

A New Bundling and Packaging Method Using Nonwoven Polylactide Based on Polymer Shrinkage by Carbon Dioxide

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Abstract: This study proposes the exposure of nonwoven fabrics to carbon dioxide for bundling and packaging purposes. The proposed process, which utilizes the shrinking property of the nonwoven fabric during carbon dioxide exposure, is demonstrated on a polylactic acid (PLA) nonwoven fabric produced by the melt-blown method. Evaluating the shrinkage induced by carbon dioxide in PLA nonwoven fabrics with varying degrees of crystallinity, it was found that increasing the crystallinity decreases both the speed and amount of shrinkage. This process is potentially applicable as a simple, inexpensive, and environmentally friendly approach for packaging food and drug products.

Keywords: shrinkage; carbon dioxide; polylactic acid; packaging; crystallinity

1. Introduction

Developing sustainable societies is a top priority in modern times. Among the key challenges is mitigating climate change [1], in particular, reducing the emissions of carbon dioxide (CO₂), a greenhouse gas that contributes to global warming. Effective uses of CO₂ that has been captured and separated from the atmosphere are also urgently sought [2,3]. CO₂ is used as a food additive and is expected to be applied as a solvent for extraction and synthesis reactions in the food and pharmaceutical industries [4]. High-pressure CO₂ is known to dissolve well in certain plastics [5] and is particularly soluble in amorphous polymers [6,7]. Studies have also explored the interaction of CO₂ with polymers, focusing on its effects on foaming behavior [8] and impregnation [9]. Consequently, several processes combining polymers and CO₂ [10], such as foaming [8], micro-particle synthesis [11], and drug impregnation processing [9], have been proposed. Research has been carried out to create foams using supercritical carbon dioxide blended with polylactic acid for high foam and low shrinkage [12].

The CO₂-assisted polymer compression (CAPC) method plasticizes fibrous plastics with CO₂ and compresses them into polymer porous bodies [13]. This method controls the porosity and other characteristics through the process conditions [14] and facilitates the encapsulation of drugs during the compression of layered fibrous sheets [15]. The method can potentially process relatively inexpensive nonwoven fabrics, extending its practical applicability. Nonwoven fabrics are mass-produced for household, medical, and agricultural products [16] with extremely low production costs. If the CAPC process could utilize low-cost fibers as raw materials, it could not only derive expensive products such as medicines but also everyday products.

Nonwoven fabrics made of fine fibers with low crystallinity, produced using the melt-blown method, have been identified as particularly well-suited for the CAPC method [17]. To further expand the applicability of this method, polymers with high melt flow rates



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(MFRs), suitable for melt-blown nonwoven fabric production, were studied. Among these materials, polylactic acid (PLA) polymers exhibited notable shrinkage behavior under non-restrictive conditions. In a CO₂ environment, this shrinkage behavior was found to resemble that of heat-shrinkable films commonly used in packaging [18], thereby introducing the concept of a CO₂ shrink film. Additionally, the fuzzy surface of the melt-blown nonwoven fabric, which likely generates high friction when layered, was hypothesized to facilitate bundling. This hypothesis was confirmed by demonstrating that the fabric could be effectively bundled by simply wrapping it around an object and exposing it to CO₂.

This study introduces an innovative method for bundling and packaging that leverages the shrinkage effect of gaseous CO₂. The research examines the shrinkage behavior of raw nonwoven fabrics under CO₂ exposure and explores how crystallinity influences both the rate and extent of this shrinkage.

2. Materials and Methods

The raw nonwoven fabric materials were fabricated from two types of PLA as follows: Ingeo Biopolymer 6252D (melt index 70–85 g/10 min (210 °C), ASTM D1238 [19], glass transition temperature 55–60 °C, melting temperature 155–170 °C) from NatureWorks (Plymouth, MN USA); and Luminy L105 (melt-flow index 70 g/10 min (210 °C, 2.16 kg), ISO 1133-A [20], glass transition temperature 60 °C, melting temperature 175 °C) from TotalEnergies Corbion (Gorinchem, The Netherlands). The melt-blown nonwoven fabrics were spun by Nippon Nozzle Co., Ltd. (Kobe, Japan). The nonwoven fabric from TotalEnergies Corbion is gradually crystallized through heat treatment. Therefore, samples of this fabric were heat-treated for 1, 3, or 10 h at 62 °C, as previously reported [17]. All nonwoven fabrics were punched out with a 16.0 mm hand punch, creating circular nonwoven fabric samples with a diameter of 16.0 mm for shrinkage analysis. Hereafter, the nonwoven fabrics procured from NatureWorks and TotalEnergies Corbion will be referred to as NW and TC0, respectively, and the TotalEnergies Corbion fabrics heat-treated at 62 °C for 1, 3, and 10 h are named TC1, TC3, and TC10, respectively. Heat treatment was performed at a temperature slightly above the glass transition temperature to promote gradual crystallization. In a previous study [17], the crystallinity of TC0 increased from 0.02 to 0.17 after heat treatment at 62 °C for 10 h.

