Biaxial Roll Milling for Sustainable Polymer Valorization and Additive-Free Performance Enhancement

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Brandi Martz, Tiffany Lee, Kennedy Brehm, Daniel Strauss, Gary Wnek,
Case Western Reserve University, Department of Macromolecular Science and Engineering
Zhigang Xu, Christopher Hale, Jagannathan Sankar
North Carolina Agriculture and Technical State University, Department of Mechanical Engineering

Introduction

Cold rolling is a well-established solid-state deformation processing method used in the steel industry to create ductile workpieces of uniform thickness. In the last 70 years, roll milling has emerged as an industrially-relevant method to enhance the mechanical, optical and barrier properties of petroleum-based polymers.

Cold rolling addresses many manufacturing sustainability concerns as it is:

- scalable
- solvent-less*
- scrap-less
- heat-minimized
- economical

*if necessary, limit to e.g. H_2O, ethanol, 2-propanol

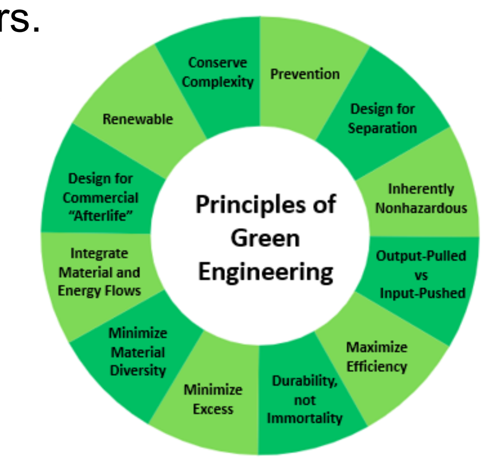

Figure 1 Principle of Green Engineering

Utilizing shear and compression, cold rolling can be used to overcome the mechanical limitations of biodegradable, recycled and blends of immiscible polymers. The present work explores the use of biaxial, single and differential speed rolling to improve the mechanical performance of these inherently brittle, low-cost, commercially available polymer classes.

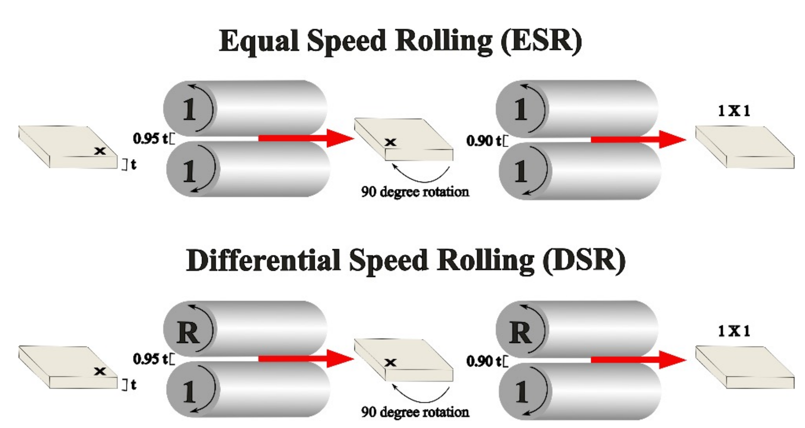

Figure 2 Roll milling processes. Top: equal speed rolling. Bottom: differential speed rolling.

Biodegradable Polymers

Poly(L-lactic acid) is one renewably-sourced alternative to petroleum-based polymers. This industrially-compostable polyester is most prevalent in the packaging, biomedical, and consumer products industries. However, widespread usage of PLLA is limited by it's inherent brittleness and low impact toughness. With rolling:

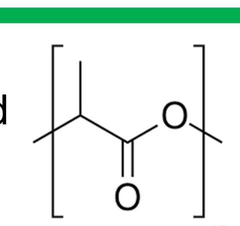

Figure 3 Chemical structure of Poly (L-lactic acid)

Amorphous (<10%)	Low crystallinity (35%)	Moderate crystallinity (45%)
5x tensile toughness	**14x** tensile toughness	**14x** tensile toughness
5x flexural toughness	**5x** flexural toughness	**5x** flexural toughness
82% increase in impact strength	**20%** increase in impact strength	**95%** increase in impact strength

