RAPIC PROJECT: TOWARD A NEW GENERATION OF INEXPENSIVE HEAT EXCHANGER-REACTORS FOR PROCESS INTENSIFICATION

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Abstract.

Process intensification (PI) in chemical production is a major concern of chemical manufacturers. This alternative technology involves transposing syntheses into continuous plug flow reactors with process intensification, leading to a multifunctional heat exchanger-reactor. In this context, the RAPIC R&D project aims to develop an innovative *low-cost* component (in the 10 kg/hour range). This project deals with the design from the local to the global scale and with testing, from elementary mock-ups to pilot scale. The present paper gives a detailed description of this research project and presents the main results on specification and definition of the reaction channel and the first simple mock-ups.

Key-words. RAPIC project; Low price, Process Intensification, Multifunctional Heat Exchanger-Reactor

INTRODUCTION

Process intensification (PI) methodology, introduced in the 1980s, was recently defined¹ as "any chemical engineering development that leads to a substantially smaller, safer, cleaner and more energy-efficient technology". As distinguished from traditional batch processes, PI consists in transposing syntheses into continuous plug flow reactors with process intensification, leading to a multifunctional heat exchanger–reactor². Many benefits are expected such as waste reduction, energy and raw materials savings, yield and selectivity increase, and cost reduction. Despite these benefits, to date only a few intensified processes are in industrial operation. Nevertheless, typical academic studies⁻⁶ of multifunctional heat exchanger–reactors in the literature report the large potential of this technology. The relatively slow transfer to industry is presumably linked to many factors, among them the lack of successful industrial demonstrations.

The RAPIC project focuses on the implementation in industry of intensified continuous unit and aims to be polyvalent and flexible (batch processes as well), robust (not too sensitive to varying conditions) and low-cost (crucial for process industrialization). The present paper details this research project and presents the main results on specification and definition of the reaction channel and the first elementary mock-ups.

RAPIC PROJECT DESCRIPTION

Main objectives

The RAPIC R&D project, started in December 2007 and funded for three years by the French National Research Agency (ANR), aims to develop an innovative but low-cost component in the 10 kg/hour range. It is original in that it must not only comply with the implementation constraints of exothermal reactions, but also be as close as possible to maturing technologies in the heat-exchanger sector by adapting them to reactive media and by respecting the cost imperatives imposed by the market. In principle, the plate reactor is composed of alternate (sandwich) coolant and reaction circuits (*figure 1*). Circuits are brazed in order to ensure good thermal contact. Thus, using enhanced plate-fin geometry available on the market, the RAPIC plate reactor has very high heat-transfer ability to ensure excellent thermal control of the reaction.



Figure 1. Schematic of RAPIC plate reactor.

We plan to use hot isostatic pressing (HIP) fabrication technology for the reaction channels so that we can produce modules with different materials (copper, stainless steel) placed around the reaction channels for efficient management of exothermal effects⁷. The complex geometries needed for process intensification are obtained by assembling machined metallic plates or bent tubes embedded in sintered metal powders. Large industrial HIP facilities available today will produce low-cost components with enhanced thermal performance.

Project organization

Over its three years, the RAPIC project will deal with design from the local to the global scale and experimentation from elementary mock-ups to pilot-scale systems. Five main tasks are involved:

- 1. Specification of the plate reactor by the end user in terms of performance level and acceptable price $(T_0 \Rightarrow T_0+3)$
- 2. Definition of the reaction channel, based on numerical simulation and analytical tests ($T_0+3 \Rightarrow T_0+18$)
- 3. Manufacture (by traditional or innovative techniques) and testing (thermal hydraulic and micromixing) of plate modules designed for reaction $(T_0+3 \Rightarrow T_0+18)$
- 4. Manufacture and test of two prototypes (1-10 L/h); several representative exothermal reactions will be tested ($T_0+18 \Rightarrow T_0+36$)
- 5. Market assessment and dissemination $(T_0+33 \Rightarrow T_0+36)$

Since the present paper summarizes work in the first project year, only tasks 1, 2 and 3 are detailed.

Partnerships

The teams in the RAPIC project are highly complementary (*figure 2*). Indeed, one end-user (Rhodia Chemicals) and one heat-exchanger manufacturer (Fives Cryo) are involved to ensure the industrial orientation of the work. In addition, French laboratories active in basic research on process

engineering (LGC) and thermal-hydraulic engineering (LTN) are involved. The Atomic Energy Commission (CEA/LITEN) is handling project coordination, component design, and manufacture of the reaction plates using HIP technology.



The involvement in RAPIC of a leading chemical group (Rhodia) as well as a parts manufacturer (Fives Cryo) desiring to diversify its industrial activities constitutes a driving force and ensures the future industrial application of the work.

SPECIFICATIONS

The specification, in terms of the chemical reaction target, has been defined by Rhodia as an exothermal reaction, with an adiabatic temperature rise of 200°C, under atmospheric pressure and temperature equal to 100°C. This is a fast reaction, with a residence time of 2-3 minutes. The reactor must be as isothermal as possible, with a maximum temperature raise of 10°C. *Figure 3* gives the required reactor volume as a function of volumetric heat-transfer performance and coolant temperature. (The chosen coolant throughput allows overcoming any heat exchange limitation in the "coolant side").



Figure 3. Specified operating conditions.

