Multi-objective optimization of tubular fixed-bed reactor for CO oxidative coupling by comprehensive simulation and machine learning

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Highlights

- Continuous reactor safety function was proposed for optimization.
- Simultaneous optimization of reactor safety and performance was done by using NSGA-II for the different heat-transfer scheme.
- Higher yield was achieved in co-current scheme with intrinsic safety.

1. Introduction

Flow direction of the coolant stream is an important factor for the heat management in the multi-tubular reactor. It is helpful in achieving the productivity with lowest value of the maximum temperature and parameter sensitivity among three basic coolant configurations: co-current, isotherm, counter-current coolant.[1, 2] Researches on the heat transfer mode of the multi-tubular reactor mainly focus on the flowrate of the coolant and lack optimization of other parameters.[3-5] However, the change in cooling mode has a huge impact on the reactor temperature distribution, which indirectly affects the reaction rate and pressure drop, and all process parameters need to be adjusted.

Here, we performed systematic studies on isothermal heat-transfer multi-tubular reactor and co-current one based on CO oxidative coupling with methyl nitrite (MN) to dimethyl oxalate process. Industrially, MN conversion is typically less than 50%, calling for further reactor improvements and optimization. Three parallel reactions are involved with two side reactions of higher activation energy than the main reaction. In a nutshell, it is necessary and representative to optimize CO oxidative coupling reactor with different heat transfer modes.

2. Methods

Non-dominated Sorting Genetic Algorithm (NSGA-II) is applied for multi-objective optimization of the simultaneous function of performance and safety. The one for performance is given by as below.

$$Obj(1) = -10Y_{DMO} + 100(1 - X_{MN}) + 500(1 - S_{DMO}) + \sigma(-\Delta P - 150)(-\Delta P - 150) \tag{1}$$

Reactor simulations were conducted by a comprehensive two-dimensional heterogeneous model to get the single-tube performance: capacity (Y_{DMO}), conversion ($\mathrm{X_{MN}}$), selectivity ($\mathrm{S_{DMO}}$) and pressure drop(ΔP). Based on the previous research, higher CO content ($\mathrm{y_{CO}}$) is favored[6], and here is fixed at 30% by the explosion limit of the reactant mixture. The upper bound of $\mathrm{y_{MN}}$, $\mathrm{y_{NO}}$, $\mathrm{T_{in}}$, $\mathrm{P_{in}}$, GHSV, coolant axial flowrate are 25%, 10%, 413.15K, 700 kPa, 5000 $\mathrm{h^{-1}}$, and 1 kg/s/tube respectively, and the lower are 12%, 5%, 373.15K, 400 kPa, 3000 $\mathrm{h^{-1}}$, and 0.001 kg/s/tube respectively. *Obj*(2) represents reactor safety, cases are divided and handled. When thermal runaway occurs, *Obj*(2) is taken as positive infinite (+*Inf*). If hotspot appears, *Obj*(2) is taken as :

$$Obj(2) = \partial^2 T / \partial z^2 \big|_{z=z(T_{g,\max})} - 4U / u_f \rho_f C_{p,f} D_t \, dT_w / dz \tag{2}$$

If the reactor is under pseudo-adiabatic operation, one can examine when the whole reactor axial temperature second-order derivative appears positive, *Obj*(2) is +*Inf*, and *vice versa* is 0.

3. Results and discussion

From Figure 2(a), it can be seen that the co-current scheme expands the predominance region, i.e. it is possible to safely enhance conversion and yield. There is a positive correlation between the *Obj*(1) and $\mathrm{X_{MN}}$, which allows to adopt $\mathrm{X_{MN}}$ as the horizontal coordinate for the change in optimal conditions. From Figure 2(b), the capacity of the reactor is higher even $\mathrm{X_{MN}}$ is the same, resulting from two reasons. Firstly, lower inlet temperature allows the reactor to accept higher inlet concentrations, as shown in Figure 3(a, b). Besides, higher temperatures of the reactor tail on the one hand require the reactor to be pressurized in order to reduce the pressure drop, and on the other hand create conditions for higher space velocity. Different from isothermal mode, the co-current one regulates performance primarily through space velocity rather than coolant inlet temperature when X_{MN} between 60-80%. Higher conversions over 80 % require higher y_{MN}, but this greatly induce safety trouble, and the reactor is more sensitive to reactant concentration.

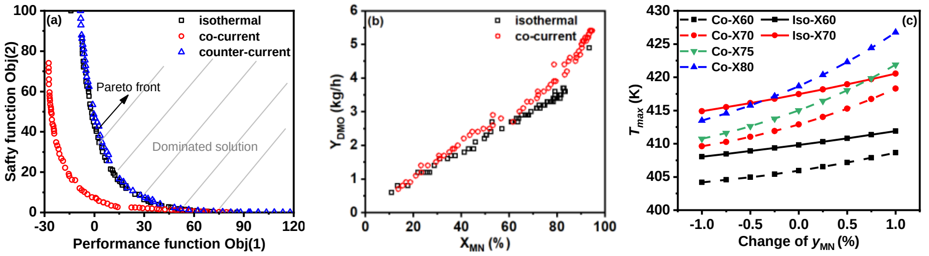

Figure 2. Optimized results with different heat transfer scheme by NSGA-II.

(a) Pareto optimal front, (b) capacity of Pareto optimal results, (c) sensitivity analysis of representative results.

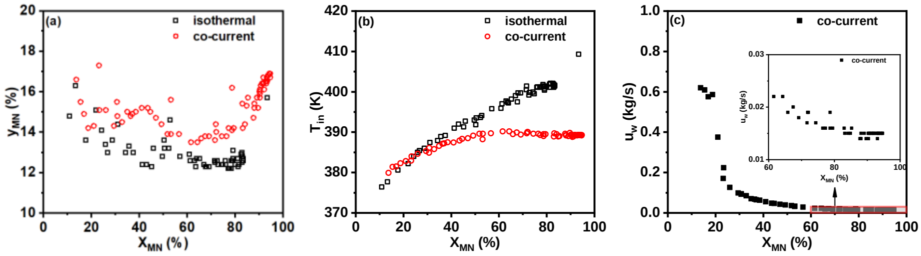

Figure 3. Optimized reactor conditions with different heat transfer scheme by NSGA-II

Sensitivity analysis of the results for different conversion in Figure 2(c) shows that the co-current reactors are more sensitive to changes in y_{MN} and that the reaction conditions can be selected by the temperature after the positive stimulus to preserve enough space for temperature rise. As a result, conditions of isothermal reactor with 70 % conversion and that of co-current reactor with 75 % conversion are chosen. As a result, the DMO capacity is increased by 26 % and 43 %, respectively, compared to the base case of the industrial reactor (0.55 $g_{DMO}/(g_{cat}\cdot h)$).

4. Conclusions

This work addressed the optimization of the multi-tubular reactor based on the rigorous modeling and genetic algorithm. The co-current heat-transfer reactor can achieve high conversion and capacity while ensuring safety operation according to the multi-objective optimization results. A global optimization of the reactor is achieved, which can assure a significant increase in reactor production.

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Keywords

Reactor optimization; NSGA-II; CO oxidative coupling.