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New process for continuous recycling of a catalyst supported by magnetic particles

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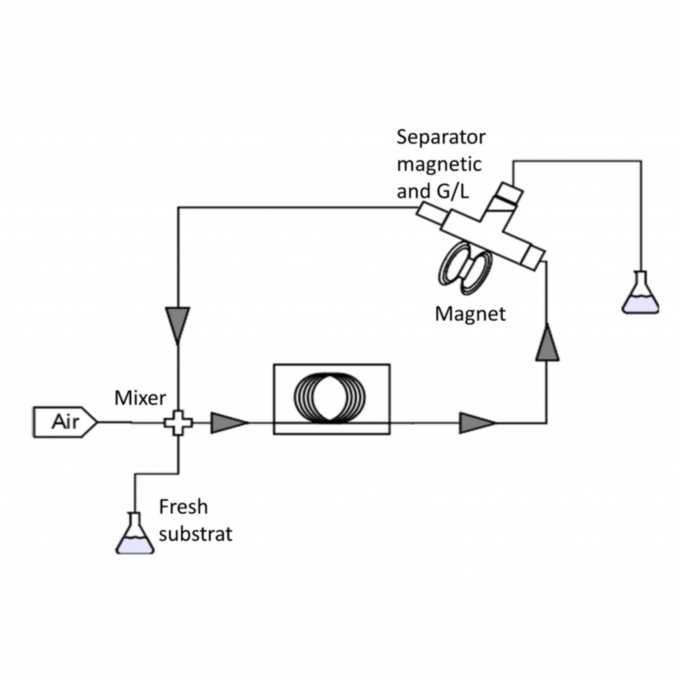
PURPOSE OF THE ABSTRACT

The NANOTRAP project proposes to design a new continuous process to obtain a permanent recycling of a catalyst based on metallic nanoparticles (gold or palladium) supported by nano or micrometric magnetic particles1. The objective is to operate a "quasi-homogeneous" catalysis chemistry with a catalyst that can not be separated by usual techniques (filtration), for example to avoid intragranular diffusional limitations observed with classical heterogeneous catalysts. The separation is then based on magnetic deviation approach using magnetic nano- or microparticles as can been seen in many studies; but the main difference in our paper is to develop the separation and the recycling of the particles in the process; while usually the separation is only showed with the interaction of a single magnet. The challenge is then to develop the separation in continuous to allow the magnetic particle to be trapped in a closed loop while the products and reactants can go in and out of the process in presence of a gas and liquid. Final goal would be to achieve a "proof-of-concept" in the presence of a multiphase reaction in a first part with model reaction (single phase in liquid or two phases at moderate temperature) and ultimately to apply the concept for catalytic oxidative depolymerization of lignins using Pd-based nanocatalysts2.

The principle of the experiment is described in the Figure 1. First, the nano/micro particles are homogenized in a reservoir by agitation. Then, the liquid is co-injected with a gas flow in a continuous tubular reactor with a diameter of 1 mm (gas/liquid system) over 2 m long. At the output of the reactor, a separator is set up to reach a 90% recycling rate on the liquid while gas is separated from the rest of the reaction. The goal is not to lose any MNP that could be carried away by gas and liquid flow. For better control of the exit flow, a T-separator was designed with an entrance path, a recycle path and an exit path. Separator's first function is to maintain MNPs in the recycle loop by deflecting their course with magnet. Its second function is to separate the gas from portion of liquid by gravity. The positions of the separator and the magnets inside the process have been optimized by an incremental "trial and error" approach as the first attempts of scaling up were quickly shown to be highly limited.

We tried several positions of the T-cross for microparticles in liquid/solid phase. Thanks to the optimization of the separator, we have succeeded in obtaining losses of MNPs limited to less than 1% by mass. We applied a down-flow because the microparticles are particularly sensitive to the gravity. But this down-flow was not appropriate in a liquid/solid/gas phase. In fact, there is a formation of a stagnant gas pocket in the separator which destabilize the flow and therefore the recyclage of MPs. We decided to use nano-sized particles, which are less sensitive to sedimentation, and by applying an up-flow, we were able to minimize particle losses and optimize gas evacuation by avoiding the formation of a stagnant gas pocket. We obtain less than 2% of losses of MNPs for 2 hours of operating. The proof of concept of magnetic recycling still needs to be validated by a model reaction which is the hydrogenation of 4-nitrophenol catalyzed by gold particles supported on MNPs3.

FIGURES



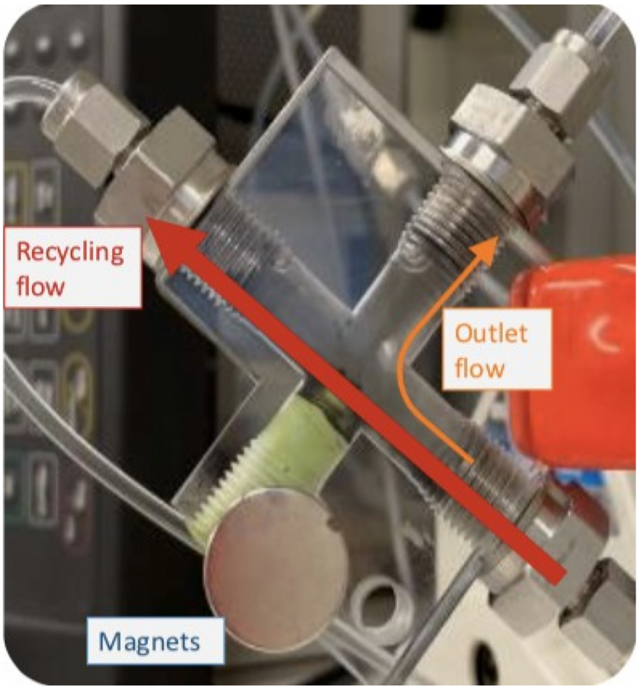


FIGURE 1

Principle of the innovative continuous process for recycling catalysts supported on magnetic particles

The recycling loop, the T-cross separator, the magnet and liquid and gaz flows are represented on scheme

FIGURE 2

Picture of T-cross separator with gas/liquid/solid.

Various path and flow are shown on the picture

KEYWORDS

Recycling procees | Magneting particles | Magnetic recycling

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