Three main sizes have been defined as production targets for the project:

- Elementary scale: 1 l/h
- Laboratory scale: 10 l/h
- Pilot scale : from hundreds of l/h up to few cubic meters/h

Only laboratory-scale units will be constructed and tested within the RAPIC project. However, extrapolation to the pilot scale is included in the choice of concept and geometry.

A detailed economic analysis made by Rhodia for industrial chemical production of 6 kTs/year is based on a methodology for transferring batch to continuous reaction. For this estimate, the reactor investment assumes a price equal to that of semi-batch technology: about 300 k \in for 27 m³ semi-batch reactor. In addition, the analysis also includes modification of the process environment, such as upstream and downstream storage tanks (larger for batch process), injection simplification, and data measurement and acquisition systems. Using these assumptions, the overall cost of batch production for 6kT/y is estimated as 4.06 M \in . For continuous processes, in order to achieve the same investment cost, the acceptable price level for the RAPIC plate reactor is about 800 k \in .

In term of manufacturing cost, we assume in a first time that the transposition from a batch process to a continuous process does not allow saving money: to meet the economical stakes, the investment cost is the key economical parameter to manage.

FIRST PLATE REACTOR CONCEPT

Simplified Modeling

Using simplified thermal-hydraulic modeling, a pre-sizing of a first plate reactor based on the specification has been made. This sizing is based on:

- The transference of CEA's know-how in nuclear fusion activity⁷ to the reaction part: integrating straight stainless steel tubes (8/10 mm) into a thermal conducting copper matrix (*figure 4*). The elementary plate size was determined by the availability of HIP vessels: 1.25 m long, 0.8 m wide.
- The use of standard straight and perforated fins (4 mm high, 0.15 mm thick) on the utility side in order to improve reactor coolant capacity.



Figure 4. Schematic of the first design of RAPIC reaction plate (gray – stainless steel; orange – copper).

Thus, in order to reach the predefined specifications, the full-size plate reactor has the following characteristics:

- 25 reaction plates (2 passes) in parallel, hence 50 plates surrounded by 51 utility plates with straight and perforated strip fins;
- Each plate includes 60 tubes (8/10 mm), with twisted tape inserts inside and external turning boxes;
- Plates are mainly copper, with stainless steel as the external skin (container). The final thickness of each reaction plate is around 20 mm.
- The final component has external dimensions 800 mm×1440 mm×1250 mm and volume 1.44 m³.

With this design, we reach a heat transfer capability of 580 kW/ m^3 .K and a pressure drop of 7 bars; the design thus meets both the required residence time and heat transfer goals. It is also a feasible technological solution.

Full prototype cost estimation

A precise cost estimate has been made for the above design

- Utility plate using offset strip fins : 165 k€
- Reaction plates: 580 k€ with 50% for materials procurement and machining

Therefore, this first design has an overall cost of 745 k€ for 6 kT/year production. This cost could be significantly reduced by enhanced design, which could lead to more compact geometries, and by manufacturing technology improvements. In any case, however, the RAPIC project has a realistic basis.

First elementary mock-ups

The objective of the first mock-up is to demonstrate that the design criteria selected for RAPIC lead to a low cost reaction plate by the choice of a mature manufacturing process and a simple geometry. The mock up size is around 320*130*25 mm, with two levels of tubes to increase compactness. The performance of the mock up will be measured in terms of heat transfer capacity, macro and micro mixing efficiency.

As shown in *figure 5*, the mock up is made of straight stainless steel tubes inserted in a copper matrix. Internal stainless steel swirls are placed into the tubes to enhance heat transfer and mixing. Tubes are placed between copper plates grooved by milling. The distribution and turning over of the fluid is ensured by two machined stainless steel turning boxes.

Once the tubes are welded to turning boxes and inserted between copper, all parts are placed into a metallic container that is sealed and degassed under vacuum. Hot Isostatic Pressing (typically under 1000°C/1000 bar/1hour) is then applied to weld all materials (tubes to distribution boxes, tubes to copper, copper to the container) by diffusion bonding. A final machining is performed on top and lower surfaces to ensure the proper flatness.



Figure 5 : Manufacturing steps for the first reaction plate mock up : a) tubes with internal swirls are placed between copper plates, b) turning and distribution boxes are welded to tube ends, c) mock up after HIP and final machining.

STRATEGY FOR REACTION CHANNEL OPTIMIZATION

So far, a feasible concept has been design and manufactured that meets all the predetermined specifications, including the price. However, this design is based on the use of straight tubes with inserts, which is not globally optimal. Thus design work continues in order to develop a more efficient, realizable and up-scalable geometry. Some orientations, based on enhanced design, have been considered:

- modification of channel cross-section shape (circular, square, rectangular)
- channels structural geometries (two-dimensional waves, three-dimensional geometries, ...)

Hence, the definition of an improved reaction plate pattern will be based on a systematic multiscale analysis, using analytical, experiments or simulation tools (*figure 6*). We should be able to define a second plate-reactor concept before the end of 2009.



Figure 6. Strategy for developing improved plate reactor geometry.

CONCLUSIONS AND FUTURE WORK

The RAPIC R&D project is at the end of its first year. The first reactor concept has been successfully designed and manufactured and will be tested in the beginning of 2009. This design is very simple and fits all the predetermined specifications. However, the potential for improvement is very high. Indeed, refining the design will impact the reactor cost significantly. The RAPIC project development strategy will follow two parallel routes:

- improving the design by using bends or inserts
- validating technological options that allow manufacture of low-cost components

At the end of the RAPIC project, two prototypes (10-50 l/h) will be available to test various chemical reactions and systems.

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