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Multilevel Phenomenological Modelling Approach to Support the Evaluation and Generation of Intensified Processes

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Abstract

The paper presents a first approach for the systematisation of the fundamentals contained in the PI developments to support their implementation into new process designs. An initial methodology based on a multilevel analysis is proposed, starting with the identification and integration of the main tasks (process stages) by the material involved, functions to be delivered and appropriate process conditions. A more detailed level deals with the refinement of each task using a phenomenological modelling approach which aids in the construction and analysis of models whose ultimate purpose is providing the insights in the process performance and understanding on how cooperative phenomena can be manipulated for the accomplishment of the design strategies that promote the intensification of processes.

Within the fundamentals of the proposed phenomenological modelling approach that guide the model structuring, the process tasks are decomposed into the relevant phases and physicochemical phenomena involved, identifying the connections and influences amongst them and evaluating the individual rates. The intensification of rate-limiting phenomena is finally achieved by diverse strategies derived from PI principles, dealing with the manipulation of driving forces.

Keywords: Process Intensification, Process Design, multilevel modelling, phenomena.

1. Introduction

Process Intensification (PI) has developed as a novel and revolutionary design approach that provides advantages in size and cost reduction and a closer approach to optimal processing conditions. However, despite the numerous advantages PI may provide, the implementation of this concept into process designs has generally been limited to the application of highly efficient equipment developments based on specific case studies. Some attempts have been made for the generation of a methodology for implementation of PI (e.g. Wood et. al. 1997). However, none of them provides specific systematic strategies to encourage intensified designs that go beyond interdisciplinary brainstorming for the generation of alternatives.

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Adaptation of processes to conventional unit operations implies the acceptance of the inherent inefficiencies of the associated equipment. PI aims to provide the ideal process environment by designing the equipment that matches the fundamental process requirements. In this way, novel process configurations and devices are developed. This work aims to understand and structure the key knowledge implicit in the PI developments in order to identify the requirements of the modelling tools to support and encourage the innovative development of intensified and integrated processes.
Generation and evaluation of these novel alternatives requires the fundamental analysis and understanding of process phenomena without the spatial constraints of conventional unit operation models and ultimately equipment. In this way, the systematic synthesis of intensified processes is not adequately supported by current approaches, requiring methods and tools that are beyond the available equipment-driven methodologies for process design and modular unit-operation-based simulation tools. A new structure of these systems is proposed by redefining the fundamental building blocks. This contribution develops a more flexible phenomena-based modelling framework with multilevel characteristics for the refinement of process models. The principles here presented should be used for the future implementation in computer-aided tools that adequately support the generation and evaluation of intensified processes.

2. Multilevel phenomenological approach for the refinement of process models

2.1 Main approaches for PI

After carrying out a performance analysis of the major PI developments, two main design principles for PI have been identified. These two key principles are complementary and should be used as the key guidelines for the implementation of PI at each step of the process development. These are consistent to the PI principles identified by Rong et al. (2004), but allow a more unified representation of the PI knowledge.

1. Synergistic integration of process tasks and coupling of phenomena. It should be understood as the definition of the most appropriate environment to maximise the expected yield by grouping/ungrouping individual processes. Separation of tasks/phenomena should be considered if the process requires to be carried out in separate stages (e.g. due to presence of competitive reactions) and in order to increase the degrees of freedom in design (Pohjola et al. 1994).
2. Enhancement of transport processes. This strategy aims to intensify the rate of limiting phenomena to overcome mass transfer, heat transfer and hydrodynamic limitations. This is achieved by means of design strategies such as reduction of transfer paths, intensification of mixing at the diverse lengthscales, novel operating conditions such as the use of alternative energy sources, novel processing media and external fields that produce extreme process conditions, and in general promoting instabilities that generate an increase in the driving forces that have an effect on the rates of phenomena (e.g. differences in concentration, temperature, velocity, etc.)

