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Setting optimal operating conditions for a catalytic reactor for butane oxidation using parametric sensitivity analysis and failure probability indices

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ABSTRACT

Safe operation of a catalytic reactor remains a sensitive issue when highly exothermic reactions are conducted and hazardous side reactions may occur. Derivation of the optimal operating conditions must include economic but also safety criteria, technological constraints, beside controllability and stability aspects. The present work introduces a criterion based on a joint failure probability index related to uncertainty in the runaway boundaries and the random disturbances of the operating parameters. The use of such a safety criterion is even more important when setting the optimal operating policy of the reactor in the vicinity of the runaway boundaries that often correspond to a high productivity. The paper indicates how an economically efficient but more prudent operating policy can be selected, by simultaneously considering the economic and safety objectives. An example is provided for the case of an industrial fixed-bed tubular reactor, of high thermal sensitivity, used for the catalytic oxidation of butane to maleic anhydride in vapour phase. The multi-objective optimization can lead to a prudent trade-off operating solution (corresponding to a failure probability of maximum 3–4%) that limits the reactor productivity. Being based on a local and global sensitivity analysis of the reactor, the proposed rule is generally applicable by minimizing the probability with which control variables violate their safety limits.

1. Introduction

Optimal setting of the operating parameters and input conditions for an industrial reactor is one of the most challenging engineering problems, as long as the chemical reactor is the core unit of any chemical plant where the products are synthesised. Chemical reactors are often the main sources of runaway risk. The high thermal sensitivity is related to the intrinsic non-linear nature of the chemical processes, due to the exponential dependence of the reaction rate vs. temperature, together with the combination of phenomena taking place involving simultaneous reactions, momentum, mass and energy transfer, often in multiphase systems. On the other hand, many chemical systems achieve their maximum performance when operating close to their limits. Such systems can be highly vulnerable to external disturbances, resulting in catastrophic changes of the expected behaviour. There is an interest to operate the systems very near their limits, in response to today's growing performance demands (reactor productivity), despite the previous observation. These systems might jump to new low-performance operating points, behaving oscillatory or chaotically, or even collapse, when facing with disturbances or design uncertainty. However, this complex behaviour could be avoided by an adequate optimal control including precise estimation of the safety limits for the control variables.

When hazardous exothermal reactions are conducted, the reactor presents a high thermal sensitivity to random disturbances in the running conditions, and its operation in the vicinity of safety limits becomes risky. A model-based analysis has to consider several aspects, when coping with the problem of reactor productivity maximization but keeping a small operating risk (i.e. a small failure probability), with fulfilling the technological constraints: a precise evaluation of the safety limits in the parametric/control variable space; estimation of the safety limit uncertainty associated to the model imperfections, reactor operation disturbances, or the calculation algorithm used; the use of a synthetic index to characterize the thermal runaway risk; the use of a multi-objective optimization to get the best trade-off between economic and safe operating policy.

The concept of *parametric sensitivity* was used to identify the critical conditions under which relatively small changes in the input variables produce significant changes in the state and output variables, to account for safety aspects in a more systematic way

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during reactor operation. As disturbances are unavoidable during operation, dangerous situations may arise around these conditions, leading to uncontrollable evolutions. This is why precise evaluation of the safety limits becomes a crucial engineering problem.

Derivation of runaway boundaries (critical conditions) of the operating variables can be made by a large variety of methods, from simple to complex ones (Grewer, 1994; Stoessel, 2008; Varma, Morbidelli, & Wu, 1999). Simple explicit methods derive the safety limits from a thermal sensitivity analysis based on engineering numbers and relationships valid for singular zero- or first-order reactions (Grewer, 1994). Such methods are not sufficiently accurate for an advanced optimization of the process or for implementing an on-line instability detector, the practical approach (but not the most efficient one) providing sufficient overdesign of the system (Seider, Brengel, Provost, & Widagdo, 1990).

Alternatively, model based evaluations of critical operating conditions of a reactor, even computationally more intensive, can offer a quite accurate prediction of the safety limits, being generally applicable irrespectively of the reactor type or process. According to Adrover, Creta, Giona, and Valorani (2007) such criteria can be classified into four categories: geometry-based criteria (interpreting the shape of the temperature or heat-release rate profile over the reaction/contact time), parametric sensitivity-based criteria (detecting unsafe conditions characterized by high parametric sensitivities of state variables vs. operating parameters), divergence-based criteria (detecting any instability or incipient divergence from a reference state-variable trajectory over the reaction time), and stretching based criteria (investigating the process divergence from the nominal condition from the dynamics of the tangent components to the state-variable trajectory). Each method presents advantages and limitations related to precision, real-time application, involved computational effort, and applicability. A detailed comparative analysis is given by Varma et al. (1999); Adrover et al. (2007); Maria and Stefan (2010a, 2010b, 2011).

