THE MAISON EUROPEENNE DES PROCEDES INNOVANTS (MEPI), AN EXAMPLE OF PILOTING AND INDUSTRIAL DEMONSTRATION FACILITY FOR THE GREEN PROCESS ENGINEERING

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Abstract. Economical, energy savings and environmental challenges require an actual technological breakthrough in process engineering, aiming with productivity, product quality, safety and reliability objectives. This explains the present growth of interest in innovative technologies (intensified devices for reaction, mixing and separation) and methods (multi-functionality, hybrid separation, batch to continuous methodology, new media …), the whole being recognized as Process Intensification. Up to now, a few of innovations has been successfully industrialized, probably due to the lack of experience and retrofitting in front of a breakthrough that always represents a technical and financial risk. There is now clearly a need for industrial demonstrations of successful PI experiments avoiding the questions of confidentiality. Consequently, a piloting and demonstration facility has been created in Toulouse in order to accelerate the implementation of PI technology in industry and the development of the Green Process Engineering. The idea is to build a data bank of success stories. The principle of this industrial technical platform lies on the association of 3 types of partners: university, equipment providers and industrial end-users.

Key-words. Process Intensification, Piloting and demonstration facility, Industrial technical platform,

WHY MEPI?

Process Intensification presents a set of often radically innovative principles in process and equipment design, which can bring significant benefits in terms of process and chain efficiency, capital and operating expenses, quality, wastes, process safety, and more[1-5]. As described by the Action Plan Process Intensification[6], introducing Process Intensification technology requires significant investment in an environment where payback times for investments need to be short. Large investments in current technology (including know-how) and competition against its reliability are limiting the introduction of Process Intensification. Managing the technical and financial risks requires developing new scale-up approaches and dedicated piloting facilities. Many barriers to implementation of Process Intensification start with the lack of a suitable piloting & demonstration facility. There are no piloting & demonstration facilities or possibilities to pilot on existing production lines. Furthermore, high (technical and financial) risks exist in the development of a first industrial prototype and in first implementation (retrofitting) of Process Intensification modules into existing production lines/plants. In the 1990s, many pilot facilities were closed down because process engineers believed new processes could be simulated with numerical modelling. However, engineers have discovered that piloting on a scale of 1:10 to 1:6 is indispensable to the demonstration of industrial and economical feasibility.

To overcome this major barrier, the need of facilities where promising Process Intensification technologies can be piloted and demonstrated on a semi-industrial scale clearly appears. To develop such an approach, the facility should offer a complete package of services (figure 1). Thus, feasibility and reliability of proposed Process Intensification processes will be confirmed at the facility on a semi-industrial scale, which will test the implementation of process changes, resolve process bottlenecks and manufacture products which can be tested in turn. The facility will also investigate scale-up processes and control, generating critical information about the behaviour of large-scale processes. And more important, integrated designs and layouts will be investigated and developed for use in the design, construction and operation of large-scale plants.

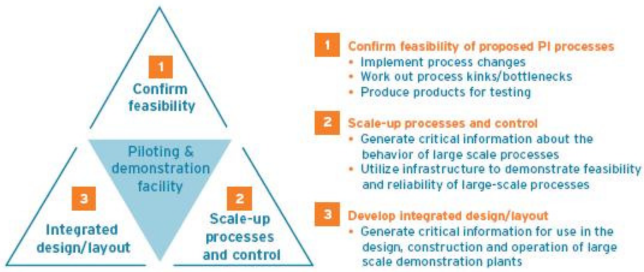

Figure 1: Objectives of piloting & Demonstration facility
Courtesy: Creative Energy, www.creative-energy.org

WHAT IS MEPI?

The "Maison Européenne des Procédés Innovants" (MEPI) is a piloting and demonstration facility for Process Intensification technologies, operating since end of 2007 in Toulouse, France. Based on the facility concept described above, MEPI presents the following features:

- An industrial technical platform combining academic and industrial R&D for piloting and industrial demonstrations for chemical and biochemical processes with a production range from 0.5 to 10 Kg/h.
- A facility for demonstration of the industrial feasibility of Process Intensification, especially in the fine-pharma sector and green chemistry,
- A place for the production of representative lots for start-ups.
- Cluster of excellence in process intensification and renewable carbon processes (agro-resources).
- A tripartite platform between university, equipment providers and industry which proposes a more "productive" relationship that goes beyond the traditional two-partner collaborations (figure 2).