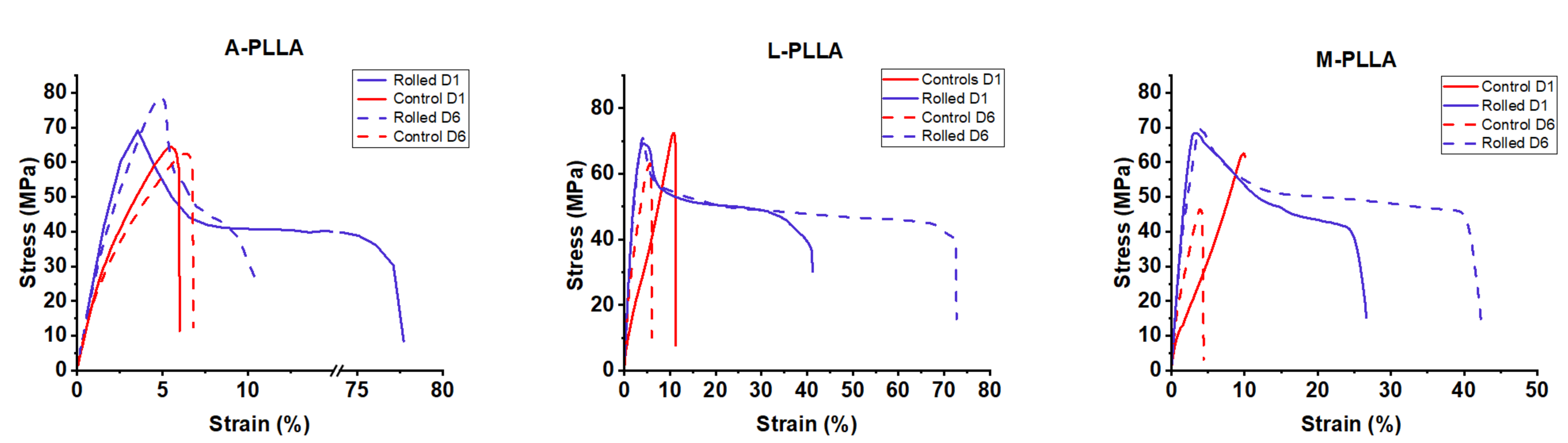

Figure 4 Tensile stress-strain curves for amorphous PLLA (left), low crystallinity PLLA (middle), and moderate crystallinity PLLA (right).

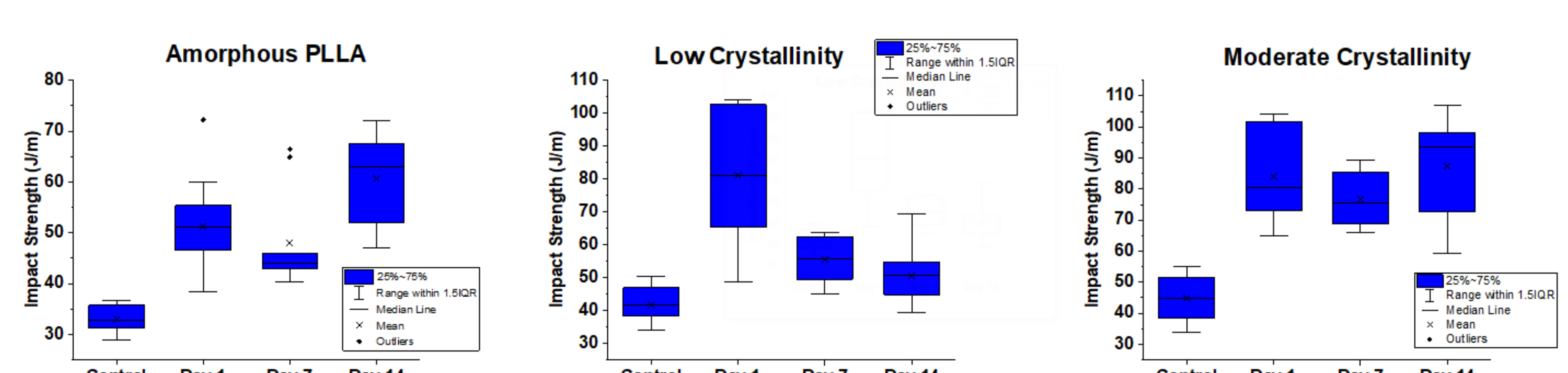

Figure 5 Impact strength for amorphous PLLA (left), low crystallinity PLLA (middle), and moderate crystallinity PLLA (right).

