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Foaming Behavior and Water Absorption-Desorption Cycle of Polylactide for Food Packaging

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***Abstract*— This study investigates the foaming behavior of polylactide (PLA), notoriously difficult to foam, using a pressure-induced batch foaming method with supercritical carbon dioxide (sc-CO_2) as the physical blowing agent. Choosing sc-CO_2 has strategic advantages due to its easily controlled critical point and high solubility in PLA, making it convenient for laboratory experiments. This research aims to explore PLA foaming with the primary aim of developing a sustainable alternative to expanded polystyrene (EPS) foams. As a bioplastic derived from renewable resources, PLA offers a promising solution to the pressing environmental concerns associated with EPS. By experimentally investigating the foaming of PLA using sc-CO_2, this study aims to contribute to developing eco-friendly and sustainable packaging solutions. Foams were prepared at different foaming pressures ranging from 14,426 kPa to 19,574 kPa with a constant temperature of 443.15 K for 135 minutes. The highest density reduction (90.645%) and superior compressive strength (5.597 MPa) were achieved. FTIR analysis indicates no residual chemical reactions in the foaming process, as seen by the absence of the -CH group vibration (2919 cm^{-1}) in the foamed PLA and the presence of the strong O-H peak (3297 cm^{-1}) only in unfoamed PLA. The water uptake test showed that the PLA foam absorbed around 12% water within the first 15 minutes, which is lower than many other biodegradable foams. It also withstood over 3 cycles of 2-hour water immersions, suggesting its potential for sustainable packaging applications.**

***Keywords*— Compressive strength; Foaming behavior; Food packaging; Polylactide; Reusability; Supercritical CO_2**

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I. INTRODUCTION

Exacerbated environmental concerns and the imminent depletion of petroleum resources have prompted a concerted effort within both industry and academia to actively explore alternative raw materials. In 2019, an alarming 22 million tons of plastics (including microplastics) were released into the environment [1], exacerbating pre-existing environmental challenges. The subsequent emergence of the COVID-19 pandemic in 2020 further worsened these issues, leading to a significant increase in plastic use, particularly in the healthcare sector due to the widespread adoption of personal protection equipment (PPE) [2]. Additionally, strict regulations applied during the pandemic triggered an intensified demand for food commodities and packaging [3], further amplifying the strain on environmental sustainability. Due to the depletion of petroleum resources, there is a growing need for eco-friendly polymeric materials that offer properties comparable to those currently available in the market.

Expanded polystyrene (EPS) poses significant environmental and health hazards due to its widespread use in disposable cups, boxes, and packaging materials, constituting 47% of its total usage [4-6]. Styrene, the primary constituent of EPS, is carcinogenic [7] and may migrate from EPS containers into food items, whether intentionally added or present as contaminants. This migration has the potential to alter the sensory characteristics of the food and pose toxic risks to consumers [8-10]. Moreover, discarded EPS products contribute to landfill waste, taking centuries to degrade [11], and may emit harmful gases like polyaromatic hydrocarbons (PAHs) and hydrochlorofluorocarbon (HCFC) if incinerated [12].

Polylactide (PLA), a polymer synthesized from renewable resources such as corn starch, rice, soy, potatoes, or whey—a by-product of the dairy industry—via ring-opening polymerization of lactide, exhibits favorable mechanical properties over extended storage periods and under controlled environmental conditions (relative humidity and temperature), durability, and transparency [13, 14]. It effectively maintains the physical and chemical attributes of packaged food while ensuring microbiological safety and facilitating natural degradation processes [15]. Numerous studies have documented polylactide as a promising biopolymer candidate poised to supplant conventional plastics across diverse applications, potentially replacing expanded polystyrene (EPS) in packaging [16]. The approval of the lactide monomer as a safe food ingredient by the Food and Drug Administration (FDA) elevated PLA's status as a green polymer of interest, further enhancing its appeal due to its low toxicity [17-21]. In a recent study on lactide-based nanocomposites by Faba, *et al.* (2022) [22], it was demonstrated that the mechanical properties of the material can be enhanced through the addition of filler, resulting in values comparable to those of polystyrene foams. Furthermore, the foamed material exhibited superior flexibility compared to its film counterpart. Specifically, the elongation at the break of polylactide foam increased due to the plasticization effect of CO_2 during the foaming process. However, it was noted that the Young's modulus and tensile strength of the PLA foam were reduced in comparison to the PLA film [22]. The lactide-based composite film, as investigated by Subbuvel and Kavan (2021) [23], exhibited promising antimicrobial properties against both gram-positive and gram-negative bacteria. This demonstrated robust properties, attributed to the synergistic effects of neem oil and curcumin, rendering the film suitable for active food packaging application [23]. Yu, *et al.* (2017) [24] investigated pure PLA and its nanocomposite water uptake when immersed. They found that water uptake decreased in the material with filler, owing to increased PLA tortuosity caused by rod-like filler shape [24].

