In-Situ CO_2 Capture Using $CaO/\gamma\text{-}Al_2O_3$ Washcoated Monoliths for Sorption Enhanced Water Gas Shift Reaction

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ABSTRACT:

In-situ capture of CO_2 allows the thermodynamically constrained water gas shift (WGS) process to operate at higher temperatures (i.e. 350^{o}C) where reaction kinetics are more favorable. Dispersed $CaO/\gamma\text{-}Al_2O_3$ was investigated as a sorbent for in-situ CO_2 capture for an enhanced water gas shift application. The CO_2 adsorbent ($CaO/\gamma\text{-}Al_2O_3$) and WGS catalyst ($Pt/\gamma\text{-}Al_2O_3$) were integrated as multiple layers of washcoats on a monolith structure. CO_2 capture experiments were performed using thermal gravimetric analysis (TGA) and a bench scale flow through reactor. Enhancement of the water gas shift (EWGS) reaction was demonstrated using monoliths (400 cells/in^2) washcoated with separate layers of dispersed $CaO/\gamma\text{-}Al_2O_3$ and $Pt/\gamma\text{-}Al_2O_3$ in a flow reactor. Capture experiments in a reactor using monoliths coated with $CaO/\gamma\text{-}Al_2O_3$ indicated that increased concentrations of steam in the reactant mixture increase the capture capacity of the CO_2 adsorbent as well as the extent of regeneration. A maximum capture capacity of 0.63 moles CO_2/kg sorbent (for 8.4% CaO on $\gamma\text{-}Al_2O_3$ washcoated with a loading of 3.45 g/in^3 on monolith) was observed at 350^{o}C for a reactant mixture consisting of 10% CO_2, 28% steam and balance N_2. Hydrogen production was enhanced in the presence of monoliths coated with a layer of 1% $Pt/\gamma\text{-}Al_2O_3$ and a separate layer of 9.4% $CaO/\gamma\text{-}Al_2O_3$; A greater volume of hydrogen compared to the baseline WGS case was produced over a fixed amount of time for multiple cycles of EWGS. The CO conversion was enhanced beyond equilibrium during the period of rapid CO_2 capture by the nano-dispersed adsorbent. Following saturation of the adsorbent, the monoliths were regenerated (CO_2 was released) in-situ, at temperatures far below the temperature required for decomposition of bulk $CaCO_3$. It was demonstrated that the water gas shift reaction could be enhanced for at least 9 cycles with in-situ regeneration of adsorbent between cycles. Isothermal regeneration with only steam was shown to be a feasible method for developing a process.

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