COMPUTATIONAL FLUID DYNAMICS-BASED MULTIOBJECTIVE OPTIMIZATION FOR CATALYST DESIGN

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Summary

For an industrial secondary methane steam reformer with regular packing, catalyst design is accomplished by an integrated optimization approach, which includes design of experiment, computational fluid dynamics (CFD) simulation, response surface method and genetic algorithm, for multiobjective optimization. Both spherical and cylindrical catalysts are studied. The reactor performance considered for the catalyst design includes the pressure drop and the hydrogen production, which constitute the binary objective functions for optimization. The optimal solutions reveal that large pore diameter, near 1 μm, should be adopted for spherical catalysts. For cylindrical catalysts, the optimal design suggests the use of 1-big-hole shape with larger particle and pore size, 10-13 mm and near 1μm, or 4-hole shape with smaller particle size of 6-8 mm.

Keywords

Rational design of catalysts.

Introduction

The design of catalysts affects the reactor performances in many aspects, including pressure drop, reaction extent, product distribution, etc.[1] CFD simulation, as an effective tool for reactor design[2], can be adopted for the optimization study of catalyst design. An integrated optimization approach[3] as shown in Fig. 1 is essential to lessen the computational work and make the task feasible. This paper presents the optimization results for the packed bed reactors using spherical or cylindrical catalysts.

CFD simulation

The reactor configuration has a tube-to-particle diameter ratio (N) of 4 and the arrangement of spherical and cylindrical particles are shown in Fig. 2 with references to Dixon et al.[4,5], where only a 120° wall segment is simulated. The commercial software Fluent is used for the simulation. The turbulent flow is simulated using the k-ε RNG model with enhanced wall treatment. The catalysts are treated as a porous media type of fluid. The reactor studied is the secondary reformer in the industrial ammonia plant using Ni based catalyst and the kinetics from De Groote and Froment[6] and Xu and Froment[7]. Grid independent studies are conducted to determine the appropriate cell sizes. User defined functions are provided for reaction kinetics and property calculations, such as the component diffusivities inside the catalyst particles to consider the Kundsen and molecular diffusions.

Integrated optimization approach

The Latin hypercube experimental design method (LHD) is first employed to determine the experimental points for CFD simulation. Response surface method is then used to establish the quadratic functions of the objective functions and the decision variables of the optimization analysis. Finally, the genetic algorithm method, NSGA-II[8], is adopted for obtaining the optimal solutions.

The binary objective functions are the pressure drop per unit length of reactor (f_1) and the hydrogen product generation rate per unit volume of reactor (f_2) and the decision variables are the shape, the particle size (d_P) and the pore diameter (d_{pore}) of the catalyst. The shapes of the equilateral cylindrical catalysts studied include solid (no-hole), one-hole, one-large-hole, three-hole, four-hole and four-small-hole. The studied ranges of the catalyst size is 6-17 mm and the pore diameter is 0.01-1 μm.

The inlet gas conditions are based on typical industrial plant's data[9]. The superficial velocity is fixed for all simulation cases.

Using LHD with 50% extra points, the experimental points for spherical and cylindrical catalysts are 9 and 16, respectively. The multiple linear regression method is applied to determine the quadratic relation between each objective function and the decision variables.

$$f = \beta_0 + \sum_{i=1}^{d} \beta_i x_i + \sum_{i=1}^{d} \beta_{ii} x_i^2 + \sum_{j=2}^{d} \sum_{i=1}^{j-1} \beta_{ij} x_i x_j + \varepsilon \qquad (1)$$

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The (R^2, R^2_{adj}) of the regression results for the spherical and cylindrical cases are higher than (99.7%, 99.3%) and (99.8%, 96.7%), respectively.

