Fabrication of Self-Standing Bio-Based Films Containing Hemicelluloses and Pectin

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It is no secret that plastics—particularly single-use plastics— are deeply ingrained into our world and lives today. This is problematic for various reasons, including health concerns (Endo, et al. 2005; Andrady, 2011; Cox, et al., 2019), marine litter (Willis, et al., 2018), and plastic waste causing disproportionate effects for those in developing countries (Browning, et al., 2021).

A commonly used type of single-use synthetic plastics is transparent, flexible films. While recycling is possible (though uncommon), there can be a decline in quality of the recycled material (Horodytska, et al., 2018). Additionally, synthetic-based plastics are non-renewable and non-biodegradable causing various environmental concerns. To potentially replace these materials with a more sustainable alternative, research focused on the development of biodegradable composite films using polymers such as pectin and hemicellulose commenced.

Pectin is a natural material obtained from plants (commonly from fruits), and it is an edible polysaccharide that is commonly used in the food industry (Chen, et al., 2020). Hemicellulose is another natural polysaccharide that is found in plants and constitutes up to 35% of the biomass in plants (Egüés, et al., 2013; Gírio, et al., 2010). Since polysaccharides are promising materials to create oxygen barriers, both pectin and hemicellulose are materials of interest for film application (Gröndahl, et al., 2004).

The purpose of the following research is to develop biocomposite films using a combination of galactomannan (a type of hemicellulose) from tara gum and different sources of pectin (apple and soybean hulls) as potential alternatives for traditional synthetic films. To develop the films, solutions containing varying ratios of tara gum and pectin (apple or soy pectin) with glycerol (at an amount of approximately 25% of total film solid weight, which was 0.5 g) as plasticizer were prepared. The compositions are displayed in Table 1. The solutions were stirred on a magnetic stirrer for 1 hour at 50 ^{0}C. The solutions were then poured into polystyrene petri dishes and dried in an oven at 35°C for 5 days. The films were then characterized using various analytical techniques.

Table. 1. Film Compositions

Hemicellulose (% of Solids)	Pectin (% of Solids)
0	100
20	80
40	60
60	40
80	20
100	0

**Note: The amount of glycerol and ultrapure water remained constant at 25 wt.% and 40ml, respectively, in all formulations.*

Preliminary characterizations were done on the films, and, for the purpose of this highlight, the most prominent results will be discussed. All characterizations took place after films remained in a desiccator at least overnight.

Film thickness was measured ten times with micro calipers on each film. The average of the ten measurements was then taken to obtain the thickness value for each composite film. Values are displayed on Table 2. The thickness ranged from 0.071mm to 0.133mm. Though

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the films were prepared with a constant mass of material, there are many different factors that influence the thickness such as drying conditions and interactions between materials used in the films (Sood & Saini, 2022), so this will require further investigation.

Table. 2. Film Thickness

Film Thickness (mm)			
Apple Pectin/Hemicellulose Composite Films		Soybean Hull Pectin/Hemicellulose Composite Films	
Film (% Pectin)	Thickness (mm)	Film (% Pectin)	Thickness (mm)
0*	0.09	0*	0.09
20	0.09	20	0.09
40	0.08	40	0.13
60	0.08	60	0.07
80	0.07	80	0.07
100	0.09	100	0.09

**100% hemicellulose films*

The contact angle of water was measured on a selection of the films. More investigation is necessary to obtain a comprehensive view of this characterization, but the results from the soybean hull pectin composite films could indicate a potential lignin contamination due to increasing zero-time hydrophobicity as pectin content increases. The film with 100% hemicellulose had a zero-time contact angle of 40.30°, while that with 100% pectin had an initial contact angle of 67.23°.

FTIR analysis was performed for each of the films, and the data from the 100% and 60% pectin films and 100% hemicellulose films is displayed on Figure 1 and Figure 2 for apple pectin/hemicellulose composite films and soy pectin/hemicellulose films, respectively. Both spectra indicate the presence of the peak at 3400 to 3300 cm^{-1}, indicating the OH stretching vibration of inter- and intramolecular hydrogen bonding (Sood & Saini, 2022), whereas the bands between 3000 and 2500 cm^{-1} is related to the CH, CH_2, and CH_3 stretching and bending vibrations (Egüés, et al., 2013). The intense peak observed around 1740 cm^{-1} corresponds to the absorption of the esterified carboxylic groups (Sood & Saini, 2022) of the pectin, whereas the absorbance band around 1200 to 980 cm^{-1} could be ascribed to the presence of hemicellulose (Cerqueira, et al., 2011). A peak around 1515 cm^{-1} in the soybean hull pectin/hemicellulose spectra can be attributed to the aromatic vibrations (Boeriu, et al., 2004). This could indicate presence of lignin in the soybean hull pectin, although further characterizations are required to ascertain the claim.

Overall, developing flexible composite films with the materials in the percentages is possible, which could provide the opportunity to diversify application fields. Moving forward, more films will be made to obtain more comprehensive data. The films will also be subjected to mechanical testing.