Despite the demonstrated attractive processing characteristics of PLA, its widespread adoption is hindered by limitations in melt strength, crystallization kinetics, thermal stability, brittleness, and moisture sensitivity. Supercritical carbon dioxide (sc-CO_2) treatment has emerged as a promising strategy to address these limitations and tailor PLA for specific applications [25-28]. As the CO_2 interacts with polylactide, slight variations in pressure and temperature during compression can have a significant impact. This interaction can induce plasticization within the polymer melt, potentially enhancing material properties, introducing new functionalities, or priming the material for specific applications [29, 30]. The appeal of sc-CO_2 technology goes beyond its eco-friendliness. Its ability to scale up to high-capacity, continuous production makes it even more attractive. This innovative technology utilizes CO_2, a non-toxic and abundant physical foaming agent, significantly reducing the need for harmful chemicals. This aligns perfectly with the growing emphasis on carbon neutrality and sustainable manufacturing, promoting resource conservation and emission reduction [31-33].

This study investigates the batch foaming of PLA using supercritical carbon dioxide as a sustainable approach for developing lightweight and stable foams. Employing a saturation-desorption technique, sc-CO_2 is first incorporated into the melted polylactide, triggering bubble formation driven by thermodynamic instability upon pressure release. Our research delves into the foaming behavior of PLA under these conditions, aiming to evaluate its potential as a viable and sustainable material for food packaging applications.

II. MATERIAL AND METHODS

A. Material

Polylactide (PLA) feedstock employed in this study was Ingeo™ Biopolymer 4043D pellets, commercially available from NatureWorks LLC. To preserve its melt processing characteristics, this grade was pre-conditioned through overnight drying at 60°C in an oven, thereby minimizing potential viscosity degradation during subsequent processing. High-purity carbon dioxide (CO_2) obtained from Linde Thailand served as the blowing agent, chosen for its suitability under supercritical conditions. The mild critical point parameters of CO_2 (71.4 kg/cm² and 31°C) facilitated its effective utilization in the foaming process.

B. Foam Preparation

Batch foaming of PLA was performed by saturating the molten polymer with CO_2 gas in a high-pressure vessel (**Figure 1**). The mixture was maintained at 443.15 K for 135 minutes for CO_2 absorption with different foaming pressures ranging from 14,425 kPa to 19,574 kPa. Subsequently, controlled pressure release to the environment at 353.15 K was employed to induce bubble formation. This pressure drops triggered thermodynamic instability, leading to the expansion and stabilization of the PLA foams.

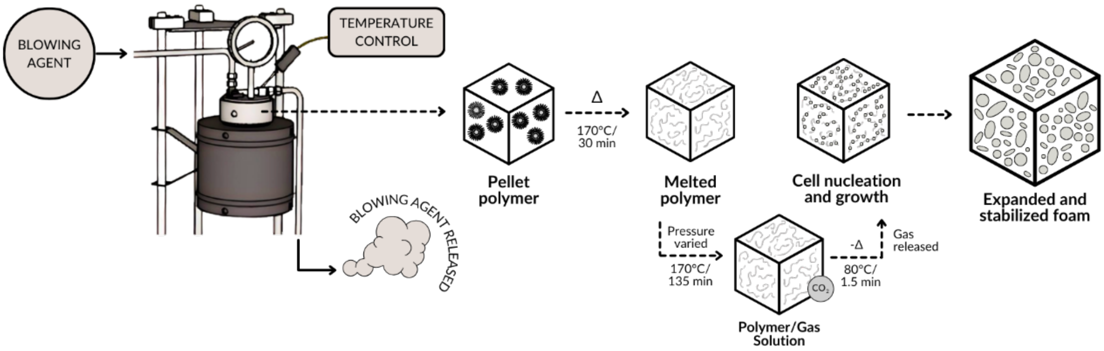

Fig. 1 Schematic diagram of experimental procedure in foaming polylactide

C. Foam Characterizations

The foams were characterized for their functional groups, density reduction, compressive mechanical behavior, and water absorption capacity, with reusability evaluated in the context of sustainability. FTIR spectroscopy in attenuated total reflectance (ATR) mode on a Nicolet iS5 spectrometer (32 scans, 4 cm^{-1} resolution) was used to identify the functional groups through analysis of their distinctive vibrational bands.