Abstract

Sustainability in the polymers industry is a multifaceted challenge that requires readily available processing technologies that can handle variable polymer feedstocks while delivering reliable, high-value products. The adaptation of metal-forming technologies, such as roll milling, offer flexible, low-cost, and practical solutions to valorize bio-based, recycled, and blended polymers without the monetary, temporal, and performance limitations of other methods. Unlike other valorization techniques, roll milling is a continuous, solventless, scrapless, and low-heat process that can be easily integrated into existing manufacturing environments. Additionally, roll milling promotes material circularity by using commercial resins and enhancing mechanical performance without additives, fillers, or copolymerization. This work highlights the versatility of roll milling, using both equal-speed rolling (dominantly compressive deformation) and differential-speed rolling (enhanced shear deformation), to improve the mechanical performance of bio-based PLLA, recycled polypropylene, and polyethylene-polypropylene blends. Results show significantly increased tensile toughness and improved impact strength across material types along with suppressed physical aging in bio-based PLLA. Overall, these findings establish roll milling as a scalable, industry-ready technology capable of converting diverse polymer feedstocks into higher-performance and commercially competitive materials.

Future Directions

The adaptation of metal-forming technologies to polymer processing offers significant potential to tailor polymer morphology and, consequently, enhance mechanical properties critical to industrial and commercial applications. Areas of further interest include:

- Different blend compositions (PS, PET, PHAs)
- Recycled PE/PP blends
- Optimization of the rolling process
- Quantification of the environmental impact of rolling

Acknowledgements and References

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1. The Recycling Partnership . *Polypropylene Recycling Coalition Annual Report 2024*; LaValley, B., DesRoberts, E., Eds.; 2024; pp. 1–19. https://recyclingpartnership.org/wp-content/uploads/dlm_uploads/2024/10/PolyPro_AnnualReport_10.30.24.pdf (accessed 2025-12-02).

Immiscible Polyolefin Blends

Polyolefins like polyethylene (PE) and polypropylene (PP) comprise upwards of 60% of all plastics production with applications in consumer products, automotive, healthcare, packaging, and construction. Small differences in their molecular structures prevent molecular-level mixing, thus producing PE/PP blends with weak interfaces and, subsequently, poor mechanical performance. With rolling:

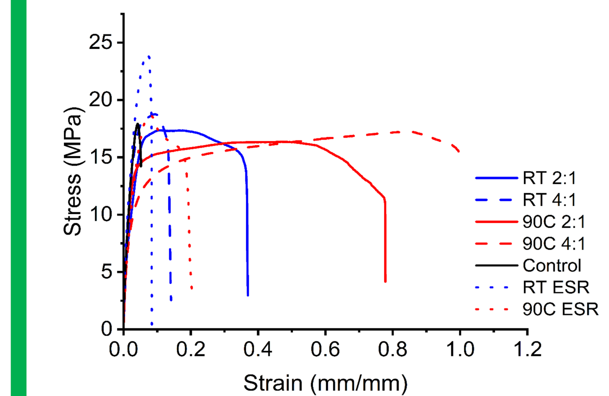

Figure 6 Tensile stress-strain curve of HDPE:PP (50:50) blend.

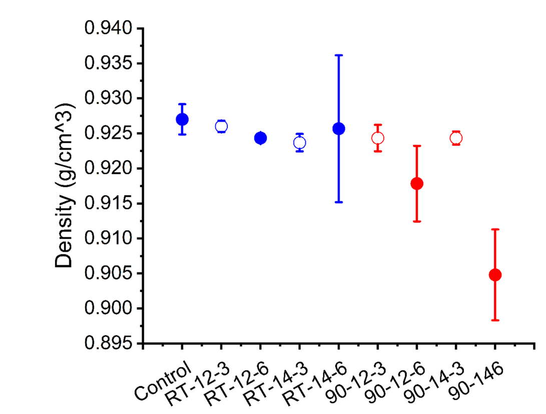

Figure 7 Density of HDPE/PP (50:50) blends

20x increase in tensile toughness (DSR 90, 4:1)
Decreased density
Fibrous morphology

