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Bio-polyurethane structural adhesive reinforced with core shell rubber for improved impact strength

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Abstract

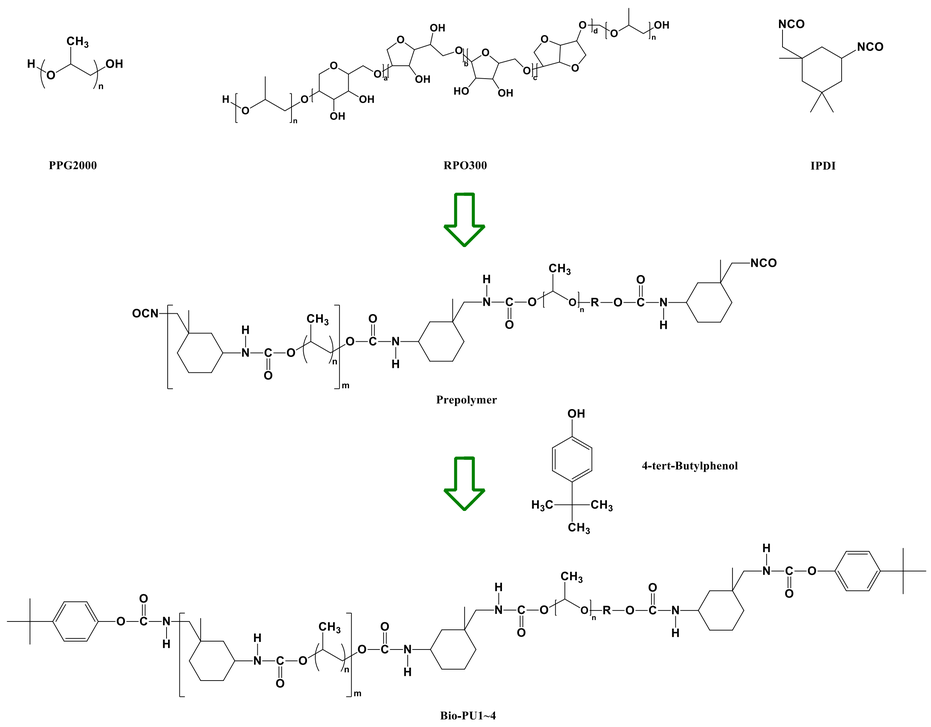
Polyurethane adhesives were synthesized using polypropylene glycol (PPG2000), isosorbide based polyol (RPO300) as polyol, isophorone diisocyanate (IPDI) as isocyanate and 4-tert-butylphenol (BP) as capping agent. The reference sample (Ref.) was synthesized by polymerizing PPG2000 and IPDI without the biomass material. Three different equivalent ratios of PPG2000 and RPO300 were evaluated: 0.9:0.1 (Bio-PU1), 0.7:0.3 (Bio-PU2), and 0.5:0.5 (Bio-PU3). The polymerization progress was confirmed through FT-IR (Fourier Transform Infrared Spectroscopy) and NCO% titration, while molecular weight measurements were performed using GPC (Gel Permeation Chromatography). Thermal properties were analyzed using TGA (Thermogravimetric Analyzers) and DSC(Differential Scanning Calorimeter) equipment. Among the three bio-based polyurethane samples, Bio-PU2 exhibited the best thermal properties and impact strength. Subsequently, 1 wt%, 3 wt%, and 5 wt% of CSR containing polyurethane acrylate in the shell were introduced into the Bio-PU2 sample to evaluate mechanical properties. Shear strength and impact strength were measured using a UTM (Universal Testing Machine).

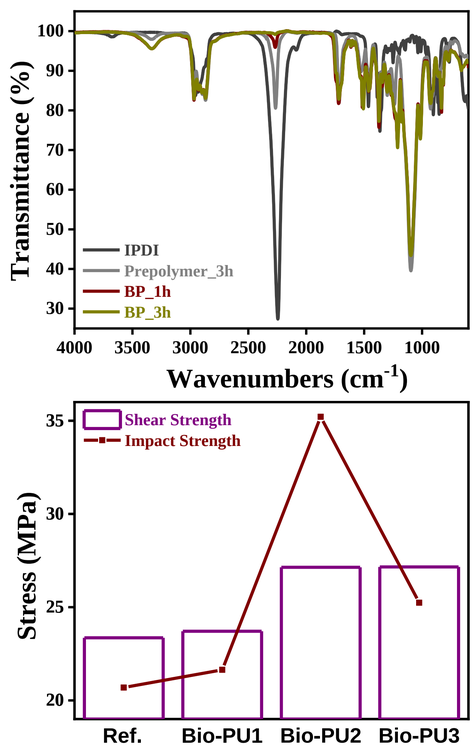
The results indicate that incorporating CSR into the biomass-based polyurethane adhesive significantly enhanced the impact strength without compromising shear strength. The optimal formulation showing the maximum impact strength while maintaining the shear strength was confirmed. In this study paves the way for the development of high-performance, environmentally friendly polyurethane structural adhesives with superior thermal and mechanical properties.