Foam density was assessed using a high-precision digital balance equipped with a density kit, following the procedures outlined in ASTM D792 [34]. Measurements were replicated three times to ensure statistical robustness, and the percentage reduction in density compared to the pellet form was calculated using the following formula:

$$\rho_{reduction}(\%) = \left(\frac{\rho_{pellet} - \rho_{foam}}{\rho_{pellet}}\right) \times 100\% \qquad \text{Eq. (1)}$$

where ρ is density in g/cm^3 for both forms (pellet and foam).

The compressive mechanical behavior of the foams was characterized in accordance with ASTM D695 [35] using a universal testing machine (H5KT, Tinius Olsen, UK) equipped with a 5000 N load cell. Measurements were conducted at room temperature with a crosshead speed of 0.8 mm/min. To ensure statistical significance, each test was repeated three times, and the reported values represent the average of these replicates.

Water uptake behavior was evaluated in accordance with ASTM D570 [36]. Foamed samples were initially dried in an oven at 110°C for 1 hour to remove residual moisture, followed by equilibration at room temperature. Assessing the long-term water absorption performance and potential reusability, the samples were subjected to three consecutive cycles of immersion for 2 hours each, interspersed with oven drying at 110°C for 1 hour between cycles. Water uptake was quantified after each cycle.

III. RESULT AND DISCUSSION

A. FTIR Spectroscopy

The FTIR analysis confirms the structure of pelletized and foamed PLA. The main characteristic of PLA can be determined at wavenumbers 1750 and 1082 cm^{-1}, corresponding to the symmetric stretching vibration (v_s) of the C=O and asymmetric stretching vibration (v_{as}) of the C=O group in the pelletized PLA, respectively. In foamed PLA, the v_s(C=O) and v_{as}(C=O) groups are observed at wavenumbers 1748 and 1079 cm^{-1}, respectively. The peaks at 1180 and 1128 cm^{-1} are associated with the v_{as}(C–O) and v_s(C–O) bonds for both pelletized PLA and foamed PLA. The C–H bond and –CH_2 group for both pelletized PLA and foamed PLA are represented by peaks at 1383 and 1360 cm^{-1}. The amorphous and crystalline zones of PLA can be observed at peaks 870 and 754 cm^{-1} for the pelletized PLA and at 868 and 753 cm^{-1} for foamed PLA [37]. The difference between pelletized PLA and foamed PLA is evident in the tensile vibration peak of –CH group at around 2919 cm^{-1}, which only appears in pelletized PLA [38]. Furthermore, the strong peak at 3297 cm^{-1} indicates the O–H stretching vibration in pelletized PLA.

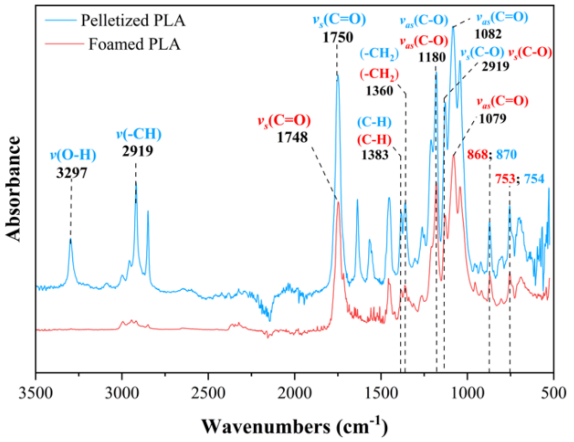

Fig. 2 FTIR spectroscopy of pelletized PLA and foamed PLA

B. Foaming Behavior

Foaming is a process that effectively reduces the density of a material by saturating gas in the polymer melt under the controlled temperature and pressure for a certain period. The cellular structure is then generated via thermodynamic instability which results in a lightweight and expanded product. The reduction in density is a crucial aspect of the foaming process and is a significant goal in developing foamed materials as it is affect the properties of the foam [39]. As we can see from **Figure 3**, the foamed product exhibits maximum density reduction (90.645 % ± 0.081) at 14,425 kPa, achieving a super-expanded foam. This indicates a trend of higher foam density with increasing foaming pressure, suggesting the production of lighter foams at lower pressures. This trend stems from two competing effects: reduced melt strength and intensified CO_2 plasticization at higher pressures [40, 41]. Higher foaming pressures increase the dissolved CO_2 concentration in the polymer, amplifying its plasticizing effect [42, 43]. This, combined with the reduced melt strength, hinders cell expansion and growth, leading to a smaller percentage of density reduction.

