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A conceptual framework for description of complexity in intensive chemical processes

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ABSTRACT

A possible conceptualisation formalism for high-level description of complex reaction systems is presented. The formalism is intended as a framework for adopting multi-scale modelling and simulation tools to the problems of reverse design of intensive processes and for design of functional materials. The formalism is exemplified through several historical cases from the fields of reaction and materials' engineering.

1. Introduction

Current approaches to development of new chemical processes and materials moved away from the traditional sequential approach, towards a more integrated approach based on mixed-skills teams, developing in parallel the chemistry, reactor technology, as well as considering life-cycle issues and considering potential routes to market and supply-chain issues. The two approaches are schematically shown in Fig. 1. The latter approach offers considerable benefits, such as *e.g.*, avoidance of scale-up problems emerging at a late process development stage, faster time to market and inclusion of environmental and toxicity criteria at an early stages of the R&D process. These benefits are achieved primarily due to a better understanding of process chemistry, which enables faster identification of process options and avoidance of potential scale-up or environmental compliance issues of the final process or a product. This approach is also entirely consistent with the principles of process intensification [1,2]: detailed understanding of a reaction mechanism, physical chemistry of the reaction system and its dynamics (rates of reaction, heat and mass transfer) enables one to generate concepts for an optimal reaction environment, maximising the reactor performance, avoiding unnecessary unit operations, reducing plant size and environmental footprint, considering alternative feedstocks. The new approach leads to faster practical implementation of novel intensified technologies. Two current examples where the new R&D approach is being used to develop and implement novel intensive processes are the large projects of the European Commission "F3 Factory" (www.f3factory.com) and "SYNFLOW" (www.synflow.eu). The two projects are aimed at developing intensive catalytic processes and their demonstration within a common platform of a flexible manufacturing process. Specifically, the development of novel flow processes based on organometallic catalysis was shown to require a complex parallel development route for which the systematic development tools are not yet available [3].

Two features of this approach, which are both essential for process intensification and important for the discussion in this paper are: (*i*) simultaneous consideration of a given problem at multiple length and time scales, *i.e.*, molecular scale processes that could practically occur at 10^{-3}–10^{3} s time-frame *vs* plant-scale over 10^{1}–10^{5} s time-frame (disregarding for now supply-chain optimisation problems which are of much large length- and time-scales), and (*ii*) the increased need for large amount of process and molecular data at an early stage of R&D. Thus, it can be argued that the transition towards a more efficient R&D model highlighted the inherent complexity of chemical processes, which was often insufficiently appreciated when a complete problem was broken down into traditional narrow boundaries of the individual scientific disciplines. Thus, the new R&D process requires the means to deal with the apparent complexity, as well as new tools providing fast access to large quantities of data and the capacity for their interpretation.

This paper provides a brief overview of the issue of complexity in chemical reaction systems and the existing concepts and methods that can be applied to this new challenge. The paper puts forward a

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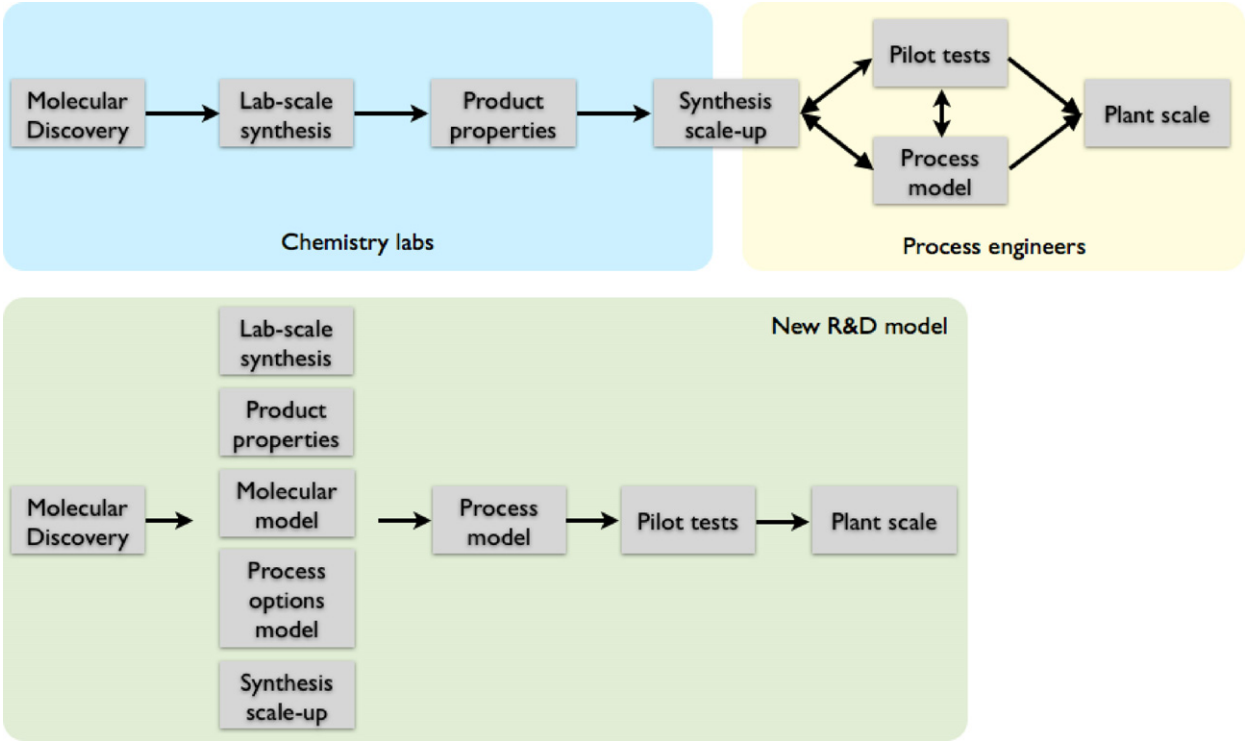