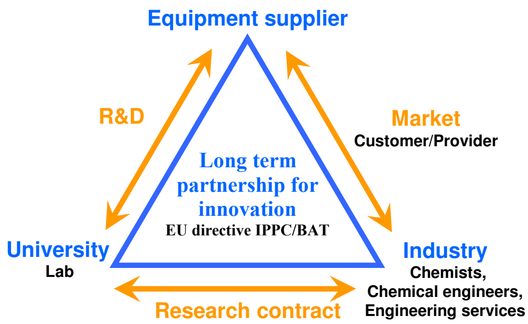

Figure 2: MEPI: a tripartite platform

In actual facts, such a relationship can be emphasized by the diversity of members of the facility, as described in table 1. MEPI is located in Toulouse, inside a place of geographical interest:

- inside the SNPE Materiaux Energetiques production site, that offers relevant infrastructures and a real safety culture,
- close to the "Cancer-Bio-Santé" pole of excellence dedicated to health industries,
- close to a dynamic and innovative university in Process Intensification.

Table 1: MEPI members

Industrials		Academics
SANOFI-AVENTIS	SNPE Matériaux Energétiques	INP Toulouse/ ENSIACET
PIERRE FABRE	ISOCHEM	Université Paul Sabatier
CORNING SA	VEOLIA	INSA
SOLIVONIC	LIBRAGEN	

MEPI OFFER & SERVICES

MEPI has been created to answer to a lack of piloting and demonstration facility in Process Intensification. In this way, the platform aims at making the link between applied research and consulting & knowledge dissemination, as described on figure 3. In fact, this link is strengthened on one hand by the strong partnership of Toulouse's University and transfer technology centers (funding members of MEPI) inside a French University network and on the other end by the role of Toulouse's University inside Europic.

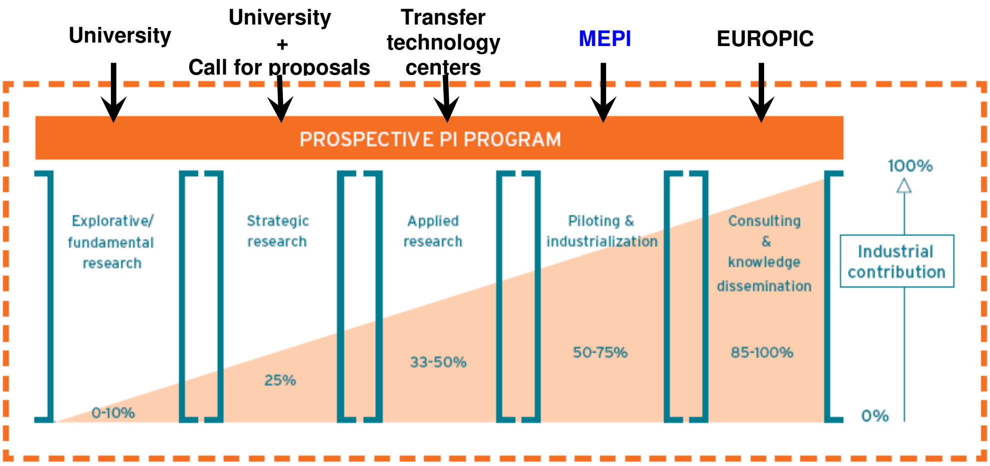

Figure 3: MEPI the missing link in Process Intensification development
Courtesy: Creative Energy, www.creative-energy.org

As Piloting and Demonstration of Process Intensification require specific information related to the considered application and/or process to satisfactorily be performed and as specific post-studies have to be carried for industrialization and scale-up, MEPI offers a large panel of services (figure 4). According to customer requirement, these "upstream" and "downstream" studies are performed directly with MEPI resources or in connection with Labs and Engineering services.