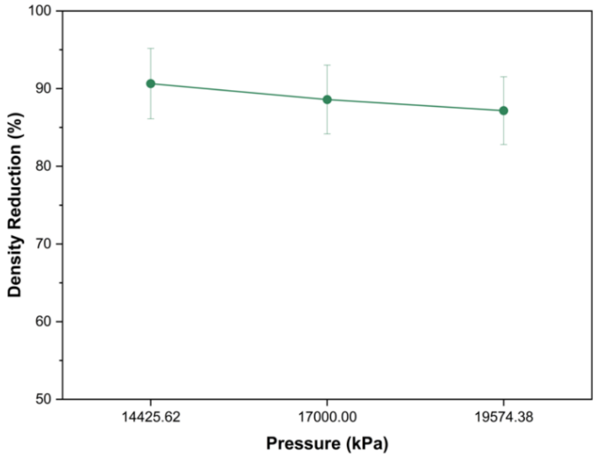

Fig. 3 Pressure dependence of PLA foam density reduction

Additionally, excessive chain mobility induced by plasticization promotes the formation of imperfect crystals with dense lamellas, acting as nucleation sites during foaming [44], further affecting the mechanical properties of the foam. An increase in nucleation site density, observed primarily at higher foaming pressures can suppress cell growth rate. This results in the formation of smaller cells, but with a higher overall cell number. Consequently, the foam exhibits enhanced deformation stress capacity due to the increased structural integrity and resistance to deformation afforded by the numerous small cells [40]. However, the initial decrease in compressive strength observed in **Figure 4(a)** might be attributed to the mid-value of density reduction percentage, where there might be only fewer closed cells available to bear the mechanical load and the rupture-prone of some cells due to the insufficient surface tension that could hinder the expansion [45, 46].

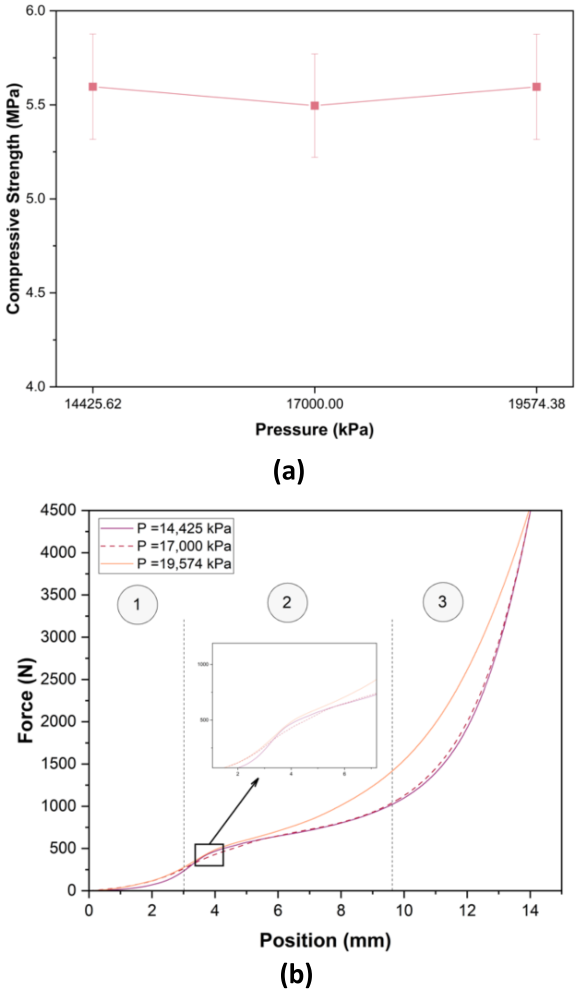

Fig. 4 Compressive properties of PLA foam under different foaming pressures: **(a)** Compressive strength of PLA foam, and **(b)** Mean force-position curve behavior of PLA foam

The force-position curve depicted in **Figure 4(b)** illustrates the mechanical response to compression loading. Three distinct phases characterize the compression process: (1) elastic deformation, (2) plateau, and (3) densification. Initially, a linear response suggests uniformity in cell structure. Subsequently, during the plateau phase, the load either increases slightly or remains constant until reaching the onset of densification, marking the point at which the structure is fully crushed [47]. The decreased compressive strength observed in the sample foamed at mid-level pressure could be further explained by the zoomed-in force-position curve in **Figure 4(b)**. Compared to samples foamed at low or high pressures, which exhibit stronger resistance to deformation due to a distinct peak in the elastic-to-plateau transition, those foamed at mid-level pressure display a linear trendline, indicating a less uniform cell structure and, consequently, reduced compressive strength.

