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Catalyst-free synthesis of high-strength, reprocessable, degradable, and shape-memory biobased Poly(β-hydroxyurethane)s

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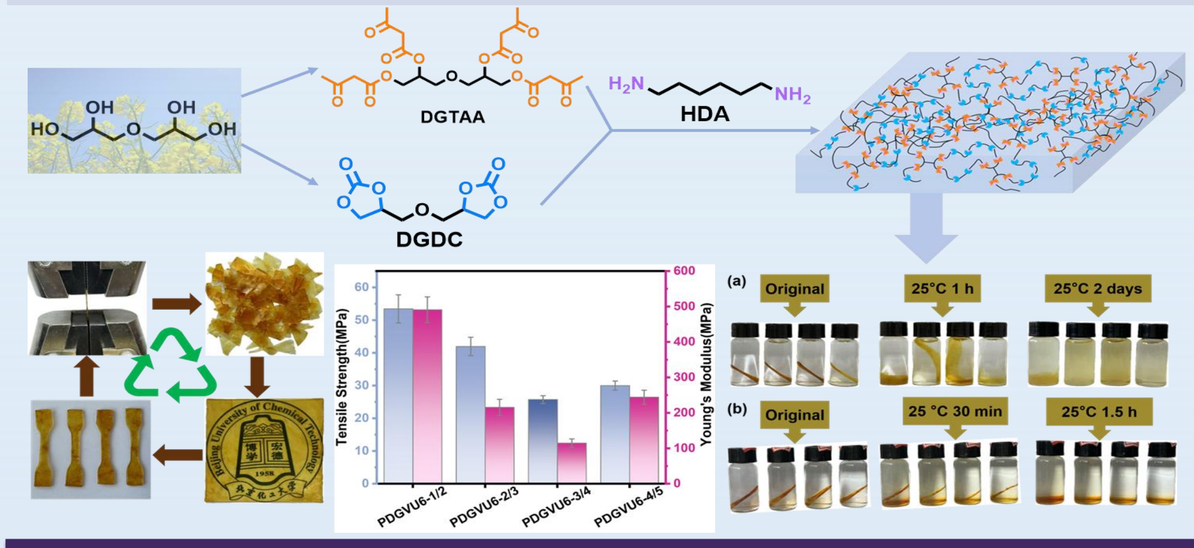
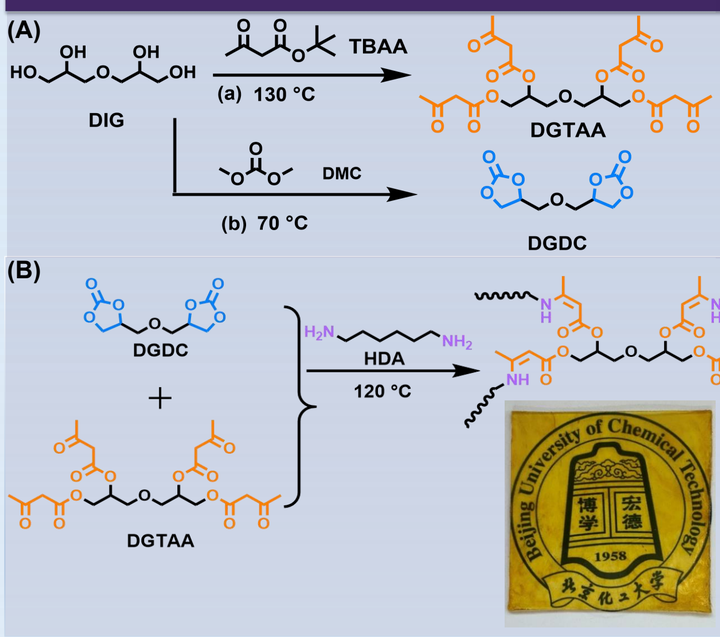
INTRODUCTION & AIM

◆ Poly(β-hydroxyurethane) (PHUs) derived from five-membered cyclic carbonates and amines, is a promising alternative to conventional polyurethanes. However, common PHUs relying on hydroxyl/carbamate exchange exhibit poor reprocessing efficiency and non-degradability [1]. This study addresses these limitations by developing high-performance, reprocessable, and degradable PHUs from renewable feedstocks, advancing in sustainable chemistry. Therefore, novel cross-linked PHU networks (PDGVU6s) with vinylogous urethane bonds [2] were prepared. This study presents an efficient method for synthesizing PDGVU6s from renewable resources, which enriches the synthetic diversity and holds a promise for further exploration in sustainable chemistry.



METHOD



Scheme 1 (A) The synthesis routes of DGTAA(a) and DGDC (b) ; (B) The Synthesis route of PDGVU6s.

(A) First converted into diglycerol dicyclic carbonate (DGDC) through a transesterification with excess dimethyl carbonate, and (B) next reacted with tert-butyl acetoacetate (TBAA) to prepare diglycerol tetraacetoacetate (DGTAA) as a green crosslinking agent. Subsequently, DGDC and DGTAA were reacted with 1,6-hexanediamine to yield a series of PHU vitrimers with vinylogous urethane (VU) bonds (PDGVU6s).

