

# ROLL-TO-ROLL PROCESSING OF SUSTAINABLE PACKAGING MATERIALS USING MODIFIED BIOPOLYMERS

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## Abstract

Packaging is a vital requirement for the effective handling of electronic, food, textile, medical, and pharmaceutical items. The demand for more environmentally-friendly, biodegradable, and sustainable materials in various engineering and scientific applications has been a driving factor behind the use of biopolymers for packaging, which traditionally has been dominated by petroleum-based plastics. One approach being investigated is the use of biopolymer-based barrier coatings on renewable and/or biodegradable non-barrier substrates, such as paper, to generate effective packaging solutions with commercially useful mechanical and barrier properties.

The objective of this work is to investigate the feasibility of a scalable fabrication process for single- and bilayer coatings of cationic polymers applied to paper that is super-calendared and lightly silicone-converted to reduce fluid penetration through the paper. A single or dual-layer slot die, integrated within a roll-to-roll system, was used to uniformly coat thin films of the biopolymers as independent or blended fluids on the paper at a flow rate and web velocity within the process window. For bilayer coatings, the modified polymers were coated simultaneously with cellulose nanocrystals (CNC) to create bilayer coatings. Barrier properties were examined by carrying out oxygen transmission rate (OTR) tests at a relative humidity of 50%. Oxygen transmission rates ranging from 2 – 30,500 cm<sup>3</sup>/m<sup>2</sup>/day were observed. Performance trends are hypothesized to arise from differences in hydrogen bonding and strong electrostatic attraction between CNC and the biopolymer coating components.

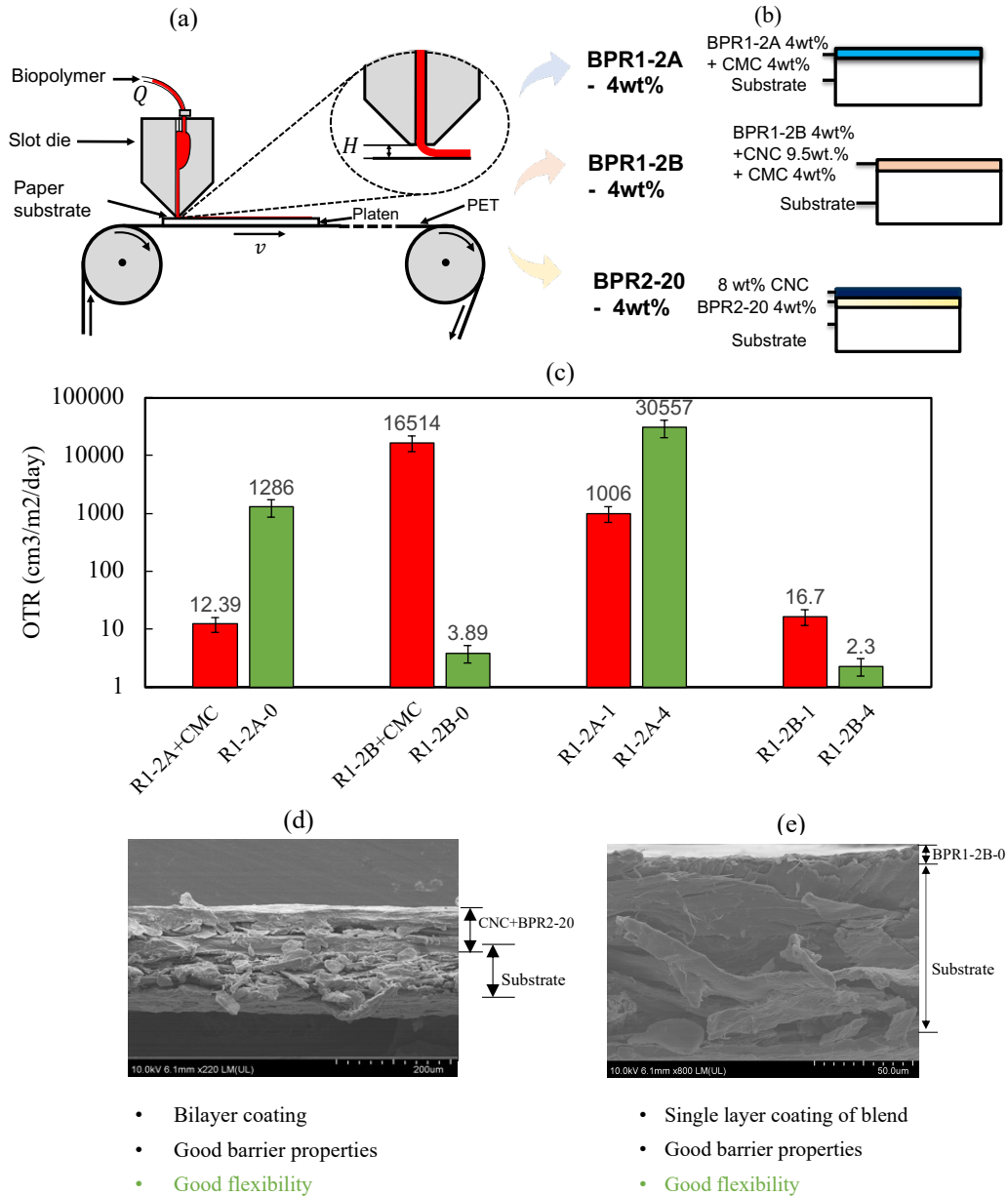
**Keywords:** Packaging, CNC, modified polymers, bilayer coating, renewable coating, slot die