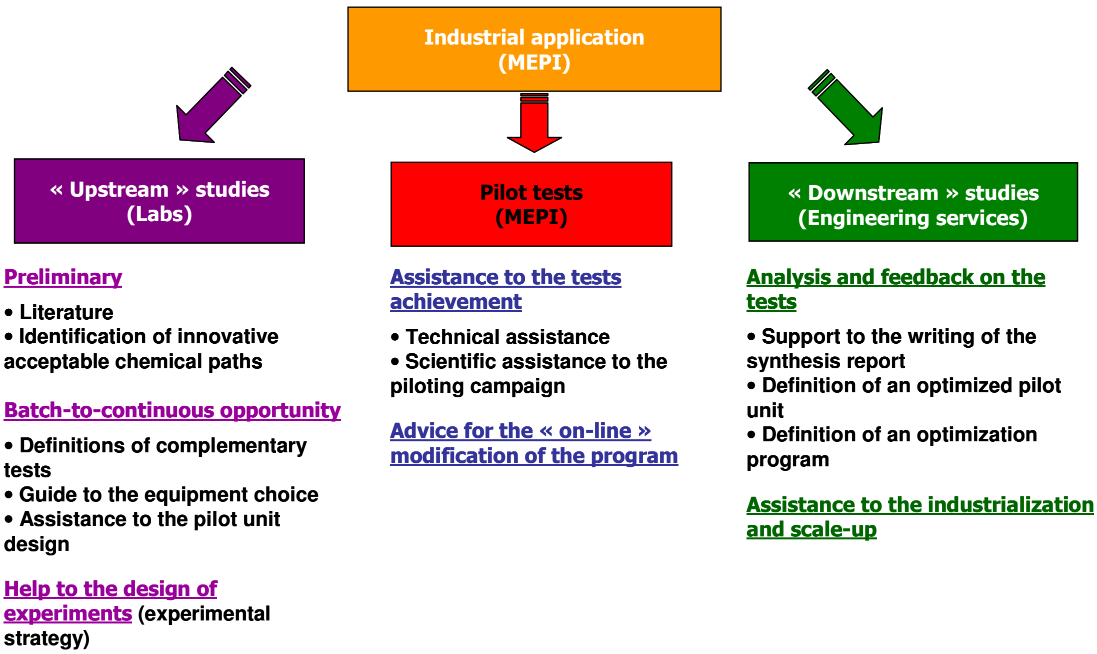

Figure 4: MEPI offer & services

MEPI APPLICATION EXAMPLE

To emphasize the benefit linked to MEPI on industrial applications, a specific Process Intensification example is presented. This example focuses on the addition reaction of Electrophile on Nucleophile in a two-phase medium (liquid-liquid). The production actually carried out in fed-batch mode is characterised by selectivity issues that lead to additional post-treatment steps. The study was initially carried out by MEPI to test the feasibility of the transposition from batch to continuous and of Process Intensification. With regards to the significant results obtained, an additional campaign was then performed to finalize and optimize the continuous process based on micro-structured reactors.

Application characteristics

The purpose of the application is to synthesize Product (P) from the addition of Electrophile (E) on Nucleophile (N), as described on figure 5. The possible addition of Electrophile on Product leads to the formation of By-Product (BP) and then to selectivity issues. Industrially, the By-Product has to comply with a less than 2% weight specification (determined by Gas Chromatography analysis).

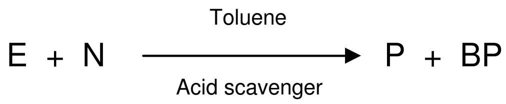

Figure 5: Reaction scheme

The reaction is carried out with toluene as solvent and organic acid scavenger to pick up chloride ions and prevent them from reacting with the product (by-product formation). According to the characteristics of reactants, the reactive medium consists of two liquid phases: Electrophile and main product in an organic layer, Nucleophile in another layer.

Industrially, the production is realized in a 4 m^3 batch reactor. For selectivity reasons, the temperature inside the reactor is kept constant to room temperature and Electrophile is continuously fed during several hours. Moreover a large excess of Nucleophile is considered (10 times compared to stoechiometry) to favour the formation of Product. Such conditions lead to a 4h operating time for the reaction, followed by many post-treatment steps:

- Water based washing steps to remove unreacted excess of raw material and acid scavenger.
- Distillation step to remove solvent.
- Distillation step to concentrate Product and remove a part of By-Product.

Process design

Process Intensification and transposition from Batch to continuous has been considered complying both By-Product specification (< 2%) and a reduction of Nucleophile excess. The process has been defined from the MEPI knowledge and experience, using Corning micro-structured reactor and leveraging Corning know-how in continuous processing and reactor design[7]. Thus, the process integrates three different parts as shown on figure 6:

- A mixing zone for the reactants injection, where the reaction is initiated.
- A residence time zone with thermal control, where the reaction completes.
- A quench zone, to prepare reaction for the post-treatment steps.

The choice and the location of the reactants feed lines has been carefully defined to avoid two-phase issues (injection of only homogeneous phase) and to comply with the pump constraints in terms of flow-rate range. With regards to viscosity, the pressure drops involved by each feed line has also been taken into account, as it strongly influences the mixing performances. Therefore, Nucleophile and Acid scavenger in toluene solution are both introduced in a first mixing structure and the resulting flow is mixed with Electrophile (also in toluene solution). Moreover, a specific Corning device has been used for the injection of Electrophile which is internally splitted into four different flows, in order to favour selectivity.

