TWO-STAGE HYDROTHERMAL LIQUEFACTION FOR MULTILAYER PLASTIC VALORIZATION

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The production of multilayer plastic films is high due to the coexistence of their mechanical and physical properties with moisture and oxygen barrier behavior that make them optimal for packaging. However, these plastic films cannot be mechanically recycled due to their multiplastic nature, nor can be valorized by pyrolysis due to the high amount of PET that cannot be tollerated. Hydrothermal liquefaction (HTL) under subcritical conditions could be useful for hydrolyzing polycondensation polymers, such as PET, the second most common polymer in multifilms, while leaving polyolefins, such as PE, the most common polymer in multifilms, intact [1]. The residual polyolefins could then be converted into liquid fuel through a consecutive supercritical HTL. In this work, the possibility to use hydrothermal liquefaction (HTL) in a two-stage process was investigated with a case study using a two-laver film made of LLDPE-PET. The first step was performed at 325 °C for 20 min and 20 wt.% of solid loading. Terephtalic acid (TPA), one of two monomers of PET, was recovered 94% as solid after alkaline extraction, while 47% of the other PET monomer, ethylene glycol (EG), was recovered in the aqueous phase. The solid residue was 98% of the LLDPE present in the starting multilayer, with a purity of 98%. The PE-rich phase was then used as feedstock for a second step in supercritical conditions. A screening for the optimal conditions of residence time and temperature was performed. The highest oil production was registered at 450 °C, 90 min and 33 wt.% solid loading. The oil yield was 47% while the gas yield was 29%. From elemental analysis, the oil formula was CH1.7 with no residual oxygen; the main compounds identified by GC-MS were linear alkanes and the boiling point (TGA) was in the naphta-gasoline range. The gas-phase consisted mainly of linear saturated hydrocarbons (CH₄ 31vol.%, C₂H₆ 27vol.%, C₃H₈ 16vol.%) with good heating value



Figure **1**– Distribution of products after subcritical step (325 °C, 20 min) and supercritical step of the PE-rich phase (450 °C, 90 min).

very high quality oil for transportation sector, respectively.

References

[1] J.S. dos Passos, M. Glasius, P. Biller, Screening of common synthetic polymers for depolymerization by subcritical hydrothermal liquefaction, Process Saf. Environ. Prot. 139 (2020) 371–379. https://doi.org/10.1016/j.psep.2020.04.040.

(LHV = 46 MJ/kg), which make the gas suitable for meeting the thermal demand of the plant or for use as steam cracking feed. The overall mass distributions are shown in Figure 1.

In conclusion, hydrothermal liquefaction has proven useful for chemical recycling of multilayer plastic films through a two-stage reaction. The two-stage configuration allows PET monomers to be recovered in good vields the monomers of PET at subcritical conditions while limiting their further degradation, making them suitable for recycling in the PET industry. The residual PE can then be valorized under supercritical conditions by producing gaseous and liquid hydrocarbons that can be used as feed for olefin production and as a