For the bundling test, strips of paper cut into long, narrow rectangles were bundled together, wreathed in NW nonwoven fabric, placed in a pressure vessel, and exposed to CO₂. For the packaging test, finely chopped paper was wrapped in NW nonwoven fabric, placed in a pressure vessel, and exposed to CO₂. In both tests, the gas was vented after 30 s of CO₂ exposure and the sample was removed to complete the process.

Figure 1 is a schematic of the equipment used in the shrinkage evaluation test. A piece of nonwoven fabric was placed between polyimide films cut to Φ18 mm and inserted into a cylindrical cylinder with an inner diameter of 20 mm. Valve V₁ was closed and V₂ was opened, lowering the piston to 10 mm above the bottom. Afterward, valve V₂ was closed, V₁ was opened for 2 s, and CO₂ was introduced from the CO₂ cylinder. The experimental temperature was 23 °C, and the pressure of the CO₂ introduced through the piping was 6.1 MPa. The time at which V₁ was closed after CO₂ introduction was defined as the temporal origin. After the specified number of seconds, V₂ was opened to release the CO₂ into the atmosphere and the sample was removed by raising the piston. Seven samples of each material were processed, with a smooth polyimide film placed between each sample. The piston merely acted as a lid, applying no load to the samples; therefore, all samples could freely shrink without restriction.

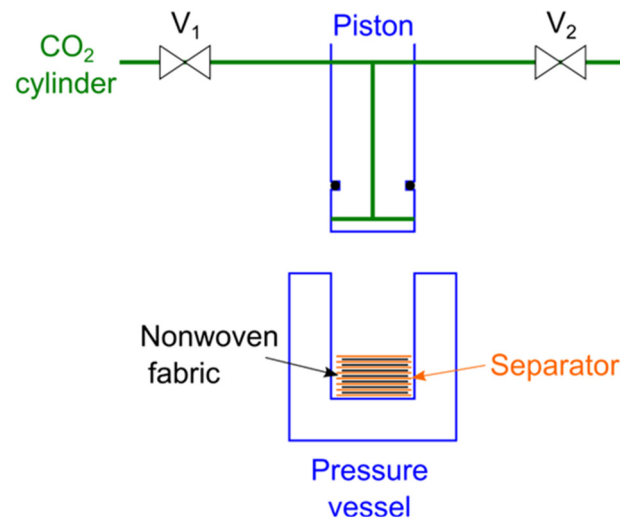


Figure 1. Diagram of the experimental setup for the shrinkage test. V₁: CO₂-introduction valve; V₂: CO₂-exhaustion valve. Samples were placed between smooth-surfaced polyimide films and stacked in the vessel.

To determine the shrinkage area of the nonwoven fabric, a piece of the fabric was placed on a 20 mm by 20 mm black square sheet of paper and covered with a glass plate. After photographing the sample, the area was calculated through image analysis. Specifically, the vertices of the black paper were extracted from the photograph and the image was converted into a square. Subsequently, the contour of the nonwoven fabric was extracted and the number of pixels within the contour was counted and converted into an area. The areas of the seven samples were evaluated for calculating the mean and standard deviation.

3. Results and Discussion

First, the results of bundling the NW nonwoven fabric are presented. Figure 2A shows the paper strips (approximately 1 mm wide) and the nonwoven fabric cut for bundling. Panels B and C of Figure 2 show a typical sample before placement in the container and after CO₂ exposure, respectively. CO₂ exposure appeared to align and tighten the paper strips. Similarly, Figure 3A shows the paper finely chopped into squares with approximate side lengths of 1 mm and the nonwoven fabric cut for wrapping. Panels B and C of Figure 3 show a typical sample before placement in the container and after CO₂ exposure, respectively. The originally wrapped section was thick and appears solid white, but after CO₂ exposure, the material shrank and adhered tightly to the contents, revealing the color of its contents. As nonwoven fabrics contain voids between fibers, liquids and gasses can permeate through these voids. This process can be potentially used to wrap coffee or herbs, creating coffee or herbal-tea tablets, respectively. The process requires no heat input and uses nontoxic CO₂, so it is safe for food applications and for encapsulating potentially heat-degradable materials.

