA/Nat-M films, (b) Young's modulus values of PLA and PLA/Nat-M films, and (c) % Elongation at break of PLA and PLA/Nat-M films (average values of 5 representatives).

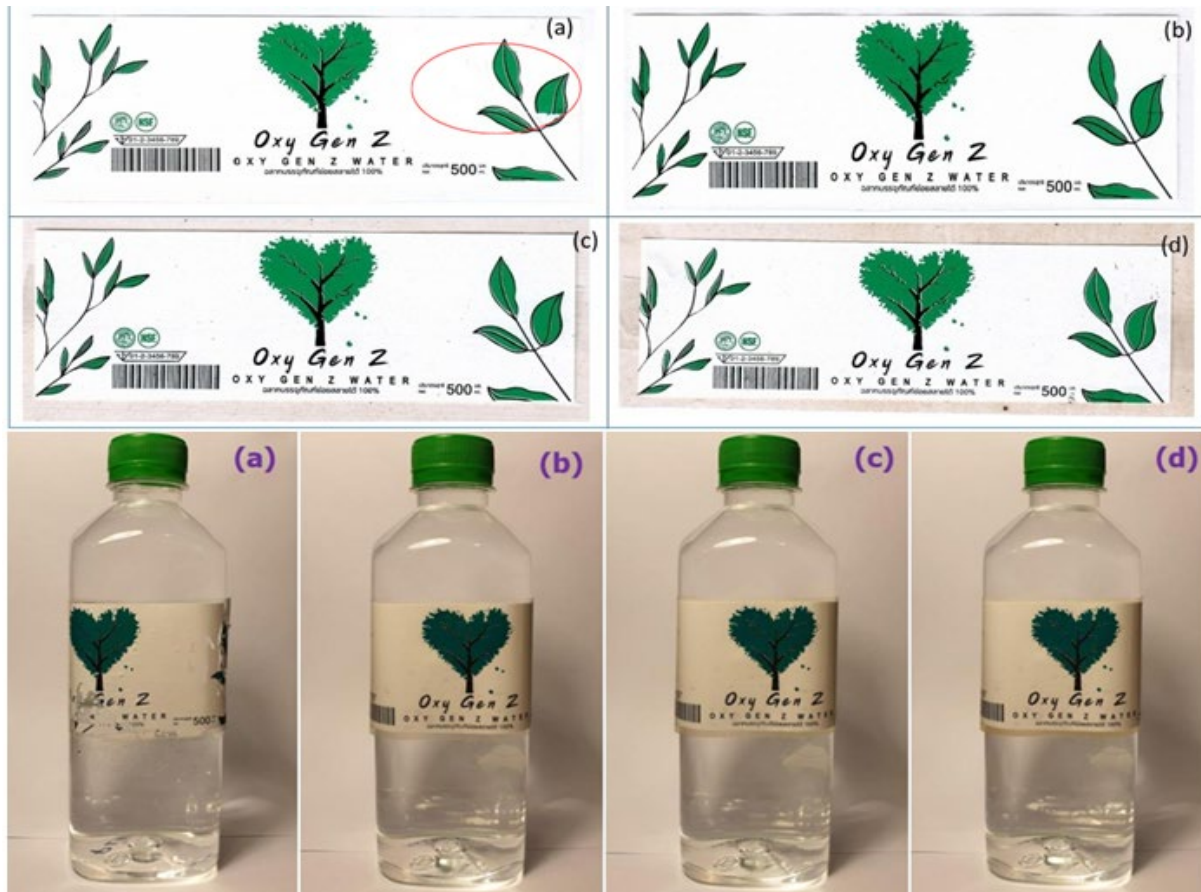


Figure 4. Screen Printing on (a) PLA film, (b) PLA/1 wt% Nat-M film, (C) PLA/3 wt% Nat-M film, and (d) PLA/5 wt% Nat-M film.