C. Water Absorption-Desorption Cycle

Water uptake, a crucial factor for PLA foam as a potential food packaging material, reflects the material's behavior against humidity and contact with water, a key consideration for any food-contact material [22]. Sample exhibiting the highest percentage in density reduction, which may be characterized as super-expanded, will be subject to further analysis to examine this behavior. As depicted in **Figure 5**, all usages show the same trend with a significant increase in the percentage of water uptake after a starting point with the highest value of 41.19% reached in the first usage, specifically at 7200s. Both second and third usage reached almost the same value at around 28.65% water uptake by the end of the same immersion time. Foaming polylactide with supercritical CO_2 significantly affects its hydrophilicity. The cell structure generated by the foaming process, driven by thermodynamic instability, creates readily accessible pathways for water molecule diffusion, enhancing the material's water uptake [22]. While the reusability demonstrated over 3 cycles suggests suitability for sustainable packaging.

It is noteworthy that while our PLA foam offers significant reusability benefits, it is important to acknowledge that its water uptake is comparatively higher than commercially available EPS. This has been reported by Yudanto and Pudjihastuti (2020) [48], who disclosed that EPS had only 2% water uptake in the first 15 minutes. A comparison with a starch/cellulose-based composite foam showed that it had a significantly higher water uptake of 25.45% within the first 15 minutes [49], indicating that PLA's inherent chemical structure is responsible for its lower water uptake. PLA primarily consists of carbon-carbon and carbon-hydrogen bonds with the ester group (–COOR) as its key functional group, which contributes to its slightly increased polarity due to the presence of oxygen atoms in its matrix [50]. This slight increase in polarity is believed to be responsible for the observed water uptake behavior.

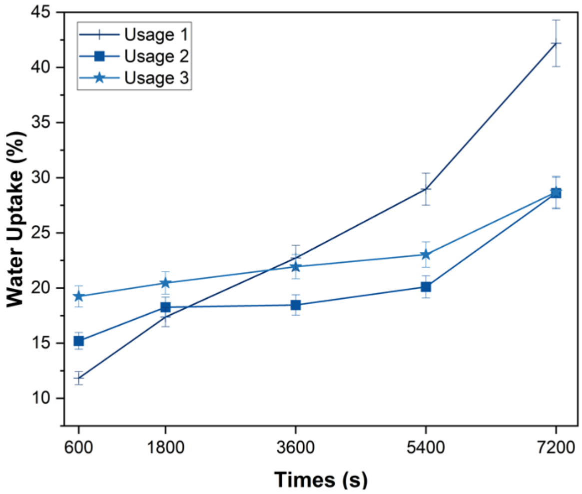

Fig. 5 Water uptake of PLA foam

IV. CONCLUSION

The present study has successfully demonstrated the ability to foam polylactide (PLA), a difficult-to-foam material, using supercritical CO_2 in a pressure-controlled batch process. Through an investigation of the effects of foaming pressure on the resulting material properties, the study aims to develop sustainable alternatives to the environmentally harmful expanded polystyrene (EPS) foams. Results revealed that PLA foams exhibited promising characteristics, including lightweight, enhanced compressive strength, and acceptable water uptake behavior, making them suitable candidates for sustainable packaging applications. FTIR analysis verified the structural integrity of unfoamed and foamed PLA, demonstrating the potential for reliable foaming processes. Foaming effectively reduced material density by incorporating gas bubbles into the polymer matrix under different pressures, resulting in lightweight and expanded products. Higher pressures resulted in denser foams, indicating that lighter foams are obtained at lower pressures. This is due to the opposing effects of CO_2 plasticization and weakened melt strength at higher pressure, limiting cell expansion and leading to less density reduction. The impact of CO_2 plasticization on compressive strength may be caused by promoting the nucleation of dense lamellar crystals, contributing to the higher observed strength. The force-position curve exhibits the typical characteristics of polymer compression: elastic deformation, plateau, and densification. Though PLA foams absorb more water than EPS, their uptake remains lower than many other biodegradable options. Moreover, their reusability and resilience across multiple cycles suggest promising potential for sustainable packaging solutions.

Further research should refine and optimize PLA foaming processes. Exploring potential blowing agents and adjusting processing parameters hold promise for enhancing desired foam properties for the feasibility and applicability of PLA foams in diverse packaging applications.

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CONFLICT OF INTEREST

Authors declare no conflict of interest to disclose.

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