In a process utilizing shrinkage in a CO₂ environment, the extent and duration of the CO₂-induced shrinkage must be elucidated. To this end, the NW PLA nonwoven fabric and TC0 PLA nonwoven fabric previously used in the CAPC study were subjected to a shrinking experiment. As heat treatment can alter the crystallinity of TC0, the impact of crystallinity on shrinkage was evaluated on samples with varying degrees of crystallinity.



Figure 2. Bundling treatment. Results of CO₂ exposure on 20 bundles of bar-shaped paper samples (approximate paper width 1 mm): (A) bundle of paper and nonwoven fabric for bundling, (B) the bundled sample wreathed in nonwoven fabric; (C) the sample after treatment.

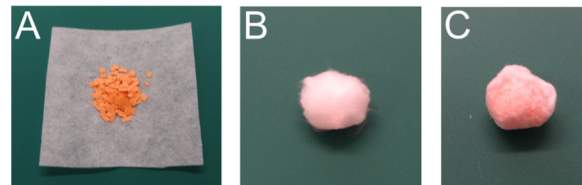


Figure 3. Packaging treatment. Results of CO₂ exposure on paper squares (approximate side length 1 mm) wrapped in the nonwoven fabric: (A) pile of paper squares and nonwoven fabric for packaging; (B) sample of paper squares wrapped in nonwoven fabric; (C) the sample after treatment.

First, the samples subjected to various heat treatments were punched out with a hand punch and their areas were reported as the averages and standard deviations of seven samples. The areas were determined as $201.0 \pm 0.4 \text{ mm}^2$ for NW, $201.1 \pm 0.4 \text{ mm}^2$ for TC0, $201.3 \pm 0.7 \text{ mm}^2$ for TC1, $202.0 \pm 0.5 \text{ mm}^2$ for TC3, and $201.9 \pm 0.5 \text{ mm}^2$ for TC10. The evaluation results of the area of all punched samples are consistent with the calculated area of a circle with a 16.0 mm diameter (201 mm^2), confirming that punching with a hand punch and image analysis are appropriate evaluation methods for the present experiment.

Figure 4 shows photographs of the samples before and after CO₂ exposure. In these images, the black backing paper was converted into a square and the contours were extracted. Owing to irregularities in the nonwoven fabric, the shrunk fabric was noncircular, so the evaluation was based on the area of the contour-extracted shape. The experimental results are summarized in Table 1. The values in the table represent the average of seven samples, and the errors indicate the standard deviation. While TC3 and TC10 showed minimal shrinkage, the remaining samples exhibited initial shrinkage, which eventually converged to a constant value. Since this behavior resembles an exponential trend, exponential fitting was applied. The data from Table 1 were plotted on a graph and the fitting results are presented in Figure 5. The markers indicate the averages and standard deviations of the seven samples, and the lines are the results of fitting with an exponential function. The results of TC3 and TC10 are not easily fitted to an exponential function and were instead fitted to a straight line, indicating no temporal change. In Figure 5, the vertical axis of the graph represents the area (201 mm^2 for the original sample). At time 0, shrinkage was already progressing as the CO₂ introduction valve had been opened for two seconds prior. After CO₂ exposure, the NW fabric originally measuring 201 mm^2 shrunk to 64% (to 128.6 mm^2). NW exhibited the highest area shrinkage among the evaluated samples. Converting the shrinkage rate to the square root of the area, the length shrinkage was determined as 80%. The higher shrinkage rate of NW compared to the TC samples was attributed to the higher MFR (indicating higher fluidity) of NW compared to TC. As the heat-treatment duration increased from TC0 to TC10, the samples became less prone to shrinkage. According to a previous study [17], the crystallinities of TC0, TC1, TC3, and TC10 are 0.02, 0.07, 0.15, and 0.17, respectively, suggesting that increasing the crystallinity decreases both the speed and amount of shrinkage. As CO₂ presumably dissolves into the amorphous regions of the polymer, the effect of crystallinity on shrinkage is unsurprising.