Experimental & Result (Bio-PU)

Scheme

Use of biomass material (RPO300) based on petroleum polyol (PPG2000) prepolymer
-. Set BP capping to Ref. for prepolymer using PPG2000 and IPDI (optimal mixing ratio design)
→ Polymerization proceeds with a total of three equivalent ratios of PPG2000 and RPO300





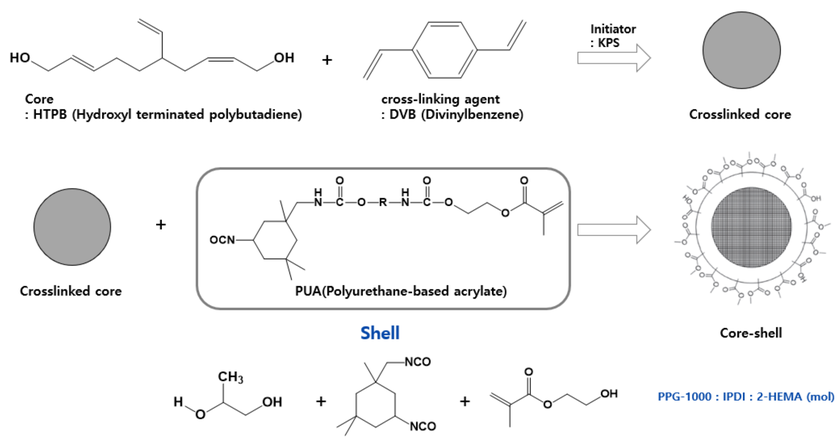
	Polyol		Isocyanate	GPC data			NCO% titration		Thermal properties		Mechanical properties	
	PPG2000 (eq)	RPO300 (eq)	IPDI (eq)	Mn (g/mol)	Mw (g/mol)	PDI (Mw/Mn)	Theoretical value	Measures	T_d	T_g	Shear Strength (MPa)	Impact Strength (MPa)
Ref.	1.0	0	2.0	7716	9706	1.26	3.44	3.68	138.5	-56.3	23.36	20.69
Bio-PU1	0.9	0.1	2.0	7934	9985	1.26	3.77	3.77	143.4	-54.4	23.71	21.64
Bio-PU2	0.7	0.3	2.0	7396	8984	1.21	4.26	4.31	153.9	-52.9	27.14	35.21
Bio-PU3	0.5	0.5	2.0	8049	10090	1.25	5.06	5.19	144.1	-35.2	27.16	25.24

Experimental & Result (CSR)

Scheme

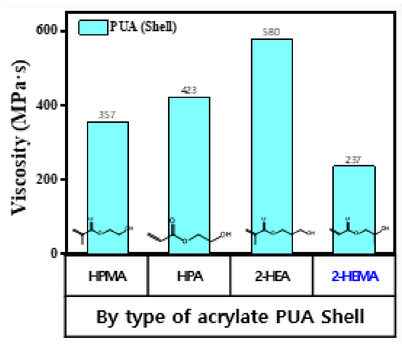
CSR (Core Shell Rubber) Polymerization

[Core] HTPB, surfactant (SDS), crosslinker (DVB), initiator (KPS) content Split
--> Derivation of optimal mixing ratio
[Shell] Using PUA (Polyurethane-based acrylate)
-. Addition of MMA after polymerization of prepolymer to lower viscosity (for the purpose of adding shell and polymerization)



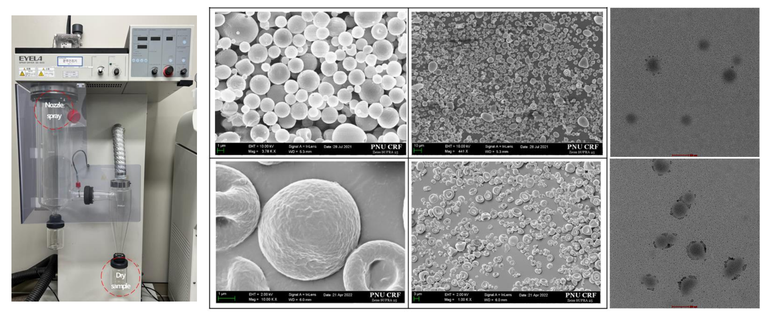
Viscosity (PUA)

Acrylate	Viscosity (25°C, mPa·s)
HPMA	357
HPA	423
2-HEA	580
2-HEMA	237



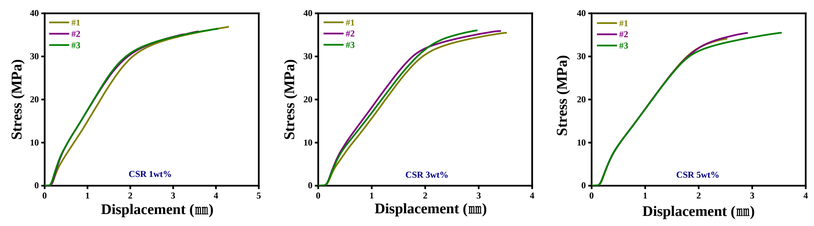
Surface analysis (FE-SEM/TEM)

Spray drying → Surface analysis result → uniform particle size of about 5 ~ 6㎛



Mechanical properties

As the wt% of CSR increases, the shear strength decreases
→ But it is maintained as much as the existing EPOXY



Conclusion

- ☐ Successful synthesis of bio-based polyurethanes and core shell rubber
- ☐ Improving impact strength of bio-based polyurethane
- ☐ Securing a constant pore size of 3-6㎛ through spray drying of CSR
- ☐ Add an appropriate amount of CSR to epoxy → Maintain shear strength

Acknowledgement

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