Lignin based materials with intrinsic recyclability

Summary

Addressing the urgent challenges posed by global warming, this study explores the design and application of lignin-based covalent adaptable networks (CANs) as a promising alternative to conventional petrochemical materials.

Traditional thermosetting materials, characterized by their irreversible crosslinked structures, are non-recyclable and often end up in landfills or are incinerated, exacerbating environmental pollution. To combat this, we propose preparing materials based on lignin, a natural biomass polymer, as an alternative. By incorporating CANs, these lignin-based materials offer both recyclability and high-performance characteristics, aligning with the growing demand for renewability and sustainability. Three chemical strategies were employed to synthesize lignin-based CANs: vinylogous urethane, imine chemistry, and transesterification. Results demonstrate that adjusting the ratio of lignin to biobased fatty acid derivatives enables tuning of the materials' mechanical properties. The vinylogous urethane network was formed under catalyst-free conditions by crosslinking lignin with acetoacetate and fatty amines, producing CANs with effective stress relaxation properties. Lignin based CANs synthesized via imine chemistry exhibited the lowest dynamic exchange activation energy, offering excellent self-healing and recyclability. Meanwhile, CANs formed by transesterification displayed significantly enhanced mechanical strength due to the formation of irreversible covalent bonds. Furthermore, a dual-network system was developed to further enhance the properties. By incorporating hydrogen bonding and semi-crystalline polymers, the system improved the creep resistance at high temperatures and storage modulus at room temperature. The potential applications of lignin-based CANs were also explored. These materials were found to be highly suitable for use as on-demand adhesives, self-healing coatings, photothermal conversion materials, and shape memory materials.

Lignin-based CANs provide a green alternative to petrochemical thermosetting materials and show broad application prospects. These findings pave the way for the development of sustainable and environmental-friendly materials toward circular economy.