Rheology and Adhesion Properties of Cellulose-Derived Green Adhesives

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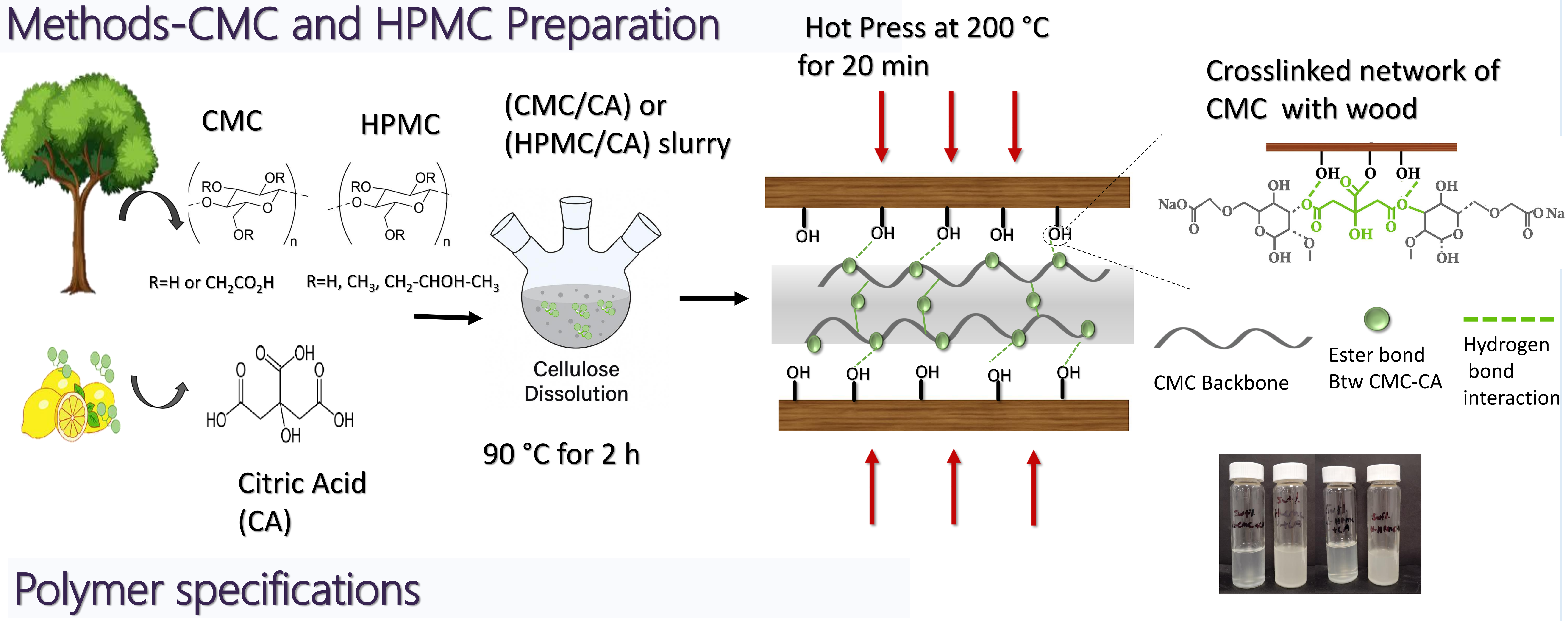
Introduction

- **Conventional wood adhesives** (e.g., UF, MUF, phenolic resins) offer strong bonding but release harmful formaldehyde, raising environmental and health concerns. **Bio-based alternatives** like carboxymethylcellulose (**CMC**) and Hydroxypropyl Methyl Cellulose (**HPMC)** are biodegradable, renewable, and can be chemically modified for improved adhesion.
- **Citric acid (CA)** is used as a green crosslinker to enhance bonding via esterification; this study examines how olecular weight and substitution degree affect adhesion to wood.