2.2 Multilevel methodology for the implementation of PI principles

Generation of PI designs follows a general equipment-independent methodology in which the fundamental process performance is analysed and the requirements are identified. Design strategies based in the PI principles are simultaneously generated. The refinement of process models is performed at 3 levels: tasks-phases-phenomena.

2.2.1 Task-oriented approach

The initial level may be defined as the required transformation (e.g. as a chemical route). Main process tasks are identified and integrated or separated (level of tasks) by examining the commonalities in (a) functions, (b) material, and (c) process conditions. A similar approach has been applied by Siirola (1995) for the well known process integration for the manufacture of methyl acetate. The task-oriented approach aims to implement concepts related to the integration of tasks and coupling of phenomena, looking at the intensification of whole processes.

2.2.2 Phenomena-based approach

Each of the main process tasks is refined by means of the phases involved and their interrelationships (level of phases). Further refinement is achieved by means of the identification of the relevant phenomena (level of phenomena) taking place within the phases and between them. Design and modelling activities at this level includes the identification of the influences amongst phenomena and assessment of the process behaviour under certain conditions, quantification of rates and identification of limiting steps. The analysis of the process at this level aims to provide with the insights into the process performance to identify the fundamental requirements. Finally, generation of design strategies is based on PI principles, which in their deepest essence consist in the enhancement of desired phenomena and discouragement of undesired phenomena by means of the manipulation of the driving forces.

2.3 Multilevel phenomenological modelling approach

Based in the General Systems Theory (e.g. Klir 1969) and as developed by several authors (Woods 1993, Marquardt 1996, Dieterich and Eigenberger 1997, Gilles 1998), two levels are identified for the structurisation of models with a phenomena-based character. (1) The "structural level" consists in region elements (physical elements defined by permeable, semipermeable or impermeable boundaries, with accumulation of material and energy as the main attribute) and connection elements (elements that serve to couple region elements and enable the exchange of material and energy). Thermodynamic phases and physical interfaces have been defined as the basic region and connection elements respectively (which is also proposed by Perkins et al. 1996 and Mehta and Kokossis 1997). However, the concept may be expanded to include any class of physical regions with accumulation of material/energy such as boundary layers, so the term "level of generalised phases" should be more appropriate. (2) The "behavioural level" corresponds to the "level of phenomena". It is defined by accumulation, generation and transport of material and energy. These phenomena-based building blocks have a related mathematical expression to evaluate their rates, which can be defined by the terms in the balance equations as described by Gerstlauer et al. (1993). In this way, a more flexible modelling environment to support development of novel processes is achieved by means of the aggregation of the terms in the balance equations representing the phenomenological building blocks. For the level of phases and phenomena, a library of models can be defined to facilitate the construction of models. A topology has been devised analogous to the one developed by Wall et al. (2001) and Drengstig et al. (1997), which is shown in figure 2 for the Paracetamol example. The topology may be understood as the representation of the process tasks that the process design has to fulfil (i.e. the process requirements).

3. Paracetamol case study

3.1 Case description

Paracetamol (n-acetyl-p-aminophenol, APAP) is produced by the acetylation of p-aminophenol (PAP) with acetic anhydride (Ac2O) and sulphuric acid (H+). The catalytic hydrogenation of nitrobenzene (NB) in the presence of strong acids leads to the formation of phenylhydroxylamine (PHA), which rearranges to form the intermediate PAP. The diverse reactions taking place in this process have been sketched in figure 1. Roles have been assigned to desired (M-main) and undesired (S-side) reactions.