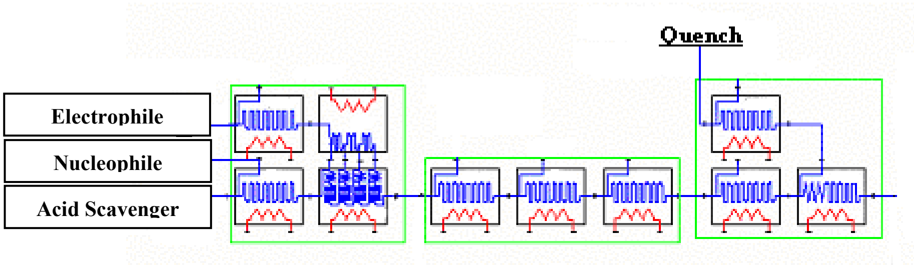

Figure 6: Intensified process based on micro-structured reactors

Optimal control

Based on the process previously designed, the study has been performed within 6 weeks, in two different steps: firstly a preliminary step to assess the feasibility and validate the process design and secondly an optimal control step to define the suitable operating conditions related to the production constraints. In this way, various parameters have been investigated: flow-rates of the different reactants, temperature, residence time and solvent use. The associated results highlight two sets of conditions of industrial interest. The first one is more unconventional as it allows the production without solvent to be carried out. Nevertheless in that case, even if all the production constraints are satisfied, the excess of Nucleophile can not be reduced. The second set of conditions lies on a heating of the reactive medium at 60°C (instead of room temperature in batch) and a good ratio between reactants. Such conditions lead to a satisfactory conversion with a By-Product concentration in the range from 0.99 % to 1.50 %. To emphasize the benefits related to this process intensification, table 1 offers a comparison between the Batch production conditions and the continuous ones. It is important to notice that in that case, with regards to the By-Product concentration at the end of the reaction, a distillation step usually performed during the post-treatment can be avoided.

Table 1: Comparison of production process

	Batch	Continuous
Reactor volume	4 m^3	100 mL
Conversion	> 95 %	> 95 %
Selectivity (By-Product fraction)	≈ 2 %	Average 1.5 % - Max 0.99 %
Excess of Nucleophile	10 equivalents	4 to 5 equivalents
Temperature	Room Temperature (1 atm)	60°C (3.5 bar)
Reaction time	4 h (including addition)	2 min
Productivity	20 T/year	17.5 T/year
Analysis	GC at reaction end	On-line Raman spectroscopy

SHORT REVIEW OF MEPI ACTIVITY

Since MEPI is operating (about 15 months), different applications have been studied and in all cases, significant improvements have been highlighted. To give an overview of MEPI possibilities, a summary of the most demonstrative applications is presented in table 2. This review doesn't aim at being exhaustive but at showing some kinds of applications that have been successfully intensified and transposed to continuous. It is important to notice that all the applications mentioned below have been carried out in Corning micro-structured reactors. Moreover, only 5 kinds of modules[7] have been used to reach the optimal design related to each application. This result emphasizes the polyvalence and flexibility features offered to process design by Corning micro-structured reactors.

Table 2: Review of some applications studied by MEPI

Reaction	Medium	Key-points	Objectives	Results	Duration
Addition of Electrophile	Two-phase (L/L)	Selectivity	Selectivity improvement	▪ Selectivity improved ▪ Reduction of reactant excess ▪ Solvent free	6 weeks
Organo-metallic reaction	Homogeneous Anhydrous (THF solvent)	Exothermicity Safety Temperature level (-70°C)	Feasibility in continuous	▪ Feasibility validated ▪ Higher temperature level (-40°C)	10 weeks
Addition of Nucleophile	Two-phase (G/L)	Safety	Estimation of the Scale down impact	▪ Conversion increase ▪ No reduction of reactant excess	2 weeks
Nucleophilic substitution	Homogeneous or Two-phase (L/L)	Selectivity Safety	Feasibility in continuous Selectivity improvement Screening and choice of base and solvent	▪ Feasibility validated ▪ Conversion increase	10 weeks
Ionic liquid synthesis	Homogeneous then Two or Three phase (G/L/L)	Exothermicity Mass transfer Reaction time	Process intensification	▪ Feasibility validated	1 week
Peroxydation reaction	Three-phase (G/L/L)	Singlet oxygen Gaz hold-up Safety	Feasibility in continuous Significant yield	▪ Feasibility validated ▪ Process optimisation (screening of solvent)	8 weeks

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