To determine the optimal reactor operating conditions with explicitly accounting for safety indices, the classical deterministic approach consists in searching for the optimal *nominal* values (or dynamic policy) of the input or manipulated/control variables (e.g. feed flow rate, inlet concentration of the co-reactants, cooling agent temperature, overall pressure), that ensure extremization of a suitable objective function (i.e. the performance objective, or 'cost' function in financial or engineering terms), by fulfilling the differential (mass, heat, momentum) balance equations, and the technological-safety constraints (Kadam, Schlegel, Srinivasan, Bonvin, & Marquardt, 2007; Li, Arellano-Garcia, & Wozny, 2008; Maria & Dan, in press; Srinivasan, Palanki, & Bonvin, 2002). Safety requirements are accounted for rather empirically by using parameter or state variable thresholds as runaway boundaries (Bonvin, 1998; Muske, Badlani, Dell'Orco, & Brum, 2004; Ruppen, Bonvin, & Rippin, 1997), or by limiting the rate of generating heat during the reaction (Toulouse, Cezerac, Cabassud, Le Lann, & Casamatta, 1996), in the classic approach. Other approaches explicitly formulate constraints to limit the temperature sensitivity vs. operating parameters, or try to keep a certain distance between the set-point and runaway boundaries to cope with the parametric uncertainty (review of Maria & Dan, in press). Recent developments check safety constraints by generating scenario-integrated process evolution via repeated simulations over a certain time horizon at the expense of a considerable computational effort (Abel & Marquardt, 2003).

This deterministic approach, even if it is computationally attractive, can sometimes lead to risky predictions for the reactor optimal running-point due to the missed uncertainty aspects, such as (Abel & Marquardt, 2003; Bonvin, 1998; Maria & Dan, in press; Smets, Claes, November, Bastin, & Van Impe, 2004): time-varying characteristics of the process (catalyst properties, raw-materials) requiring a continuous model up-dating; a high degree of uncertainty in model structure and its parameters (due to incomplete experimental information and adopted simplifying hypotheses), in operating variables (due to random disturbances), and in constraint formulation (many times based on heuristic/direct observations).

When parametric uncertainty is considered during reactor optimization, the system runaway (failure) probability is associated to the probability with which stochastic control variables/operating parameters will overstep the runaway boundaries during operation. Consequently, the performance criterion to be extremized and the process variables are expressed in stochastic terms, thus a *robust* control solution being obtained. There are two ways to develop the stochastic approach.

i) Some researches are focused on the "risk of not getting the expected value of the performance criterion" predicted by the optimal operating solution, or the risk of violating the constraints due to random perturbations of parameters or initial conditions ("differential sensitivity analysis", or "robust model predictive optimal control"; Li et al., 2008; Nagy & Braatz, 2004; Srinivasan et al., 2002; Terwiesch, Agarwal, & Rippin, 1994). The open-loop optimization (i.e. without on-line state feedback for improving the model) in the stochastic sense is computationally expensive, requiring derivation of the expected value of the optimization criterion, its variance, but also sensitivities of model functions and constraints vs. stochastic control variables (Li et al., 2008), irrespectively of the formulated criterion (usually of economic nature; Diehl, Bock, & Kostina, 2006; Nagy & Allgöwer, 2007). Consequently, several simplifying alternatives have been developed: a) minimize the worst-case objective (i.e. the "minmax" approach, or "best-worst-case"), by minimizing the expected value of the (multi-) objective function (Diehl et al., 2006; Nagy & Braatz, 2004); b) consider a trade-off solution between *nominal* (deterministic) and *robust* (stochastic) performance objectives; for instance, the parametric uncertainty can be considered only when minimizing the sensitivity of the performance index vs. parametric random variations; c) optimization of the "performance quality" that can be achieved with a specified level of confidence (Terwiesch et al., 1994). Several implementation details have been reported because the stochastic approach is computationally intensive. For instance, because minimization of the expected value of the performance criterion, and of its variance, requires a repeated numerical integration over the whole multi-dimensional parametric space (of known distribution), a first- or a second-order variance approximation has been proposed (Nagy & Allgöwer, 2007).

ii) Other researches are focused on defining safety objectives to be simultaneously optimized with economic goals when searching for a feasible problem solution. Safety indices are included in a deterministic way (based on the distance between the running point and the runaway boundaries, or based-on state variable sensitivities vs. operating parameters), or in a stochastic way (by evaluating the probability that output variables violate imposed upper/lower bounds of the technological constraints; Li et al., 2008).

All such strategies suffer from the lack of a systematic model-based derivation of the safety limits and their associated uncertainty to be further considered in minimizing the risk of violating the safety limits during reactor operation by the presence of random disturbances.