With the population size of 50 and the generation number of 200, the multiobjective optimization using NSGA-II[8] provides 50 optimal solutions. For the spherical catalyst case, the optimal solutions indicate that pore diameter is not the deterministic variable and the largest pore diameter should be used, conversely, the particle diameter gives rise to the trade-offs between the binary objective functions as shown in Fig. 3. For the cylindrical catalyst case, the optimal solutions are shown in Fig. 4. The catalyst shape is either one-large-hole or four-hole. For the former, the larger pore diameter of 1 μm and particle size of 10~13 mm should be used. For the latter, the pore diameter effect is not significant and the particle size should be 6~8 mm.

Conclusions

The study has successfully accomplished the multiobjective optimization of the catalyst design for regular packed bed reactors by an integrated optimization approach utilizing the CFD simulation. The optimal solutions preferentially suggest the shape, particle size and pore diameter of the catalysts.

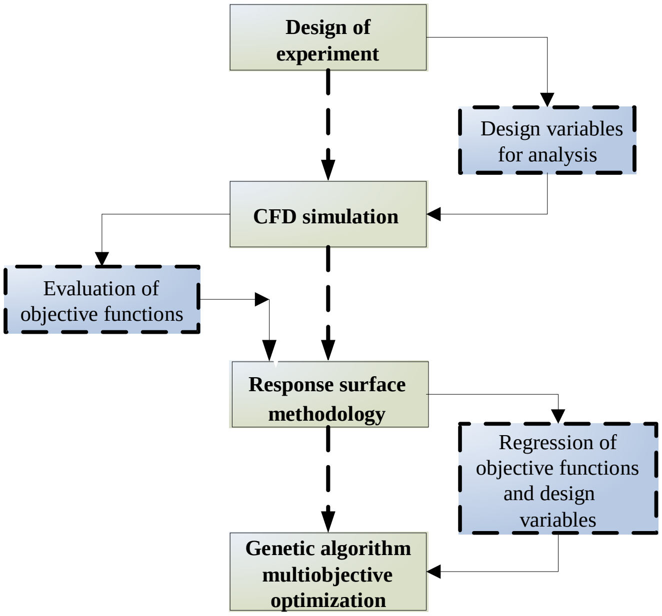
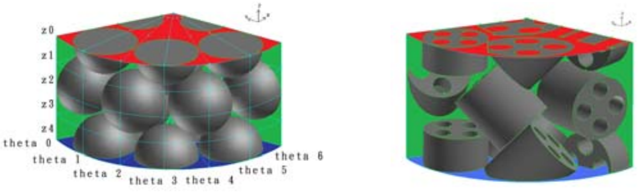
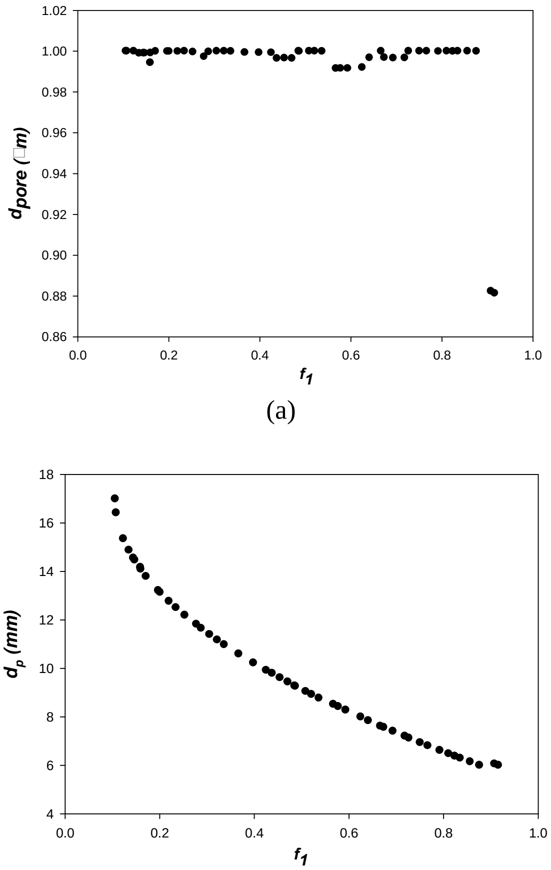