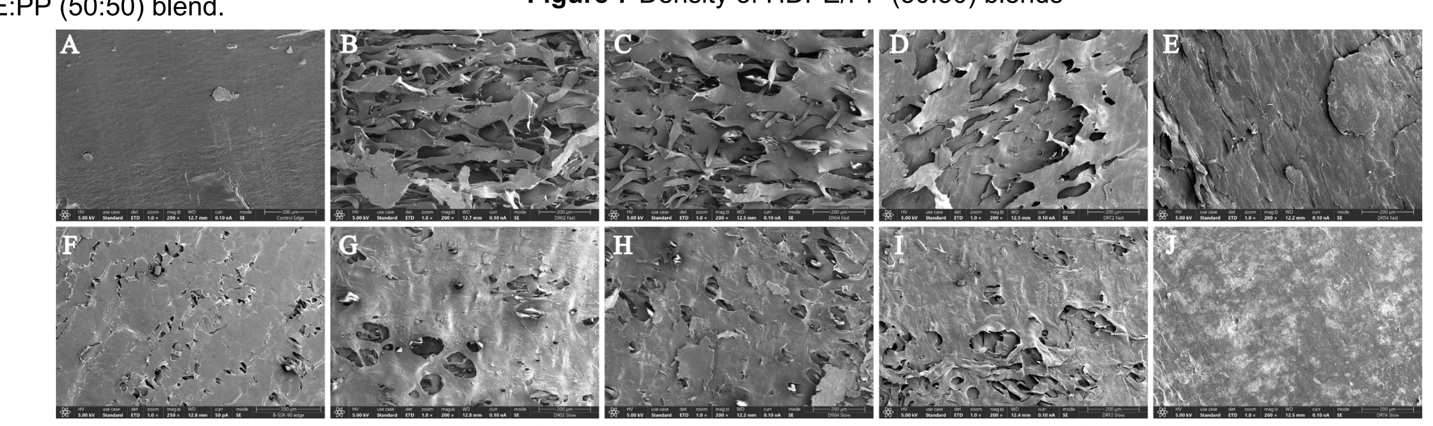

Figure 8 SEM images of rolled surfaces at 200 um. A: Control, B: 90C 2:1 Fast, C: 90C 4:1 Fast. D: RT 2:1 Fast, E: RT 4:1 Fast, F: ESR 90, G: 90C 2:1 Slow, H: 90C 4:1 Slow, I: RT 2:1 Slow, J: RT 4:1 Slow

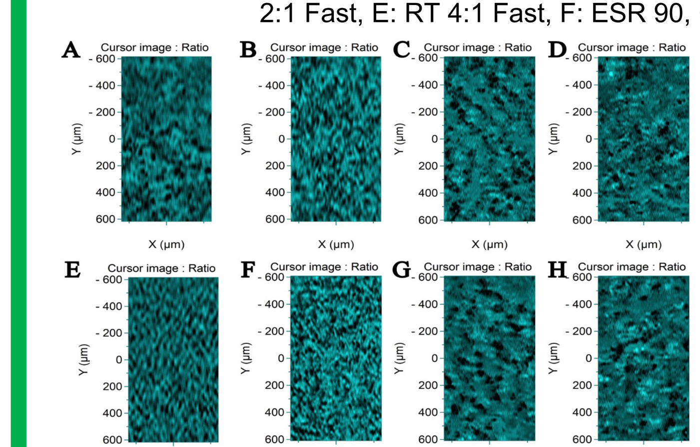

Figure 9 Confocal Raman Spectroscopy of samples. A: Control fracture surface, B: DSR-90, 2:1 fracture surface, C: DSR-90, 2:1 Slow rolled surface, D: DSR-90, 2:1 Fast rolled surface, E: ESR RT rolled surface, F: DSR-90, 4:1 fracture surface, G: DSR-90, 4:1 Slow rolled surface, H: DSR-90, 4:1 Fast rolled surface.

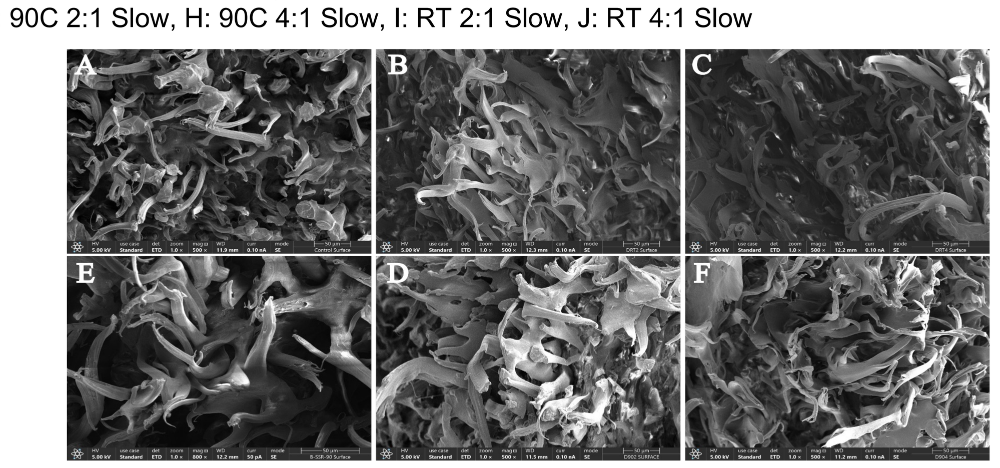

Figure 10 SEM images of HDPE:PP (50:50) samples at 50 um. A: Control samples, B:Single speed rolling, C: 90C-2:1 speed ratio, D: RT-2:1 speed ratio, E: 90C 4:1 speed ratio, F: RT-4:1 speed ratio