Fig. 1. Illustration of a traditional sequential and the current parallel R&D processes.

potential conceptualisation framework, that enables better understanding of complex hierarchical and temporal problems within the context of chemical processes. The basis of the framework has been established and developed in the neighbouring disciplines of systems engineering, but apparently had not crossed the boundary into chemical processing.

Examples of the problems aimed by the proposed methodology are the integrated intensified catalytic reactor systems combining several functionalities, *e.g.*, three-phase reactors-heat exchangers for selective oxidation reactions [4] and other catalytic microstructured reactors [5]. The proposed framework is generic and offers itself for developing specific computational tools on the basis of the existing methods. As presented, the proposed framework is more generic (higher level), than the recently published conceptualisation framework for multiscale modelling developed by Yang and Marquardt [6]. The relevance of the two alternative approaches to each other is also discussed.

Section 2 of the paper provides several examples of the types of problems in chemistry and chemical processing that may be addressed through application of the proposed systematic analysis methodology. Section 3 describes the high-level conceptualisation framework as a tool for systematic analysis of complex problems. Finally Section 4 provides analysis of several historic examples within the conceptual framework.

2. *Complexity* and *systems* in chemistry and processes

The term 'complexity' is usually applied to situations involving multiple interacting components. An accessible review gives an up-to-date list of different measures and definitions of complexity [7]. Here we focus on complexity as quality and understand it to include the presence of strongly non-linear behaviour within a chemical system under consideration, and the presence of emergent behaviour in complex chemical systems. The concept of emergence is central to definition of complex systems [7] and is highly relevant to chemistry, chemical processes and industry. The most obvious examples within the field of chemical reactions that fit this description are phenomena displaying complex structure-, composition- and time-dependant behaviour, *e.g.*, time-dependent self-organisation of micelles [8], oscillatory reactions, *e.g.*, of the Belousov-Zhabotinsky type [9], multiple steady-states and thermal runaway in exothermic catalytic batch reactions, and many more across the very different fields of chemistry.

Recently the term 'systems chemistry' emerged in the field of synthetic organic chemistry with respect to understanding of kinetics and reaction pathways within mixtures of reactive intermediates [10,11], and extended to analysis of possible syntheses routes using networks theory [12]. Such systems are not unlike metabolic pathways within biological reaction networks, which are described as classical 'complex systems' [7]. Hence is the similarity in terminology with systems biology [13]. In both cases the term 'systems' is used to highlight the presence of emergent behaviour and the need for a different treatment to that of simpler problems. The difference in the approach is the use of holistic analysis of a system's behaviour, rather than a reduction of complexity to the simpler linear relationships, which is necessarily associated with the loss of information. However, the emergent behaviour is relevant not only to organic synthesis, but also to almost any type of problems in chemistry and chemical processes. Hence, there is a need to expand the tools of systems analysis beyond organic synthesis. Recent literature advocates for development of such methods to aid in the advancement of *sustainable* chemical processes [14,15].

A step-up in complexity of a system is a problem including not only a reaction network, but also the reaction environment. This may include solvent effects, effects due to the presence of several phases within a reaction environment, effects due to self-organisation, *etc*. Thus, the presence of immiscible or partially miscible fluids affects the immediate reaction environment in the large class of interface reactions. The presence of free surfaces induces strong transformations in molecular geometry and ener-