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Intensification principle of a new three-phase catalytic slurry reactor. Part II: Eco-efficiency and techno-economic performances

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ABSTRACT

In two papers, the concept and the performances of a new continuous intensified reactor named RAPTOR® (French acronym for Reactor with Polyvalent Rectilinear Stirred Reactor with Optimised Transfer) are presented.

Based on flow, heat and mass transfer characterisations and real hydrogenation experiments, Paper I presented a simple analytical model based on characteristic times that enables to explain the intensified performances compared with a semi-batch stirred reactor and to generalise the operability, rapidity and the flexibility of this minireactor. In Paper II (this article), the model is used to evaluate in a comparative study the eco-efficiency and the techno-economical advantages of a continuous process involving a RAPTOR® versus a classical batch process equipped with a stirred reactor. Economical, environmental aspects are considered as well as productivity, safety and process control.

1. Introduction

This is the second part of a two-part article on an intensified reactor designed for multiphase reactions. This new reactor called RAPTOR® (French acronym for "Réacteur Agité Polyvalent à Transfert Optimisé Rectiligne" or "Polyvalent Rectilinear Stirred Reactor With Optimised Transfer") is analysed as catalytic three phase slurry. A previous paper in this journal [1], described the reactor performance, i.e. typical application examples, mass transfer characteristics, flow patterns and a reaction regime analysis. Furthermore, a simplified reactor model based on characteristic times has been presented which served to explain experimental performance. Part II (this paper) deals with economic and environmental efficiency as well as techno-economical aspects including productivity, process safety and control. We decided to also focus on these practical or commercial rather than scientific aspects as they typically represent important barriers to process intensification and explain why very few intensified reactors have actually made it to commercial scale [1,2].

So far, very few articles have been published which approach intensified reactors from the techno-economic or environmental angle. Some papers treat this subject on a rather general basis, not going into detail [3]. Other studies, more detailed, focus on homogeneous reactions in microreactors [4–8]. The reason why only few articles of this kind exist is mainly the fact that the majority of intensified reactor concepts are unrealistic on commercial scale for practical and economic reasons. The high numbering-up requirement due to limited productivity often leads to high CAPEX with long payback periods. Reactor plugging, flow distribution, flexibility and process control can represent practical obstacles. Finally lack of experience regarding reactor design, -operation and -safety for intensified conditions slow down the changeover from batch to intensified continuous processing [1].

For the purpose of this work we would again like to focus on the exemplary process of hydrogenation of ortho-cresol. Operation conditions for this metal-catalysed, gas–liquid reaction are summarised in Table 1. Herein the observed process parameters for continuous production are compared to those of a theoretical batch process of similar productivity.

These quite impressive but partly unexplained observations prompted a reactor characterisation on a more general basis. In this context the following questions were and will be addressed:

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Table 1
Reactor performances for the catalysed hydrogenation of ortho-cresol [1].

	Batch Reactor	Raptor minireactor
Volume	6 m^3	<0.001 m^3
Productivity	300 t/year	300 t/year
Pressure	15 bar	200 bar
Temperature	100 °C	170 °C
Dilution (solvent fraction)	75%	None
Operation time	4 h	3 min
Catalyst concentration	4%	0.4%
Conversion	95%	>99.9%
Adiabatic temperature rise	100 °C	925

Part I [1]:
- Why can fast and strongly exothermic reactions be carried out without dilution?
- How can total conversion be attained despite agitation/gas-flow (backmixing)?
- Why is the reactor faster despite the small catalyst concentration?

Part II (this paper):
- Are small-sized reactors "inherently" safe despite intensified operation conditions?
- How can control obstacles associated with intensified operation be handled?
- How can large scale batch productivities be attained?
- Finally, is the eco-efficiency driver strong enough for the changeover from batch to continuous?

2. Safety and control

A model assisted HAZOP safety study has shown that the Raptor can be considered as safer when compared to corresponding large scale batch reactors. However, inherent safety, an attribute often assigned to small reactors, especially microreactors, should be considered with care: potential hazards may be much more frequent due to the intensified conditions. Consequences, however, are much less severe. Furthermore the fact that the reactor's inert mass (material of construction) is large when compared to the processed material delays thermal runaway and provides more time for countermeasures. Finally, small size offers additional measures of intervention like instant drainage, quenching or blockhouse installation. Hence, rather aspects indirectly related to the reactor size allow to conclude that the Raptor is safer. For further details we would like to refer to the corresponding publications [8,9].