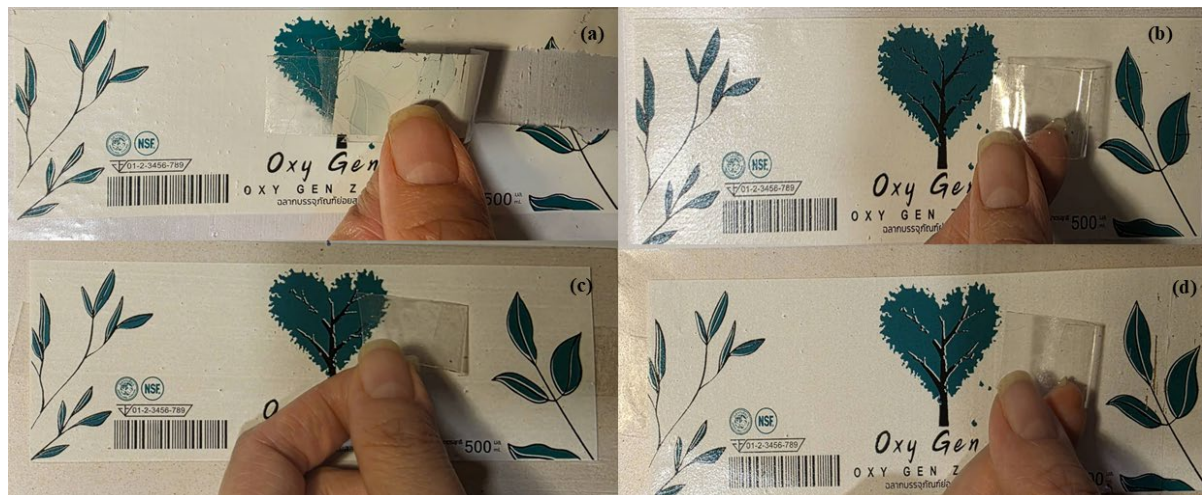


Figure 5. Tape test results of (a) PLA film, (b) PLA/1 wt% Nat-M film, (C) PLA/3 wt% Nat-M film, and (d) PLA/5 wt% Nat-M film.

Figure 4 Screen-printed “Oxy Gen Z” labels on PLA film and PLA/Nat-M films with various Nat-M loadings. The print quality and ink adhesion differed noticeably among the samples. The pristine PLA film (a) exhibits poor printability, with significant ink peeling-off and poor ink transfer due to its low surface energy and poor wettability. In contrast, the PLA/1 wt% Nat-M film, (b) shows improved print quality with a more continuous and uniform printed pattern, indicating enhanced surface interaction between the ink and the modified substrate. The PLA/3wt% Nat-M film, (c) demonstrates further improvement,

producing sharp, well-defined images with no signs of ink detachment, suggesting that a moderate Nat-M content contributes to change in surface polarity and roughness which promotes better ink adhesion. The PLA/5 wt% Nat-M film, (d) presents the highest print quality and adhesion strength, with intact, dense color patterns and no cracking or fading. The higher Nat-M loading appears to optimize surface compatibility with the ink binder, resulting in durable and stable printing suitable for product labeling applications.

3.3.2 Color tone accuracy and ink adhesion test

The color tone accuracy of the printed PLA and PLA/Nat-M films was tested to determine their ability to reliably replicate colors after printing. The results show that all PLA/Nat-M films exhibit color tones closely matching the original digital artwork, with 5 time print/sample. This indicates that the addition of Nat-M did not compromise color fidelity and may have contributed to more uniform ink absorption and color rendering on the film surface. Ink adhesion was evaluated based on the amount of ink removed by the tape, following the ASTM D3359 rating scale, which ranges from 5B (no removal, excellent adhesion) to 0B (greater than 65% ink removal, poor adhesion). The test was conducted in triplicate for each sample, and average ratings were reported to ensure reliability shown in Figure 5.

The ink adhesion of pristine PLA film and PLA/Nat-M films was evaluated according to ASTM D3359. Pristine PLA film exhibits 100% ink removal (rating 0), indicating poor ink adhesion. In contrast, all PLA/Nat-M films exhibit 0% ink removal (rated 5), reflecting a significant improvement in ink adhesion due to the Nat-M addition. This enhancement may be attributed to the plasticizing effects of Nat-M which reduces dipole-dipole interaction among PLA polymer chains, promoting greater ink absorption into the film surface.