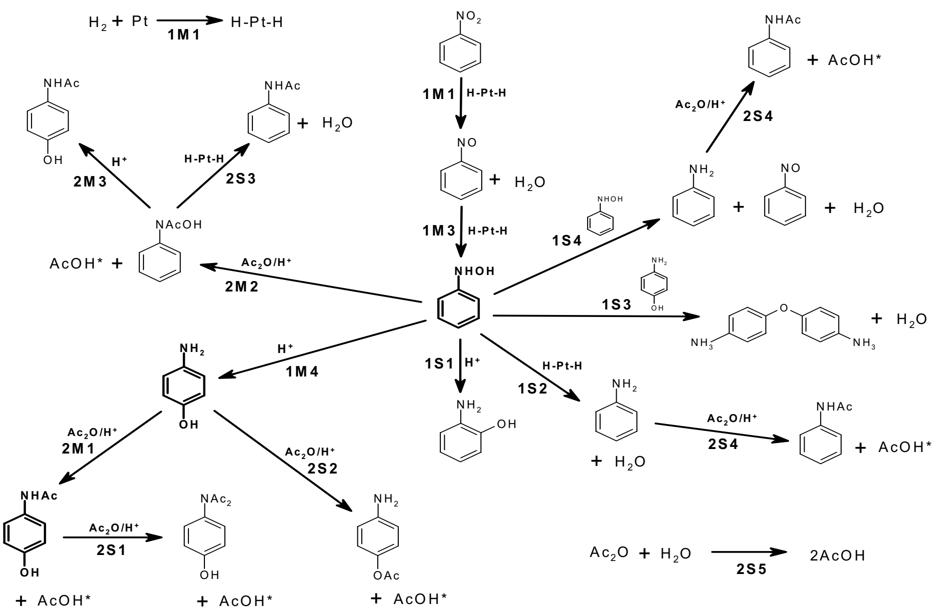

Figure 1. Reaction network for the production of Paracetamol (n-acetyl-p-aminophenol)

3.2 Task identification and integration

If the process is performed in one single step (or single vessel without internal boundaries), several side-reactions appear, consuming reactants and reducing the overall yield. Further separation of the product and byproducts, and separation and recycle of reactants introduces additional costs. Process can be split to perform hydrogenation and acetylation in independent steps. If intermediate PAP is isolated from the hydrogenation environment before acetylation it is possible to eliminate 2S3 by isolating 2M2. 2S4 and 2S5 are also stopped. Furthermore, hydrogenation should be split into two stages by removing PHA from the hydrogenation media (H-Pt-H) immediately after its formation in order to avoid 1S2. These tasks could be performed in one single vessel only if internal boundaries are introduced in such a way that in-situ separation of products avoids further side-reactions, e.g. by means of membranes or with another selective technique (extraction, adsorption, etc.) with an accurate control in the flow patterns.

3.3 Phenomena-based process representation

The hydrogenation step is represented in fig. 2 by means of the phenomena based concepts previously defined and the topology proposed. Wettability of the catalyst particles is the main point to consider when defining the contacting phases to be represented. Selection of the phase contact may have design reasons such as elimination of further reactions, e.g. over-hydrogenation of PHA (1S2) by removing the product from the neighbouring reaction zone (catalyst surface). PHA is more soluble in the inorganic phase, so if catalyst is surrounded predominantly by the organic phase (i.e. NB), PHA is forced to be transferred to the outer inorganic phase, reducing possibility of over-hydrogenation. If 1S2 is potentially eliminated by means of the in-situ separation of PHA, reaction with H+ to produce PAP may be carried out at the same time (in a single vessel). Furthermore, the availability of NB for the molecular contact and reaction with adsorbed hydrogen is higher due to shorter NB transfer paths. Additionally, NB can be recycled with the catalyst by decanting the inorganic phase.

