

**Wolfson Department of Chemical Engineering Seminar****Wednesday, April 29th, 2026 at 13:00**<https://gtiit.zoom.us/j/95891622686>**Catalytic Decomposition of Polyolefin Plastics via Ex Situ and In Situ Approaches for High-Value Products****Sijie Li****PhD Seminar**Advisor: Prof. Moshe Sheintuch, Prof. Xi Gao
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Plastic depolymerization usually relies on harsh conditions, including high temperatures, elevated pressures, strong acids or bases, and external hydrogen, resulting in high energy consumption and environmental concerns. Here, we show that tuning catalytic active-site environments enables selective upcycling of polyolefin plastics into either hydrogen or jet-fuel-range hydrocarbons. For hydrogen production, a grain-boundary-rich $\text{Fe}_2\text{O}_3/\text{Al}_2\text{O}_3$ catalyst outperformed Fe_2O_3 , Al_2O_3 , and their cascade combination, delivering $50.53 \text{ mmol g}^{-1}$ plastic H_2 from HDPE, corresponding to more than 70% recovery of the hydrogen in the plastic. Characterization indicates that the grain boundary enhanced the adsorption of gaseous intermediates and promoted in situ formation of FeAl_2O_4 , which facilitated proton adsorption and C–H bond cleavage. Further improvement was achieved with FeAl_2O_4 @biochar containing tetrahedral Fe sites, which produced $57.98 \pm 1.96 \text{ mmol g}^{-1}$ plastic H_2 from practical PP mask waste, equivalent to 86.25 H% in PP. DFT calculations show that tetrahedral Fe is more favorable for C–H activation and that dehydrogenation is kinetically more facile than C–C bond cleavage, making it the main source of H_2 . For liquid-fuel production, a sequentially modified HY-SMP zeolite developed enlarged supermicropores and more accessible Brønsted acid sites. Under solvent- and hydrogen-free ambient-pressure conditions at 200–240 °C, jet-fuel-range hydrocarbons yield reached 77.39% for PP, 80.92% for LDPE, and 81.10% for HDPE. DFT further reveals that the $\text{C}_5 + \text{C}_7$ route is favored by the lowest β -scission barrier owing to optimal stabilization of a secondary carbenium-ion-like transition state. Overall, this work establishes a structure–reactivity framework for directing polyolefin upcycling toward either high-yield hydrogen or valuable liquid fuels.