Fig. 1 Integrated optimization approach



(a) Spherical catalyst (b) Cylindrical catalyst

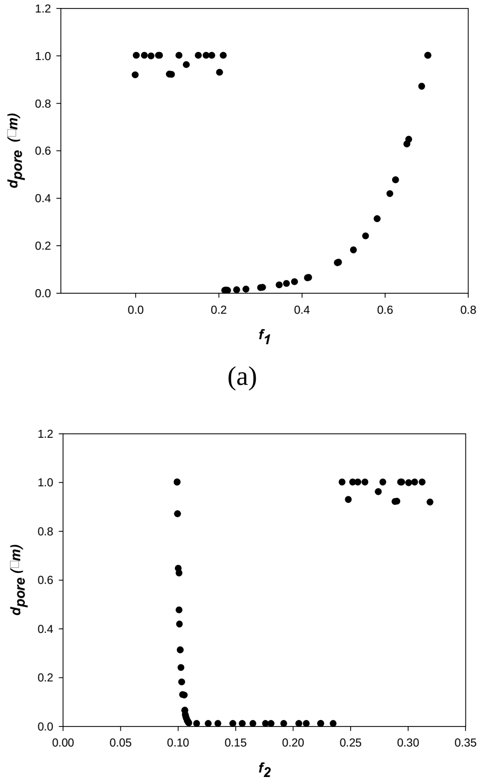
Fig. 2 Catalyst arrangement



(a)

(b)

Fig. 3 Optimal solutions for spherical catalyst case



(a)

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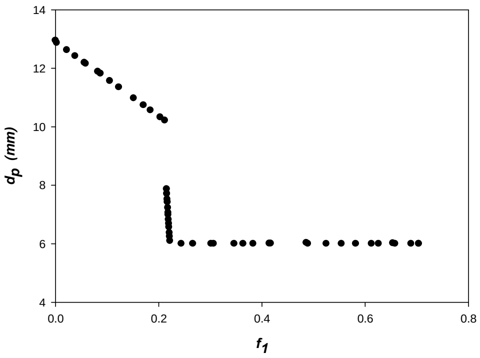
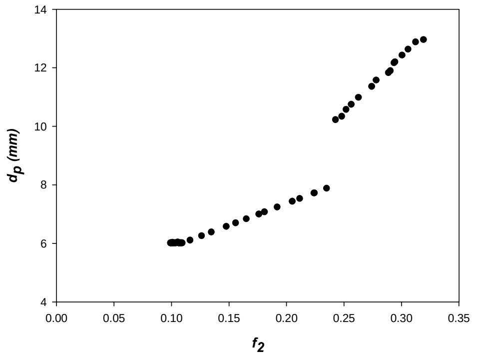
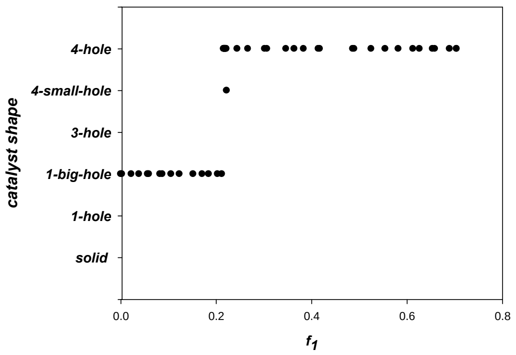


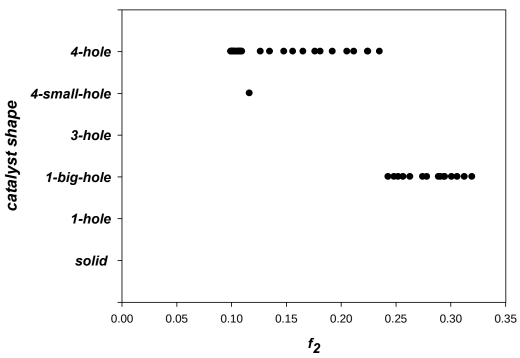
(c)



(d)



(e)



(f)

Fig. 4 Optimal solutions for cylindrical catalyst case

Acknowledgement

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