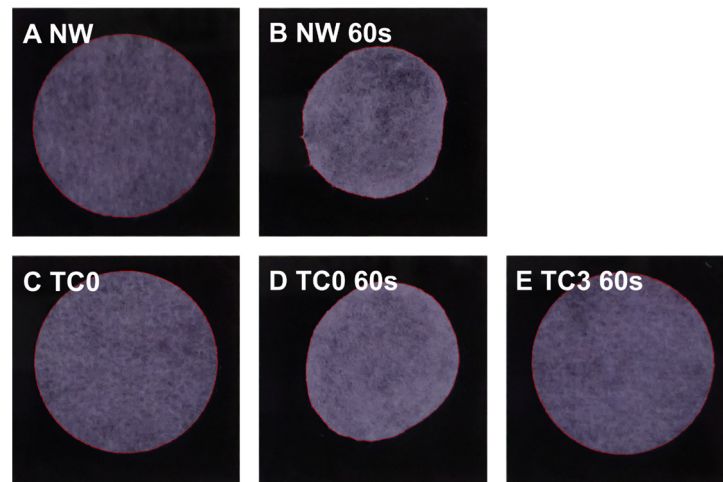


Figure 4. Photographs of the samples before and after shrinkage: NatureWorks (NW) nonwoven fabric (A) before CO₂ treatment and (B) after 60 s of CO₂ treatment; TotalEnergies Corbion (TC) nonwoven fabric without heat treatment (C) before CO₂ treatment and (D) after 60 s of CO₂ treatment; TC nonwoven fabric heat-treated for 3 h (E) after 60 s of CO₂ treatment. The black squares (side length 20 mm) were corrected to squares from an image taken with a digital camera. The areas of the nonwoven fabrics were estimated by counting the number of pixels in the image enclosed by the extracted outlines (highlighted in red around the nonwoven fabrics).

Table 1. Results of shrinkage experiments. Errors are standard deviations of 7 samples.

Time [s]	Area of NW [mm ²]	Area of TC0 [mm ²]	Area of TC1 [mm ²]	Area of TC3 [mm ²]	Area of TC10 [mm ²]
0	162.3 ± 8.1	168.0 ± 4.6	189.2 ± 5.1	195.9 ± 2.9	201.1 ± 0.7
5	130.0 ± 6.1	152.5 ± 3.2	184.6 ± 9.3	199.1 ± 3.8	202.4 ± 3.2
10	134.8 ± 3.1	143.8 ± 5.2	189.7 ± 10.7	191.0 ± 10.3	194.9 ± 5.1
15	120.8 ± 8.6	148.8 ± 7.0	183.2 ± 8.5	197.0 ± 7.9	201.2 ± 2.9
20	123.0 ± 9.4	143.2 ± 8.0	188.8 ± 6.4	195.4 ± 12.9	201.4 ± 3.8
25	123.0 ± 15.4	153.1 ± 10.1	178.3 ± 6.8	197.8 ± 5.1	203.3 ± 1.4
30	132.8 ± 6.9	150.8 ± 6.0	185.7 ± 9.6	196.4 ± 5.3	201.5 ± 2.9
40	132.7 ± 12.4	145.5 ± 8.2	174.5 ± 7.7	198.1 ± 2.6	203.5 ± 2.4
50	128.8 ± 12.0	144.3 ± 12.1	186.8 ± 7.6	195.0 ± 5.0	203.6 ± 1.5
60	133.9 ± 6.2	148.0 ± 9.3	183.3 ± 11.0	200.1 ± 3.1	204.1 ± 1.5

NW: NatureWorks nonwoven fabric; TC0: TotalEnergies Corbion nonwoven fabric without additional heat treatment; TC1, TC3, and TC10: TC0 heat treated at 62 °C for 1 h, 3 h, and 10 h, respectively.

From the observed decrease in heat shrinkage, the fundamental cause of shrinkage was inferred as strain accumulation during spinning. The nonwoven fabric was produced using the melt-blown method, in which the molten polymer is extruded from a nozzle, stretched by air, and rapidly cooled. The strain accumulated during this process is probably relieved by shrinkage when the polymer is plasticized by CO₂ and gains fluidity.

Figure 6 shows the scanning electron microscopy images of several samples. As the samples were not pressed, the indentations formed by the compression of the fibers observed in a previous CAPC study are absent in these images. Even after exposure to CO₂ for 60 s, the NW nonwoven fabric resembled its untreated counterpart. The TC samples also presented no obvious changes after exposure to CO₂ for 60 s.