Aim

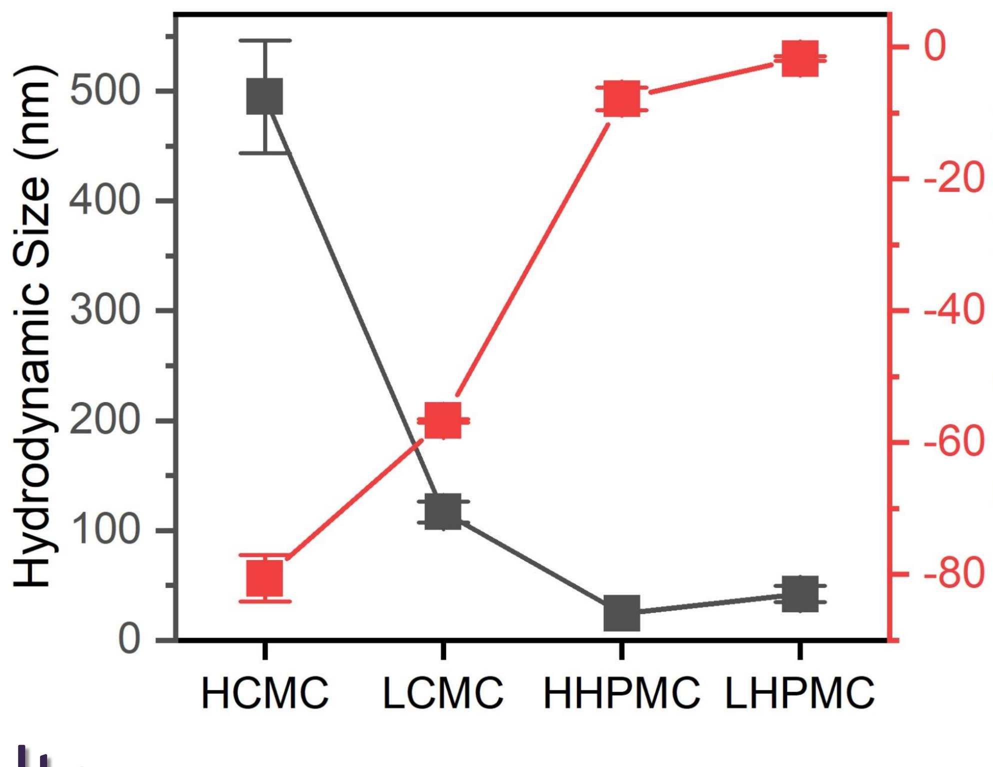
- This study aimed to explain the performance of cellulose-based wood adhesives including the different cellulose derivatives high-viscosity carboxymethyl cellulose (**H-CMC**), low-viscosity carboxymethyl cellulose (**L-CMC**), high-viscosity hydroxypropyl methylcellulose (**H-HPMC**), and low-viscosity hydroxypropyl methylcellulose (**L-HPMC**) by systematically investigating effects of molecular weight, degree of substitution, and free –OH content. By controlling viscosity, the study highlights how higher MW and moderate DS which lead to higher free –OH content and stronger polymer–substrate interactions — play a more dominant role in improving shear strength.