## 1.0 Methods:

Biopolymers have been coated on renewable material substrates to improve their barrier properties<sup>1-5</sup>. In this work, modified polymers were coated on paper along with other biopolymers such as cellulose nanocrystals (CNC) and carboxy-methyl cellulose (CMC). The modified polymers used in this work were BPR1-2A, BPR1-2B, and BPR2-20. BPR1-2A solution was blended with CMC, while the BPR1-2B solution was blended with CMC and CNC. The blends were coated on paper using a single layer slot die integrated within the roll-to-roll system as shown in Figure 1 while BPR2-20 was coated simultaneously with CNC using a dual layer slot die<sup>5</sup> (similar to Figure 1a, but with two slots). The coated solutions were dried in an oven at 90 °C for about 15 minutes. The flexibility of the materials was assessed using a fold Kit test which is similar to the standard Kit test<sup>7</sup> but in this case, the reagents were applied to the coated materials along the edges that have been folded once.

## 2.0 Results:

The quality of the coatings was assessed by oxygen transmission rate (OTR) tests and fold Kit tests. Figure 1c illustrates the results for blends at different weight ratios. The OTR test results indicate that BPR1-2B-4 performed best with an OTR value of  $2.3 \text{ cm}^3/\text{m}^2/\text{day}$ . The bilayer sample BPR2-20+CNC also exhibited good barrier properties with an OTR value of  $11.7 \text{ cm}^3/\text{m}^2/\text{day}$ . The materials also had fold Kit numbers greater than 10 indicating good flexibility. The SEM images in Figures 1e and 1f show the cross-sections of the coated samples for BPR2-20+CNC and BPR1-2B-0 respectively. The coatings show promising results and potential for renewable packing applications.



**Figure 1:** Modified polymers used for coating on paper to improve barrier properties. (a) Slot die coating technique was used to coat the polymers on paper. (b) Polymers BPR1-2A, and BPR1-2B were coated along with CNC and CMC as blends while BPR2-20 was coated as a bilayer along with CNC. (c) OTRs as low as  $2.3 \text{ cm}^3/\text{m}^2/\text{day}$  were obtained for the blends. The plots show the blends at different weight ratios. (d) SEM image of cross-section of bilayer BPR2-20 with CNC coated on paper, (e) SEM image of cross-section of BPR1-2B blend coated on paper.

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### References:

1. Ji, Y., Waters, S., Lim, E., Lang, A. W., Ciesielski, P. N., Shofner, M. L., Reynolds, J. R., & Meredith, J. C. (2022). Minimizing Oxygen Permeability in Chitin/Cellulose Nanomaterial Coatings by Tuning Chitin Deacetylation. *ACS Sustainable Chemistry and Engineering*, 10(1), 124–133. <https://doi.org/10.1021/acssuschemeng.1c05051>
2. Satam, C. C., Irvin, C. W., Lang, A. W., Jallorina, J. C. R., Shofner, M. L., Reynolds, J. R., & Meredith, J. C. (2018). Spray-Coated Multilayer Cellulose Nanocrystal - Chitin Nanofiber Films for Barrier Applications. *ACS Sustainable Chemistry and Engineering*, 6(8), 10637–10644. <https://doi.org/10.1021/acssuschemeng.8b01536>
3. Yu, Z., Ji, Y., Bourg, V., Bilgen, M., & Meredith, J. C. (2020). Chitin-and cellulose-based sustainable barrier materials: a review. *Emergent Materials*, 3, 919–936. <https://doi.org/10.1007/s42247-020-00147-5/Published>
4. Yu, Z., Ji, Y., & Meredith, J. C. (2022). *Multilayer Chitin–Chitosan–Cellulose Barrier Coatings on Poly(ethylene terephthalate)*. <https://doi.org/10.1021/acsapm.2c01059>
5. Jung, K., Ji, Y., Jeong, T. J., Ciesielski, P. N., Meredith, J. C., & Harris, T. A. L. (2022). Roll-to-Roll, Dual-Layer Slot Die Coating of Chitin and Cellulose Oxygen Barrier Films for Renewable Packaging. *ACS Applied Materials and Interfaces*, 14(39), 44922–44932. <https://doi.org/10.1021/acsmi.2c09925>
6. Ding, X., Fuller, T. F., & Harris, T. A. L. (2013). Predicting fluid penetration during slot die coating onto porous substrates. *Chemical Engineering Science*, 99, 67–75. <https://doi.org/10.1016/j.ces.2013.05.039>
7. Hubbe, M. A., & Pruszynski, P. (2020). Greaseproof paper products: A review emphasizing ecofriendly approaches. *BioResources*, 15(1), 1978–2004. <https://doi.org/10.15376/biores.15.1.1978-2004>