3.4 Surface wettability analysis by water contact angle (WCA)

Surface wettability of the PLA/Nat-M films was evaluated using a contact angle goniometer (OCA 20, Dataphysics Instruments, Germany). A droplet of distilled water (5 μ L) was carefully placed on five different positions of each film surface. The water contact angle (WCA) was measured at each position, and the average value was

reported to represent the surface hydrophilicity or hydrophobicity of the sample. Wettability is typically quantified by measuring the water contact angle (WCA) which was measured as presented in Table 4 and Figure 6.

Pristine PLA film typically exhibits a water contact angle of around 95°, indicating a hydrophobic surface, as contact angles above 90° are considered hydrophobic and resist wetting by water. However, the additions of modified natto or Nat-M into PLA with concentrations of 1%, 3%, and 5% result in a decrease in WCA to values below 90°, reflecting a transition toward a more hydrophilic behavior. Among these, the 5% PLA/Nat-M blend demonstrates the lowest contact angle, suggesting it possesses the highest degree of hydrophilicity.

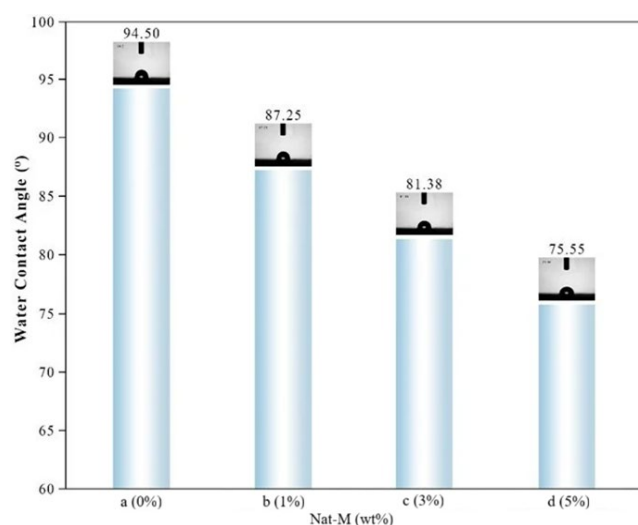


Figure 6. WCA (a) PLA film, (b) PLA/1 wt% Nat-M film, (c) PLA/3 wt% Nat-M film, and (d) PLA/5 wt% Nat-M film.

Table 2. Evaluation of ink adhesion of PLA film and PLA/Nat-M films (ASTM D3359).

Sample	Ink removal [%]	Adhesion rating (ASTM D3359)	Observation
PLA/Nat-M 1%	0	5	Excellent ink adhesion
PLA/Nat-M 3%	0	5	Excellent ink adhesion
PLA/Nat-M 5%	0	5	Excellent ink adhesion
PLA	100	0	Poor ink adhesion

Table 3. Water contact angle (WCA) of PLA film and PLA/Nat-M films.

Sample film	Contact angle [°]	Droplet image
PLA	94.50 ± 0.5	
PLA/1 wt% Nat-M	87.25 ± 1.2	
PLA/3 wt% Nat-M	81.38 ± 1.8	
PLA/5 wt% Nat-M	75.55 ± 2.5	

Table 4. Summary of ink abrasion resistance of PLA film and PLA/Nat-M films.

Film	Pressure [lbs]	Rub cycles	Ink removal [%]	Ink loss
PLA	2	100	100	fading
PLA/1 wt% Nat-M	2	100	5	Slight smearing
PLA/3 wt% Nat-M	2	100	0	-
PLA/5 wt% Nat-M	2	100	0	-

3.5 Ink abrasion resistance test

Printed PLA films exhibit lower resistance to surface abrasion, while the incorporation of Nat-M significantly improved durability under both 2 lbs and 4 lbs pressures. Under 2 lbs pressure and 100 rub cycles, each sample was tested in five replicates, pristine PLA showed complete ink removal (100%), indicating poor abrasion resistance. PLA/Nat-M films exhibit improved anti-abrasion performance: PLA/1 wt% Nat-M film has 5% ink removal with slight smearing, while

PLA/3 wt% Nat-M and PLA/5 wt% Nat-M films show no visible ink loss, demonstrating enhanced ink durability.