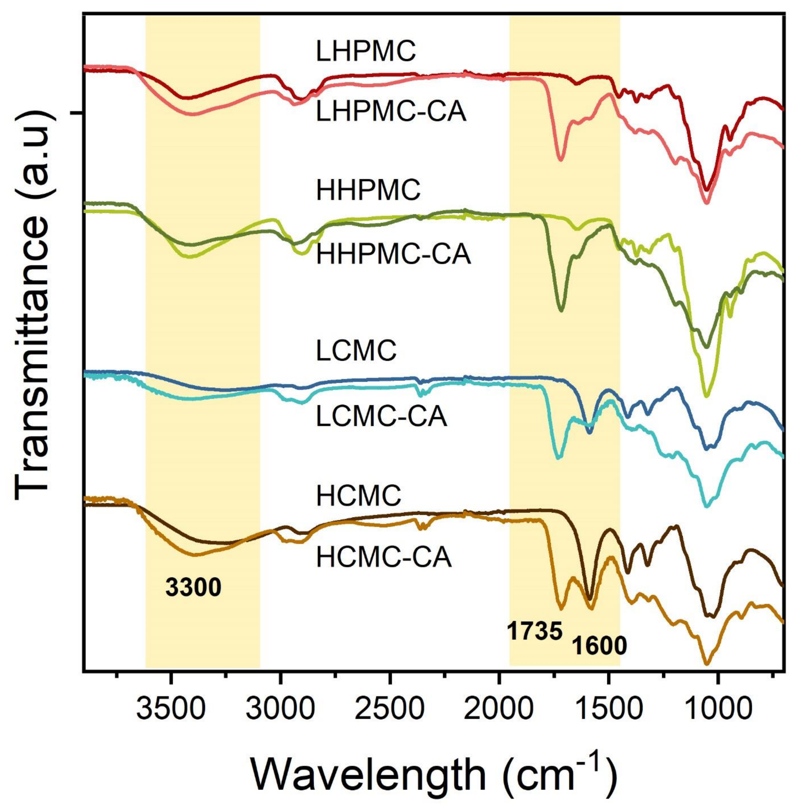
Methods-CMC and HPMC Preparation



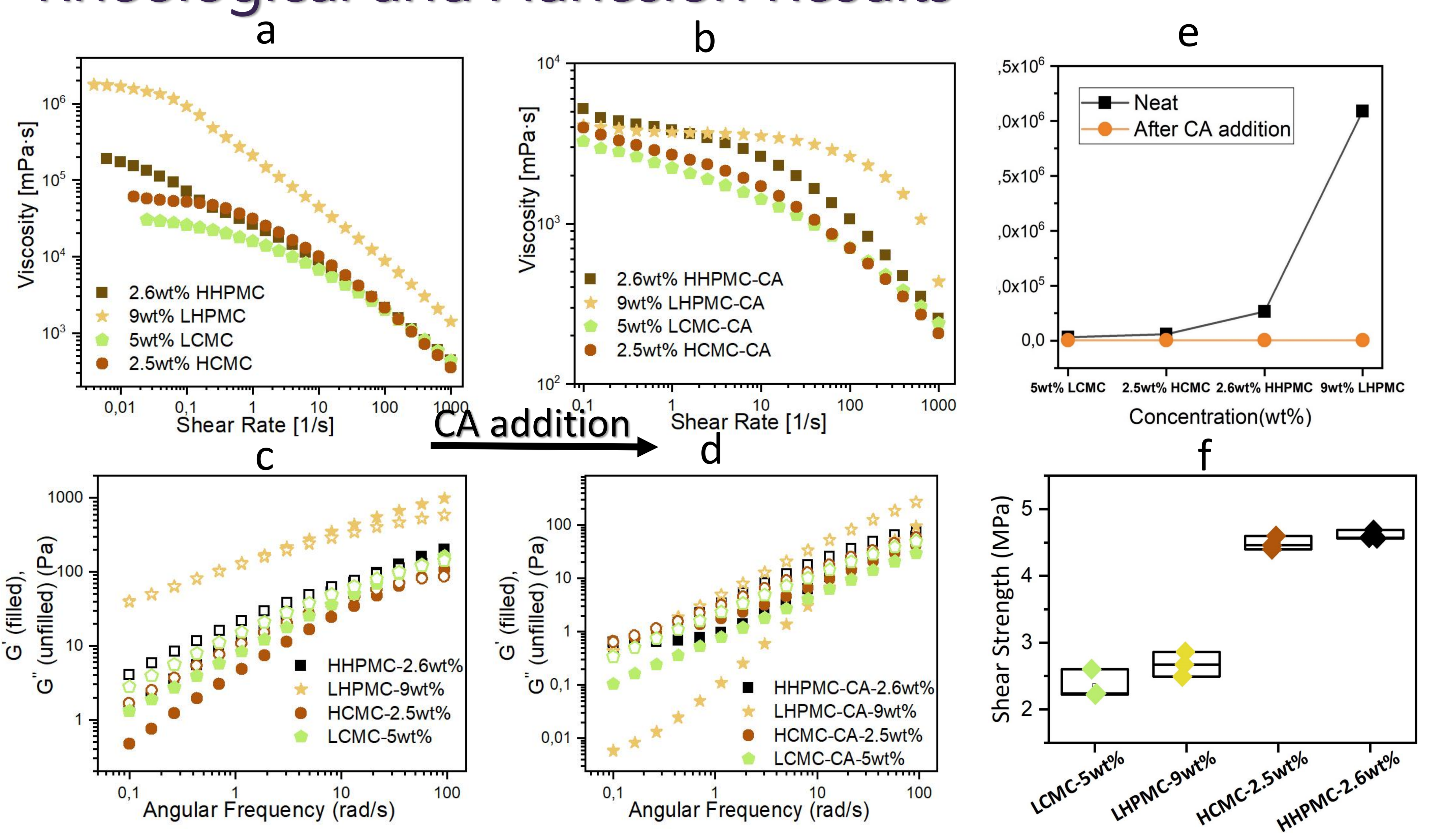
Polymer specifications

Polymer	MW (g/mol)	[η] (dL/g)	DS	Free OH per chain
L-CMC	2.83×10^5	12.51	0.67-CM	3280
L-HPMC	3.11×10^5	23.16	0.66-MeO 0.30-HP	3162
H-HPMC	6.57×10^5	46.65	0.66-MeO 0.18-HP	7336
H-CMC	4.53×10^6	178.97	0.97-CM	20739





Rheological and Adhesion Results



CA addition reduces viscosity and viscoelastic moduli (G′, G″) across all samples, especially in L-HPMC. While H-HPMC and H-CMC maintain higher moduli due to stronger chain interactions and molecular weight, L-CMC and L-HPMC show more pronounced loss, reflecting weaker network structures and lower entanglement.

Conclusion

- **H-HPMC** and **H-CMC** maintain balance between **flowability (post-CA)** and **network formation**, enabling deeper wood penetration and efficient crosslinking — explaining their superior shear strength.
- In contrast, **L-HPMC** and **L-CMC**, though rich in free –OH groups, shows lower adhesion—likely due to poor substrate interaction and limited chain entanglement. These results highlight that beyond viscosity, both polymer architecture and functional group accessibility critically influence adhesive efficiency.

References

(1) Shang, X.; Li, C.; Xi, X.; Wei, Z.; Wu, Z.; Lei, H.; Du, G. Biomass cellulose-based wood adhesive with improved water resistance and bonding strength by esterification reaction. *Industrial Crops and Products* **2024**, *222*, 119667.
(2) Cai, L.; Chen, Y.; Lu, Z.; Wei, M.; Zhao, X.; Xie, Y.; Li, J.; Xiao, S. Citric acid/chitosan adhesive with viscosity-controlled for wood bonding through supramolecular self-assembly. *Carbohydrate Polymers* **2024**, *329*, 121765.

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