The key role of reaction engineering in catalytic recycling of HDPE and PP

 $\underline{\mathsf{S}}$. Jaydev 1 , K. Chikri 1 , M. Usteri 1 , A. J. Martín 1 , G. Pagani 1 , J. Pérez-Ramírez 1*

¹ETH Zurich

High-density polyethylene (HDPE) and polypropylene (PP) account for nearly 50% of all plastic waste produced. Life cycle assessments point at their catalytic hydro-conversion into light alkanes as a potential long-term environmentally and economically viable solution.^[1] Investigations have largely focused on catalyst development, despite the presumably large relevance of reaction engineering due to the distinctive nature of plastic as reactants. As non-Newtonian fluids when melted, their highly viscous and shear-thinning nature (**Fig 1a.**) may dictate mass transport properties not properly described by classical theories for internal and external mass transport limitations, underscoring the role of rheology to assess catalyst-feedstock contact^[2]. Systematic studies over a reference Ru/TiO₂ catalyst revealed and rationalized distinctive responses to fluid velocity, flow pattern, and catalyst sieve sizes. Distinctive flow patterns imposed by stirrer geometry are described by cyclic motion of the feedstock in axial flow, bringing about greater catalyst-feedstock contact, leading to higher conversion, whereas sideways motion in radial flow leads to the catalysts being settled at the bottom (**Fig. 1b**). Multiphase CFD simulations and highspeed imaging revealed that the lower viscosity of PP under operating shear (**Fig. 1a**) favour a more homogeneous distribution of catalyst particles in the reactor, in contrast to the more viscous HDPE, for which catalyst sieve size becomes key (**Fig. 1c**). The aforementioned observations indicate that reaction engineering is thus of utmost importance for the evaluation of catalytic performance. Consequently, this study also identifies operating conditions characterized by nondimensional numbers for minimizing the influence of viscosity of HDPE and PP, under which catalyst are recommended to be evaluated (torque > 10 N.cm with axial flow, $T > 473$ K).

Fig. 1a Chemical structure and viscosity (under changing torque) of the two feedstocks (PP, $MFI = 4$, HDPE, MFI = 2.2). **b** Yields of different fractions for PP and HDPE under varying mixing patterns **c** Yields of different fractions for PP and HDPE under varying sieve size. Reaction conditions: $T = 498$ K, $P = 20$ bar H₂, $t = 4$ h and catalyst/plastic = 5 wt.%

[1] A.J. Martín, C. Mondelli, S. D. Jaydev, J. Pérez-Ramírez, *Chem* **2021**, *7*, 1487-1533. [2] J. Ge, B. Peters, *Chem. Eng. J.* **2023**, *466*, 143251.