The results, presented in Table 4 show that pristine PLA film illustrates 100% ink removal, reflecting complete ink fading upon abrasion. In contrast, PLA/1 wt% Nat-M film exhibits slight smearing with only 5% ink removal. Furthermore, PLA/3 wt% Nat-M and PLA/5 wt% Nat-M films demonstrate no visible ink removal, indicating a significant improvement in ink resistance as a result of Nat-M addition.

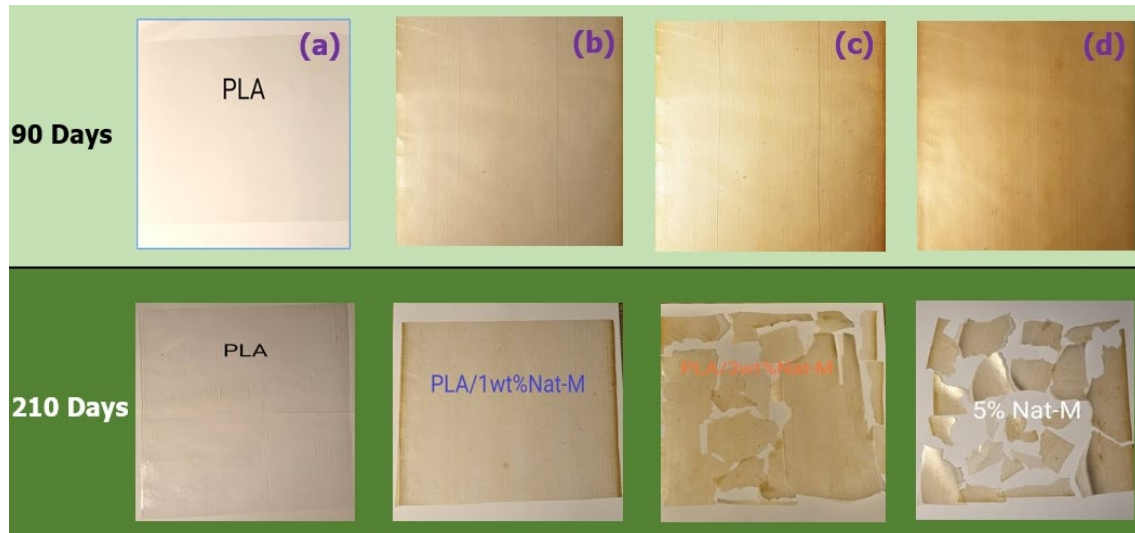


Figure 7. Photos of decomposed films taken after soil burial test (a) PLA film, (b) PLA/1 wt% Nat-M film, (C) PLA/3 wt% Nat-M film, and (d) PLA/5 wt% Nat-M film after 90 d and 210 d.

