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Partial regeneration of Ni-based catalysts for hydrogen production via methane cracking part II: modeling and optimization

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Abstract: High purity, carbon monoxide-free hydrogen and filamentous carbon can be produced by thermo-catalytic cracking of methane. Carbon filaments continue to grow until the catalyst deactivates because of carbon encapsulation. Regeneration of catalyst is important to maintain a continuous process. Our work on optimization of the partial regeneration method showed that activity of the catalyst can be sustained for longer times by gasifying not all but some extent of the deposited carbon. In the previous work, a kinetic model was developed to be able to understand the reaction mechanism of deactivation of fresh catalyst at the molecular level. The objective of this study was to develop a well-fitted kinetic model for deactivation of regenerated catalyst. A kinetic model that consists of surface reactions, filament formation, and deactivation was developed for the regenerated catalyst. It is assumed that reaction parameters at the molecular level do not change when the burn off degree is at a moderate extent but these parameters changed drastically when burn off degree is vastly increased or decreased. Rate constant for the encapsulation reaction is adjusted for the simulation results to be representative of the experimental results (typically specific weight of carbon (g C/g Ni) vs. time on stream). The system of differential algebraic equations consists of the steady-state equations for all surface intermediates, an algebraic equation of dissolution/segregation, diffusion equation, and the site balance equation. The system has been solved without assuming any rate-determining step or most abundant surface intermediates. Parameter estimation procedures were repeated for the deactivation cycles of regenerated 5 wt% Ni/γ-Al_2O_3 catalyst. The basic idea that underlies the model is that every carbon atom will diffuse through the nickel particle and participate in the formation of carbon filaments until the catalyst deactivates. Specific weight of carbon is calculated using the rate of carbon diffusion.

Key Words: Thermo catalytic cracking of methane, hydrogen, filamentous carbon, partial regeneration, Ni/γ-Al_2O_3 catalyst, kinetic modeling, estimation of rate constant.

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