RESULTS & DISCUSSION

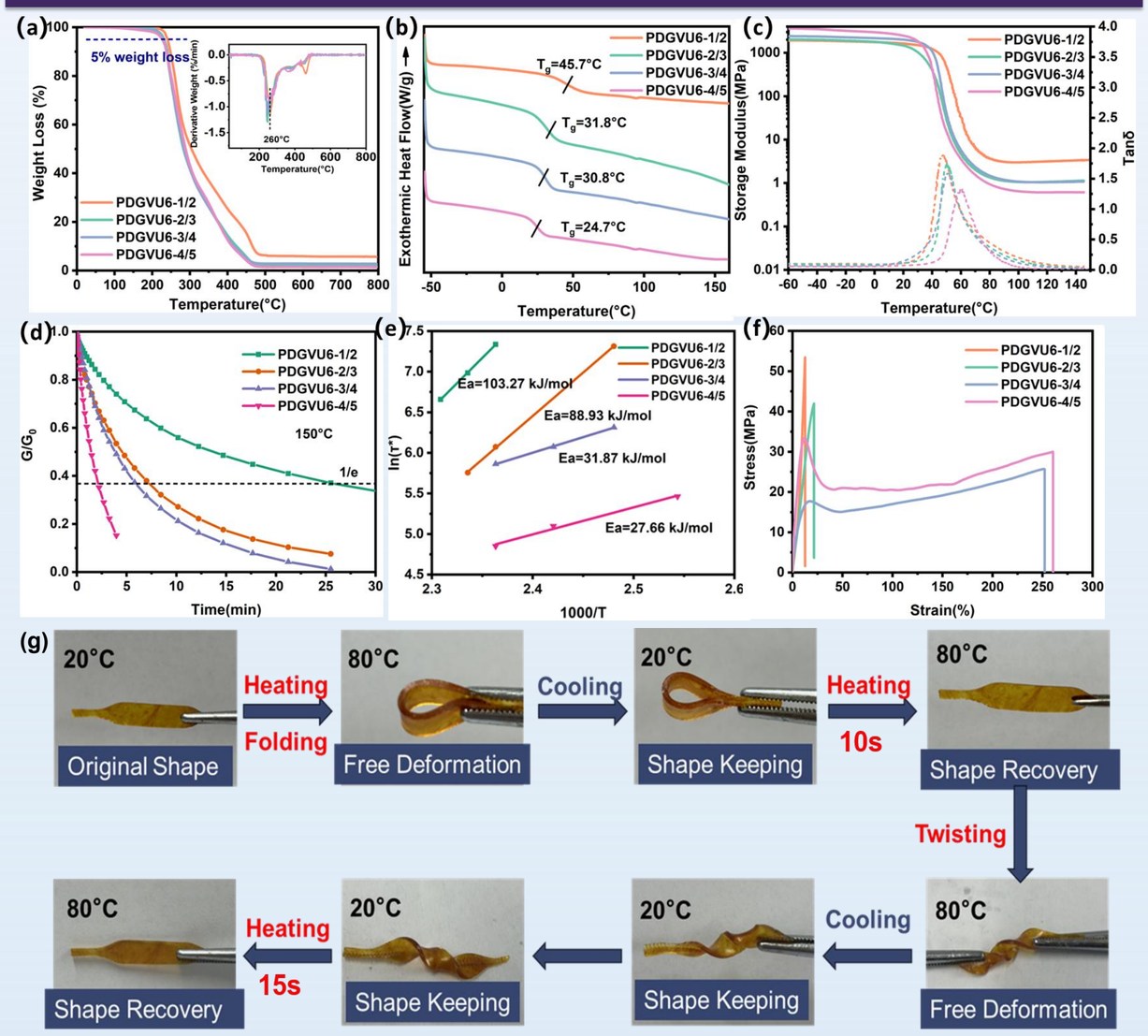


Figure 1 (a) TGA, (b) Second heating DSC, (c) DMA, (d) Stress at 150° C, (e) Fitting results of Arrhenius equation obtained from the stress relaxation and (f) Stress-strain curves of PDGVU6s.

CONCLUSION

- In this work, we have reported a green route to synthesize a bio-based NIPU using diglycerol as a nontoxic and renewable feedstock, achieving 100% bio-based content in the final product.
- The obtained PDGVU6s with dual dynamic covalent bonds exhibited relatively low activation energy (27.66 kJ/mol).
- The introduction of VU linkages endowed PDGVU6s with outstanding tensile strength (up to 53.4 MPa) and high reprocessing efficiency (~100%, 30 min at 150 ° C), successfully addressing the trade-off between mechanical performance and reprocessability while enhancing sustainability.
- PDGVU6s possessed excellent self-healing ability (95% repair efficiency after 24 h at 70 ° C), shape memory property and chemical degradable properties.
- This work established a green and sustainable synthetic strategy for novel NIPUs with VU bonds using bio-based feedstocks.

FUTURE WORK / REFERENCES

References

[1] Zhao X.-L., Tian P.-X., Li Y.-D., Zeng J.-B., 2022. Biobased covalent adaptable networks: towards better sustainability of thermosets. *Green Chem.* 24, 4363–4387.

[2] Van Lijsebetten F., De Bruycker K., Spiesschaert Y., Winne J. M., Du Prez F. E., 2022. Suppressing Creep and Promoting Fast Reprocessing of Vitrimers with Reversibly Trapped Amines. *Angew. Chem. Int. Ed*. 61, e202113872.

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