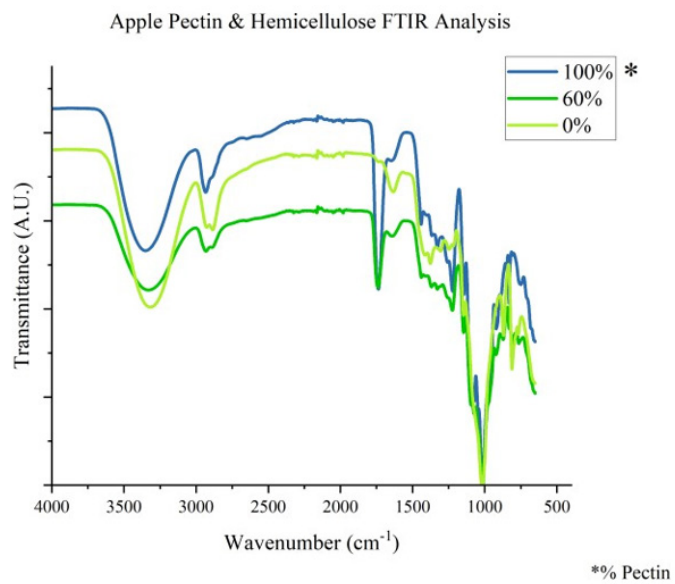

Fig. 1. FTIR Spectra of Apple Pectin Composite Films

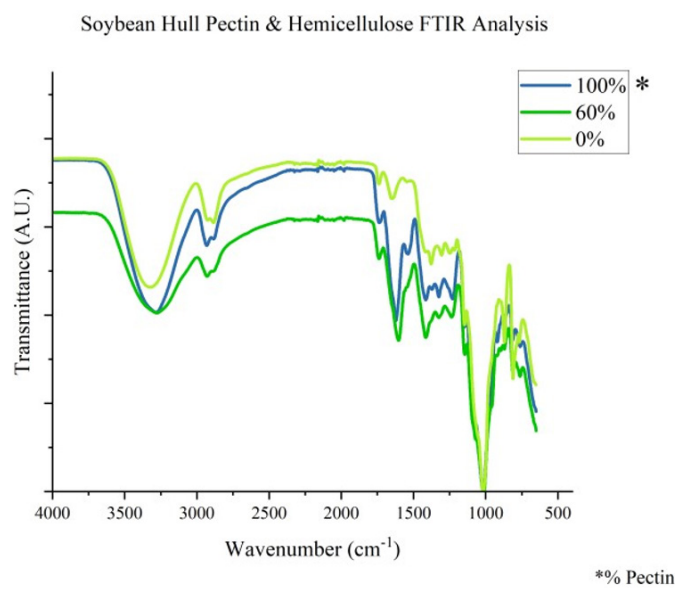

Fig. 2. FTIR Spectra of Soybean Hull Pectin Composite Films

Statement of Research Advisor

Evie has actively developed strategies to develop formulations to cast composite films, while overcoming challenges related to deaeration of the solutions and homogenization to improve film formation. Additionally, Evie has planned and executed experimentation, data collection and analysis. During this period, Evie has shown a great level of maturity in carrying independent work, as well as critical thinking.

- Maria Soledad Peresin, College of Forestry, Wildlife and Environment

References

[1] Andrady, A. L. (2011). Microplastics in the marine environment. Marine pollution bulletin, 62(8), 1596-1605.

[2] Boeriu, C. G., Bravo, D., Gosselink, R. J., & van Dam, J. E. (2004). Characterisation of structure-dependent functional properties of lignin with infrared spectroscopy. Industrial crops and products, 20(2), 205-218.

[3] Browning, S., Beymer-Farris, B., & Seay, J. R. (2021). Addressing the challenges associated with plastic waste disposal and management in developing countries. Current Opinion in Chemical Engineering, 32, 100682.

[4] Cerqueira, M. A., Souza, B. W., Simões, J., Teixeira, J. A., Domingues, M. R. M., Coimbra, M. A., & Vicente, A. A. (2011). Structural and thermal characterization of galactomannans from non-conventional sources. Carbohydrate polymers, 83(1), 179-185.

[5] Chen, Y., Xu, L., Wang, Y., Chen, Z., Zhang, M., & Chen, H. (2020). Characterization and functional properties of a pectin/tara gum based edible film with ellagitannins from the unripe fruits of Rubus chingii Hu. Food chemistry, 325, 126964.

[6] Cox, K. D., Covernton, G. A., Davies, H. L., Dower, J. F., Juanes, F., & Dudas, S. E. (2019). Human consumption of microplastics. Environmental science & technology, 53(12), 7068-7074.

[7] Egüés, I., Eceiza, A., & Labidi, J. (2013). Effect of different hemicelluloses characteristics on film forming properties. Industrial Crops and Products, 47, 331-338.

[8] Endo, S., Takizawa, R., Okuda, K., Takada, H., Chiba, K., Kanehiro, H., ... & Date, T. (2005). Concentration of polychlorinated biphenyls (PCBs) in beached resin pellets: variability among individual particles and regional differences. Marine pollution bulletin, 50(10), 1103-1114.

[9] Gírio, F. M., Fonseca, C., Carvalheiro, F., Duarte, L. C., Marques, S., & Bogel-Łukasik, R. (2010). Hemicelluloses for fuel ethanol: a review. Bioresource technology, 101(13), 4775-4800.

[10] Gröndahl, M., Eriksson, L., & Gatenholm, P. (2004). Material properties of plasticized hardwood xylans for potential application as oxygen barrier films. Biomacromolecules, 5(4), 1528-1535.

[11] Horodytska, O., Valdés, F. J., & Fullana, A. (2018). Plastic flexible films waste management–A state of art review. Waste management, 77, 413-425.

[12] Risch, S. J. (2009). Food packaging history and innovations. Journal of agricultural and food chemistry, 57(18), 8089- 8092.

[13] Sood, A., & Saini, C. S. (2022). Red pomelo peel pectin based edible composite films: Effect of pectin incorporation on mechanical, structural, morphological and thermal properties of composite films. Food Hydrocolloids, 123, 107135.

[14] Willis, K., Maureaud, C., Wilcox, C., & Hardesty, B. D. (2018). How successful are waste abatement campaigns and government policies at reducing plastic waste into the marine environment?. Marine Policy, 96, 243-249.

Authors Biography

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