In yet another work recently published a new process control strategy is presented which is adapted to the special characteristics of intensified mini-reactors and has been studied for the Raptor as test example. An efficient hierarchical control structure was applied which responds rapidly to set point changes in the intensified minireactor and which uses simple to measure input variables. When compared to classical PID-controllers the control characteristics could be greatly improved [10].

3. Productivity

Intensification of heat and mass transfers is very often obtained in small reactors which presents high surface to volume ratios. As the net productivity is proportional to the ratio of the reactor volume to the operation time required to complete the reaction, it is often objected that this miniaturisation is detrimental to minireactor's productivity. To compete with conventional large reactors, operation time or residence time in minireactors needs to be very small [11].

3.1. Theoretical analysis neglecting changeover times

Let us first consider the idealised case of a batch process without changeover times. The productivity of the semi-batch reactor for ideal operation, i.e. negligible changeover-times can be expressed in as follows:

$$P_{B,ideal} = \frac{V_L C_{A0} X_A}{t_{OP}} = \frac{V_L p}{\nu He t_{MT}} \tag{1}$$

Based on the simplified model presented in part I [1], the following equation is derived for the productivity of the continuous reactor (considering (19) in [1] and $\tau_l = V_L/q$):

$$P_C = q_L C_{A0} X_A = q_L C_{A0} \frac{\tau_L}{t_{MT}} \xi = \frac{V_L p}{\nu He t_{MT}} \tag{2}$$

Hence, for mass transfer control, the productivity of continuous and batch reactor are the same and directly related to the operating pressure of hydrogen, the volume of the reactor and the mass transfer characteristic time but independent from initial concentration of the liquid reagent or the volumetric throughput q_L.

So far we considered the reactor dimensions to be fixed. The following study investigates how scale-up of the reactor dimensions affect dilution and productivity. Eqs. (2) and (1) consider that the productivity is directly proportional to the reactor volume, which in turns is a function of the vessel diameter D. However, since the surface to volume ratio decreases, more dilution is required in order to meet the safety constraints which is counter productive with respect to productivity. In order to determine the maximum vessel diameter for a certain safety constraint, e.g. $\Delta T/(\Delta T_{ad})_{pure} < 10\%$ (Eq. (18), [1]) marks the starting point for the evaluation. In this equation the characteristic time of heat transfer is the only property that depends on the vessel diameter. The surface to volume ratio of typical agitated vessels can be expressed as $A/V_L = 5.3D$ (cylindrical vessel with $L = D$) which turns relation (11, [1]) into:

$$t_{HT} = \frac{\rho c_p V_L}{UA} = \underbrace{\frac{\rho c_p}{5.3\,U}}_{R_B} D \tag{3}$$

Introducing (3) in (18, [1]) leads to

$$\frac{\Delta T}{(\Delta T_{ad})_{pure}} = \xi_{pure} \left(\frac{R_B}{t_{MT}}\right) D \left(1 - \exp\left\{-\frac{t_{MT}}{R_B} \frac{X_A}{\xi_{pure} F} \frac{1}{D}\right\}\right) \tag{4}$$

which is implicit with respect to D and must be solved iteratively. However, due to the similar behaviour, relation (21, [1]) may be used as first approximate instead of (18, [1]), which leads to the explicit expression (7). The productivity would then be calculated from (1) and (7) for a cylindrical vessel ($V_L = \varepsilon_L D^3(\pi/4)$):

$$P_{B,ideal} \approx \frac{\pi}{4} \varepsilon_L \frac{1}{R_B^3} \left[\left(\frac{\Delta T}{(\Delta T_{ad})_{pure}}\right)^{-1} - F\right]^{-3} \left(\frac{C_{A0}}{\xi^2}\right)_{pure} t_{MT}^2 \tag{5}$$

For the continuous reactor (21, [1]) a cylindrical shape is assumed ($A/V_L = 4/D$) which leads to

$$t_{HT} = \frac{\rho c_p V_L}{UA} = \underbrace{\frac{\rho c_p V_L}{4U}}_{R_C} D \tag{6}$$

Introducing (6) in (21, [1]) gives

$$D = \frac{t_{MT}}{\xi_{pure} R_C} \left[\left(\frac{\Delta T}{(\Delta T_{ad})_{pure}}\right)^{-1} - F\right]^{-1} \tag{7}$$