A Parametric Investigation of Impregnation and Drying of Supported Catalysts

Xue Liu, Benjamin J. Glasser* and Johannes Khinast
Department of Chemical and Biochemical Engineering, Rutgers University, Piscataway, NJ, 08854
* bglasser@soemail.rutgers.edu

Introduction

Supported catalysts are essential components of many industrial processes with applications, ranging from catalytic converters to fuel cells. They are generally required because of their high surface area, reduced amount of active agent (usually an expensive metal such as Pd, Pt or Ru), and high thermal stability. The performance of a catalytic process is intimately related to the catalyst design. There are four main categories of catalyst profiles, uniform, egg-yolk, egg-shell and egg-white. The choice of the optimal catalyst profile in the support is determined by the required activity, selectivity, and by other characteristics of the chemical reaction (kinetics, mass transfer). It is generally believed that the metal profile is controlled by the conditions that are applied during impregnation where the metal contacts the solid support for the first time. However, experimental work has shown that drying may also significantly impact the metal distribution within the support [1]. Therefore, to achieve a desired metal profile we need to understand both impregnation and drying.

Materials and Methods

For impregnation, we assumed that the support is pre-wetted so capillary forces are not considered and the distribution of the metal precursor on the support is due to diffusion and adsorption. We also developed a drying model, which takes into account the convective flow of the liquid solvent due to capillary forces, the diffusion of the metal due to the gradient of concentration, metal adsorption onto the support, and the convective and diffusive transport in the gas phase [2,3]. The initial input of the drying model comes from the results from impregnation simulations. The system of equations is discretized by a finite volume method, and the resulting ordinary differential equations are solved using LIMEX [4], a solver for highly stiff differential–algebraic equations.

Results and Discussion

In the present work, uniform or egg-shell profiles are obtained by impregnation of one component onto the support. The effect of impregnation time, diffusion coefficient and adsorption constant is investigated for the impregnation procedure. A temporal development of the catalyst distribution is shown in Fig 1. Clearly, an egg-shell profile can be obtained for a relatively short impregnation time, while a uniform profile can be observed if the impregnation time is long enough. The speed of the impregnation front increases with an increase in the diffusion coefficient, and decreases with an increase in the adsorption constant (not shown). The effect of the drying temperature on the redistribution of the metal is shown in Fig 2 with an initial condition observed in Fig 1 (t=10000s). It can be seen that the egg-shell profile can be enhanced under the fast drying condition and slow drying leads to a uniform distribution. Egg-white or egg-yolk profiles are obtained by competitive impregnation of two components. We found the adsorption constant and the initial concentration of the coimpregnant metal have a strong effect on the final distribution of the desired metal.

Significance

Our main goal is to deepen our understanding of the impact of impregnation and drying on the final catalyst profile through mathematical modeling. We investigated the importance of the physical properties of the solid support (permeability, particle size), liquid solution (diffusion, initial metal precursor concentration) and operational conditions (humidity and temperature in the drying air), and studied the effect of interactions that exist between the dissolved metal and the solid support (physical adsorption, crystallization). The parametric investigation can improve our understanding of the preparation of supported catalysts.

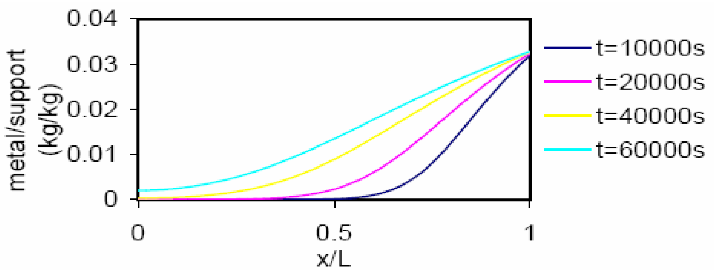

Figure 1. Impregnation front verse time

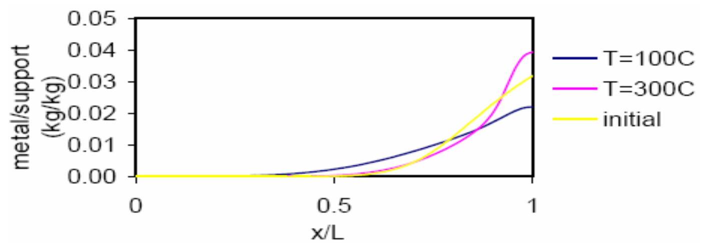

Figure 2. Effect of drying temperature on the metal redistribution

References

1. Lee, S.Y. and Aris, R. *Catalysis Reviews-Science Engineering* 27, 207 (1985).
2. Lekhal, A., Glasser, B.J., and Khinast, J.G. *Chemical Engineering Science* 56, 4473 (2001).
3. Lekhal, A., Khinast, J.G., and Glasser, B.J. *Ind. Eng. Chem. Res.* 40, 3989 (2001).
4. Deuflhard, P., Hairer, E., and Zugck, J. *Numerische Mathematik* 51, 501 (1987).