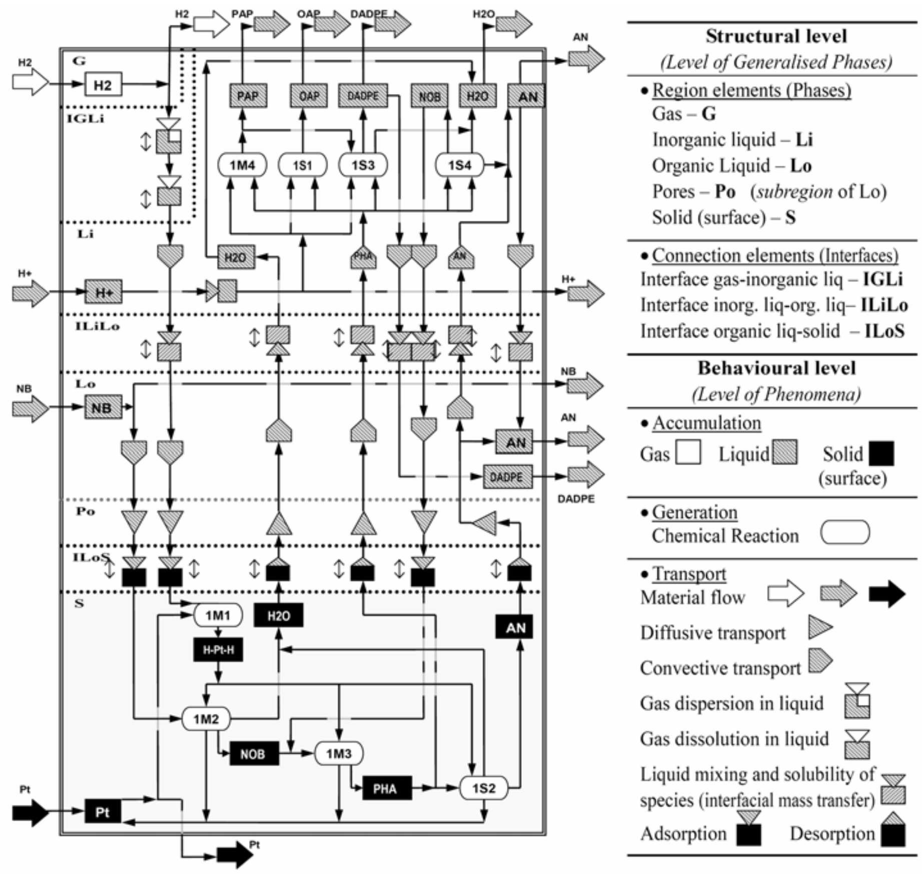

Figure 2. Phenomena-based representation for the first stage in the Paracetamol process

3.4 Implementation of PI principles as design strategies

Side reactions 1S2, 2S3, 2S4 and 2S5 have been eliminated by means of the task identification and integration principles. It is possible to generate strategies to reduce or eliminate the remaining undesired reactions by means of a more fundamental analysis of the process to estimate rates and identify main driving forces. Reaction 1M4 for the production of PAP competes with 1S1, 1S3 and 1S4. Reactions 1M4 and 1S4 are fast reactions compared to 1S1 and 1S3, so the former can be encouraged over the later by applying intensive mixing to avoid mass transfer limitations, and hence increasing selectivity. In this particular case, reaction 1M4 is exothermic and 1S4 is endothermic, so heat removal may encourage 1M4 and discourage 1S4 (and also endothermic reaction 1S3 if still present). Strategies to intensify heat transfer may be applied such as reduction of transfer paths (microtechnology) and intensive mixing plus intense heat removal. Similar considerations can be applied for the acetylation step, in which reaction 2M1 has a higher rate compared with 2S1 and 2S2. In order to avoid over-acetylation of APAP (2S1) a cocurrent contacting pattern in a plug flow may enable the reduction of Ac2O/H+ concentration as the reaction proceeds. In particular this reaction is slow so very long reactors may be needed, and the use of an oscillatory baffled reactor may represent the best option. Current process uses stirred tank reactors run by 8 hours in which there is a major possibility of over-acetylation.

4. Conclusions

The proposed phenomenological approach allows a more fundamental analysis for the identification of the process requirements and generation of design strategies. The phenomena-based building blocks do not constrain process to conventional unit operation/equipment boundaries. They can be aggregated or disaggregated according to the best performance and novel designs can be generated. These lower building blocks are the generalised phases, their connection, and the phenomena taking place in them. Phenomena can be basically defined by the terms from the balance equations, providing a link with the mathematical modelling. The structuring fundamentals and the link to PI principles here presented should be used as a theoretical framework for the modularisation of most classes of chemical processes, enabling maintainability and reusability of models. The structure here developed should serve for the future computer implementation in a more flexible system that adequately supports innovative design.

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