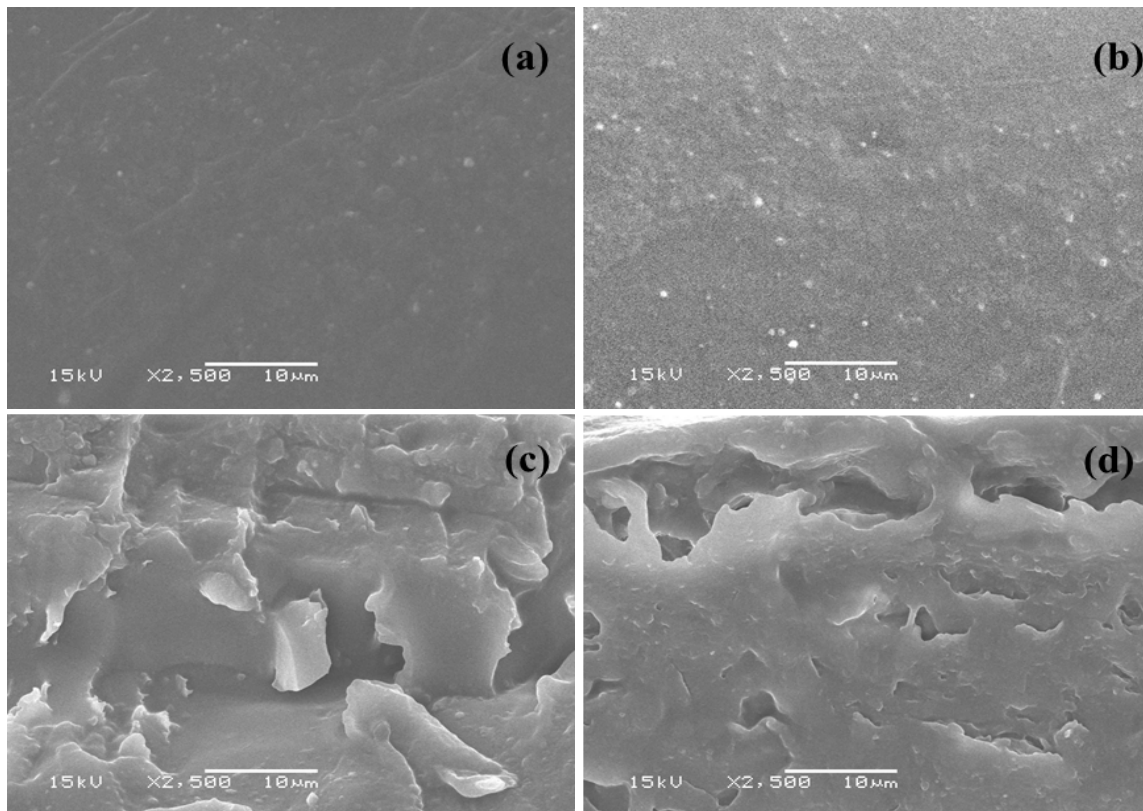


Figure 8. SEM micrographs of film surface taken 210 days after soil burial tests: (a) PLA film, (b) PLA/1 wt% Nat-M film, (C) PLA/3 wt% Nat-M film, and (d) PLA/5 wt% Nat-M film.

3.6 Decomposition characteristics of films

3.6.1 Physical appearance after soil burial test

The soil burial test, conducted according to ASTM D5338, was performed on PLA film and PLA/Nat-M films with contents of 1 wt%, 3 wt%, and 5 wt% for periods of 90 d and 210 d, as shown in Figure 7. The results indicate that pristine PLA film exhibits only slight yellowing even after 7 months of burial. In contrast, PLA/Nat-M films showed more pronounced yellowing and increased brittleness due to degradation, with the effect becoming more evident as the Nat-M content increased from 1 wt% to 5 wt% after 3 months in soil burial. After 7 months, PLA/Nat-M films with 3 wt% and 5 wt% exhibit noticeable fragmentation, with the 5 wt% sample showing the most deterioration.

3.6.2 Morphology

The morphology of the soil burial films was studied using SEM, as illustrated in Figure 8. The findings show that buried PLA/Nat-M films with 3 wt% and 5 wt% Nat-M suffered substantial natural degradation after 7 months in burial soil, whereas no significant degradation was detected in pure PLA films. As demonstrated, PLA deterioration during soil burial tests is proportional to Nat-M content; the higher the Nat-M content, the faster the PLA degrades. The presence of Nat-M in PLA matrix is responsible for the water absorption of PLA/Nat-M films, which allows bacteria in soil undergoing hydrolytic processes to break down PLA polymer chains.

4. Conclusions

This study successfully demonstrated that the addition of polyglutamate coated natto or Nat-M into PLA film could improve several properties. The addition of Nat-M not only improved ink adhesion but also influenced the thermal and mechanical properties of PLA, as well as its biodegradability. Nat-M served as a plasticizer and a reinforcing material for PLA, as demonstrated by a decrease in glass transition temperature (T_g) and melting temperature (T_m), while simultaneously enhancing the Young's modulus of the PLA films. Furthermore, ink abrasion resistance tests revealed that PLA/3 wt% Nat-M and PLA/5 wt% Nat-M samples exhibited no visible ink loss, demonstrating enhanced ink adhesion when compared to pristine PLA film. The increased surface wettability expands the possible applications of PLA/Nat-M films in biodegradable packaging materials. More research is needed to understand the aging characteristics, biodegradation behavior, and environmental impact of PLA/Nat-M films in commercial conditions.

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