Recycled Polymers

Approximately 2 billion pounds of polypropylene waste Is generated in the US each year.[1] The loss of Mechanical properties associated with mechanical recycling, the high cost of collecting, sorting, and transporting recyclable materials, and downstream issues associated with chemical compatibilizers result in only 2% of waste returning to a similar quality application. With rolling:

3258% increase in Clear tensile toughness
542% increase in Red Dyed tensile toughness
197% increase in Blue Dyed Impact Strength

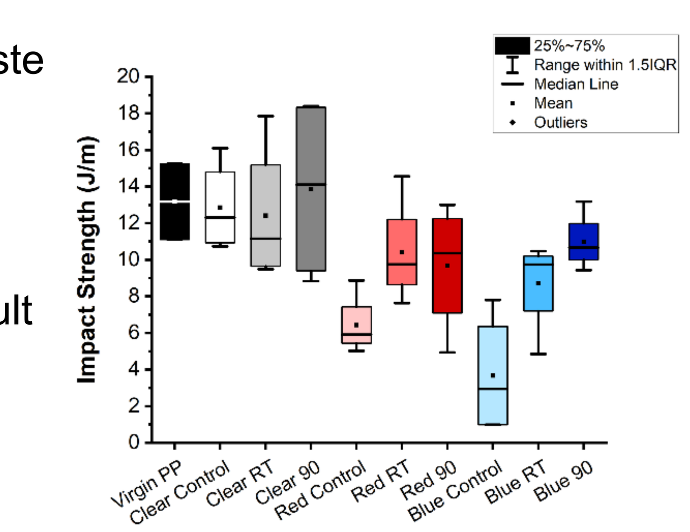

Figure 11 Impact strength of recycled PP

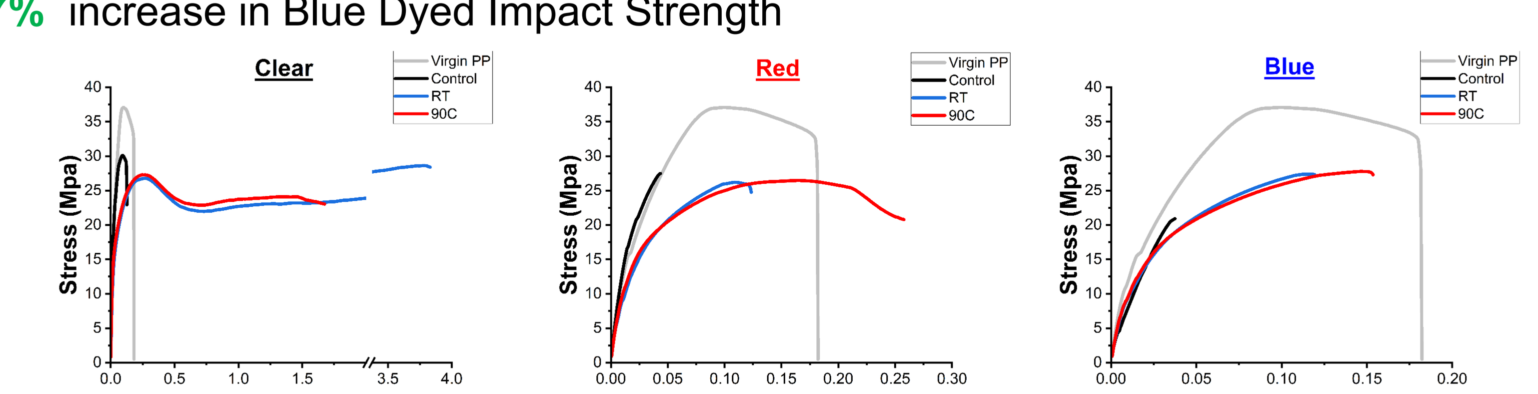

Figure 12 Tensile stress-strain curves of recycled PP. Clear PP (left), Red-dyed PP (middle), Blue-dyed PP (right).

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