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**HYBRID TANDEM CATALYTIC CONVERSION PROCESS
TOWARDS HIGHER OXYGENATE EFUELS**



E-TANDEM - Deliverable report

D 4.1 – Recycling of hybrid catalyst system

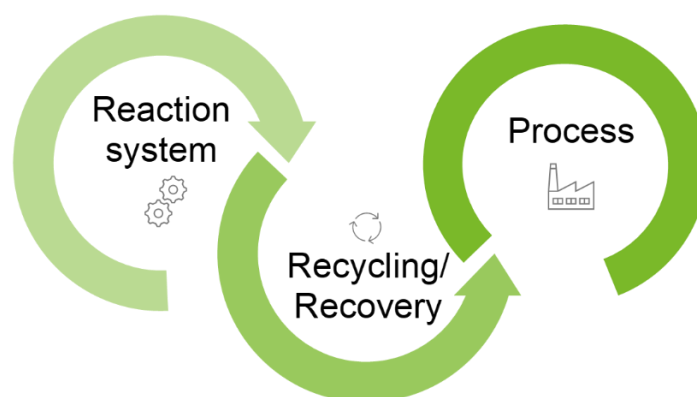


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Publishable summary

Carbon-neutral, high-energy density e-fuels are crucial to de-fossilize long-haul, heavy-duty road, marine, and aviation transport sectors. The Horizon Europe E-TANDEM project will develop and validate, at bench scale, the first direct process for the selective production of carbon-neutral higher oxygenate e-fuels (HOEF) for the marine and heavy-duty transport sectors. This oxygenated fuel is directly produced from CO₂ as the sole carbon source, and renewable power as the sole energy input, in a once-through hybrid catalytic conversion process integrating three major catalysis branches: high-pressure electrocatalysis syngas production coupled to a tandem catalytic e-syngas conversion encompassing thermocatalysis with solid catalysts and chemocatalysis with molecular complexes. The project will demonstrate the new e-fuel production process at bench-scale and assess its capacity to cope with fluctuating energy inputs.

Work package 4 experimentally validates the new e-fuel production concept, in a continuous mode, at bench-scale in a custom made miniplant. Moreover, it models, simulates and optimizes the operation of an engineering demonstration plant with 1 MW capacity in silico. Essential for the experimental validation of the e-fuel production process, as well as a critical design input for process simulation, is to develop a strategy to effectively retain the two different types of metal-based catalysts which cooperate in the hybrid, tandem syngas conversion process step, ensuring no metal losses or product contamination.



This deliverable report 4.1 describes the work done to experimentally identify an optimal concept for recycling of the hybrid catalyst system for the CO/H₂ conversion. A solid Fischer-Tropsch catalyst and a molecular reductive hydroformylation catalyst are combined in the tandem reaction, which proceeds in a slurry-phase stirred-tank reactor. Therefore, two different recycling concepts for both catalysts were developed. The solid Fischer-Tropsch catalyst shall remain in the reactor at all times during an experiment to keep its activity and preventing from the clogging of flow lines in the miniplant, downstream from the reactor. For the molecular catalyst, different recycling strategies like multiphase approaches, immobilization, distillation and organic solvent nanofiltration (OSN) are discussed and evaluated in terms of their applicability for catalyst recycling in the tandem reaction. The most promising approach is found to be the use of membranes in an organic solvent nanofiltration (OSN) process. Compared to alternative methods like distillation or separations in biphasic media, OSN shows a high energy efficiency, is particularly amenable to continuous process operation and proceeds at mild temperatures, preventing thermal decomposition of the organometallic complex molecular catalyst. For the selection of a suitable membrane candidate a computational and an experimental approach are adopted, leading to the identification of a polymeric membrane which exhibits cobalt rejection of 99.7 %.



The report demonstrates that the process can be operated with a very low loss of catalyst metal ($<0.1\%$) over 100 hours, as a combined cobalt loss of 0.08% from both the heterogeneous and homogeneous catalysts has been calculated.

Furthermore, the recovery of the product, especially the Fischer-Tropsch by-product water is discussed. A decanter is used to separate the organic product phase and the aqueous phase that will be fed back as a reactant for the electrolysis after a conditioning step. This report summarises the downstream process for the E-TANDEM project, making it possible to proceed with the next step, the demonstration of the process at bench-scale.