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A NEW ERA IN OLEFIN METATHESIS: CATALYSIS TOWARDS A SUSTAINABLE FUTURE

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Growing concerns surrounding plastic waste management necessitate the development of diverse strategies and technologies to support the transition toward sustainable, circular materials. One particularly promising approach to addressing the persistent challenge of fossil-based plastic waste - such as polyethylene - is the design of innovative catalytic processes that upcycle waste into more sustainable, value-added products. Among these, olefin metathesis catalytic systems have emerged as powerful tools for transforming durable polyethylene waste into valuable chemical intermediates, which can be further utilized to produce high-value materials, including chemically recyclable and biodegradable plastics.

This work focuses on the development and application of N-alkyl-substituted Cyclic(Alkyl)(Amino)Carbene (CAAC)-Ru catalysts with "inverted" chemical structures, as well as bicyclic (Alkyl)(Amino)Carbene (BICAAC)-Ru complexes, both of which demonstrate enhanced activity in isomerization metathesis (ISOMET) reactions. These catalysts are used in tandem with a ruthenium-based double bond isomerization co-catalyst, (RuHCl)(CO)(PPh₃)₃. The process begins with the slow pyrolysis of post-consumer polyolefin waste, generating pyrolysis oils enriched in olefins. In the subsequent ISOMET step, the tailored olefin metathesis catalysts are combined with the isomerization catalyst to selectively convert these olefins into propylene.

The resulting olefin metathesis/isomerization catalyst systems show remarkable efficiency in converting post-consumer plastic waste into propylene. Notably, successful scale-up to a 900 mL reactor system and repeated batch testing confirmed the long-term operational stability of the catalyst system.

The metathesis of sterically crowded alkenes remains a significant challenge due to reduced catalyst accessibility and reactivity at the hindered double bonds. These substrates often exhibit sluggish reaction rates, poor conversions, or lead to catalyst deactivation. Overcoming this limitation requires the development of more robust and selective catalyst systems capable of tolerating steric bulk while maintaining high activity and functional group compatibility. Advances in ligand design - especially the development of novel bicyclic(alkyl)(amino)carbene (BICAAC)-based catalysts - offer promising solutions to the challenge of metathesis involving branched polymers, paving the way for broader applications in polymer chemical recycling. This talk will highlight recent progress and remaining challenges in olefin metathesis-based approaches to polymer recycling.