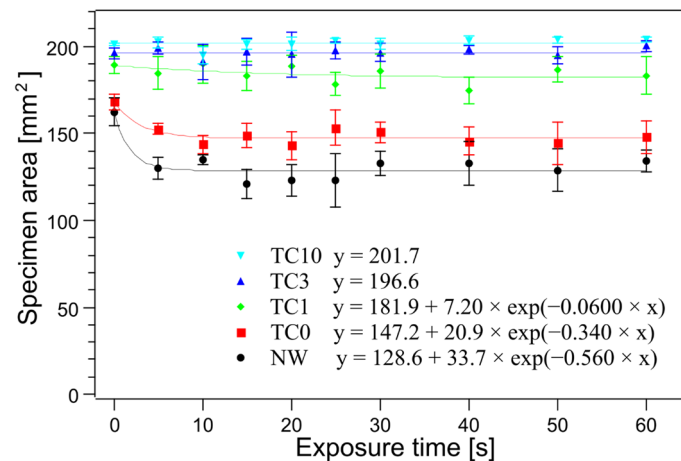


Figure 5. Results of shrinkage experiments. Markers are the means of 7 samples. Error bars are standard deviations and lines are least-squares fitting results. Fitting results are shown in the figure. NW: NatureWorks nonwoven fabric; TC0: TotalEnergies Corbion nonwoven fabric without additional heat treatment; TC1, TC3, and TC10: TC0 heat treated at 62 °C for 1 h, 3 h, and 10 h, respectively.

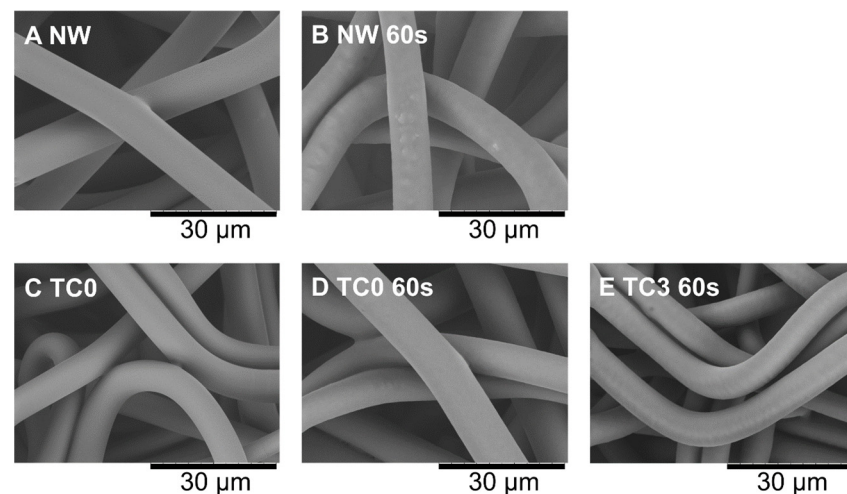


Figure 6. Scanning electron microscope (SEM) images at 3000x of NatureWorks (NW) nonwoven fabric (A) before CO₂ treatment and (B) after 60 s of CO₂ treatment; SEM images of TotalEnergies Corbion (TC) nonwoven fabric (C) without heat treatment and before CO₂ treatment, (D) without heat treatment after 60 s of CO₂ treatment, and (E) after 3 h of heat treatment and 60 s of CO₂ treatment.

4. Conclusions

It was demonstrated that nonwoven fabrics fabricated from PLA using the melt-blown method are suitable binding and packaging materials under CO₂ exposure. This process is extremely simple and is as follows: the material only needs to be wreathed or wrapped and exposed to a CO₂ environment. The process exploits the shrinkage property of nonwoven fabric during CO₂ exposure. Under the same CO₂ exposure conditions, a material with lower crystallinity shrank more quickly and more extensively than the same material with a high crystallinity. As the CO₂ exposure treatment involves no heat, it is expected to be suitable for binding or enclosing heat-sensitive items. In addition, as the nonwoven fabric is a porous material, it is potentially applicable to drug delivery and other applications requiring the controlled release of enclosed agents in liquid. The proposed method shows potential for encapsulating dry goods such as coffee or herbal teas. However, further investigation is required to assess the structural stability and potential organoleptic effects of PLA-based packaging when in contact with hot liquids.

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