報告番号	甲　第　14233　号

主　論　文　の　要　旨

論文題目　Numerical Study on Chemical Reactions and Transport Phenomenon in The Solid Catalyst Fixed Bed for Methanation of Carbon Dioxide and Dry Reforming of Methane
(二酸化炭素メタネーションおよびメタン乾式改質触媒固定床における化学反応と移動現象に関する数値解析的研究)

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論　文　内　容　の　要　旨

Carbon capture and utilization and carbon capture and storage (CCU and CCS) are promising strategies for the energy crisis and climate change. The present study focuses on the CO_2 utilization process to do further investigation. Two promising processes for CO_2 utilization were investigated, which are CO_2 methanation and dry reforming of methane (DRM) processes.

CO_2 methanation is one of the promising technologies for CO_2 utilization. It should be mentioned that, however, the temperature runaway phenomenon is the critical problem that limits industrial application for the CO_2 methanation due to the exothermic nature. This section applied the bed dilution method to solve the temperature runaway phenomenon in the CO_2 methanation process. Based on the lattice kinetic scheme-lattice Boltzmann method (LKS-LBM), the effect of bed dilution structure on temperature

profile and the carbon conversion rate was investigated. Numerical results indicated that under the restriction of a constant number of catalysts, the temperature profile could be decreased by increasing the volume fraction of inert particles. Based on the critical temperature (800 K), the configuration is optimal when the volume percentage of inert particles is equal to 33.3 %. Moreover, the optimal inert particle distribution was found, which can not only control the peak temperature below the critical temperature but also improve the carbon conversion rate by almost 18% compared with the structure without dilution under the same circumstance.

After that, a CFD model was developed to investigate comprehensively CO_2 methanation process in a shell-and-tube reactor. The model was verified by available experimental data. The effects of several operating factors on the temperature profile and carbon conversion were explored: gas flow rate, feed gas composition, catalyst pellet thermal conductivity, cooling phase intake temperature, and phase transition refrigeration. The results indicate that Increasing gas flow rate results in an increase of temperature profile and carbon conversion. Similarly, decreasing the inlet mole fraction of CO_2 also can promote temperature profile and carbon conversion. Lowering the inlet temperature of the cooling phase can reduce the peak temperature in the reactor, but the carbon conversion will also drop sharply. Moreover, catalyst pellet thermal conductivity is a significant parameter in CO_2 methanation process. Increasing catalyst pellet thermal conductivity can reduce peak temperature in the reactor and maintain almost the same level of carbon conversion.

The main issue For the DRM process is coking, which results in catalyst deactivation. In this section, based on a multi-component LBM model, macro-mesopores structure in a catalyst pellet was applied to investigate the coking process. A catalyst pellet with

macro-mesopore structure was constructed by the modified random generation of macro-meso pores (RGMMP) algorithm. The effect of three pore structure parameters on catalytic performance was explored: the catalyst porosity, the ratio of mesopore volume to macropore volume, and the ratio of average macropore diameter to average mesopore diameter. The result indicates a competitive relationship between the number of active sites and intraparticle diffusion. The optimal value of catalyst porosity and the ratio of average macropore diameter to average mesopore diameter can be found. Moreover, with a constant porosity limitation, The number of active sites is scarcely affected by the pore volume ratio of mesopore to macropore. The intraparticle diffusion is the key limiting factor for carbon deposition. Therefore, the optimal values for catalyst porosity, the ratio of mesopore volume to macropore volume, and the ratio of average macropore diameter to average mesopore diameter were found to be 0.7, 0.5 and 5, respectively, for a reaction condition (Ni/Al_2O_3 catalyst, operating temperature: T=923.15 K, operating pressure: P=1 bar, CH_4/CO_2 feed ratio=1:1).