LITHIUM SILICATES FOR ISOTHERMAL SORPTION-ENHANCED STEAM METHANE REFORMING

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Conventional steam-methane reforming (SMR) accounts for the roughly 50% of the world's supply of hydrogen. The process consists of separate SMR and WGS steps with complex heat integration, the economics of the process are strongly influenced by scale. This has inspired efforts of process intensification via sorption-enhanced reforming (SE-SMR), in which the CO_2 is chemically fixed to a suitable sorbent, thus driving the equilibrium to produce more hydrogen at lower temperatures. The selection of sorbent is key: for CaO sorbent, efforts have reached the pilot stage. With this technology, capital and operating costs have shown significant reductions from current technology; however, the CaCO3 adsorbed phase must be heated to 800 temperature-swing process still requires extensive heat integration and favors large economies of scale.

Lithium-based CO_2 sorbents, notably lithium orthosilicates (Li_4SiO_4, or LOS), have been evaluated as candidates for SE-SMR. In contrast to CaO, however, the adsorbed product (Li_2CO_3 and Li_2SiO_3) does not require a temperature change to reactivate the sorbent, thus enabling a simpler pressure-swing adsorption process. At Villanova, we developed a novel surfactant-assisted approach to preparing a nanostructured LOS has been developed by adding a polyether amine surfactant to the sol-gel preparations. Preferred LOS compositions sorb 26 wt% CO_2 within 1 minute from a 50 vol% CO_2/N_2 stream and show no observed loss in a 33 wt% working capacity in over 30 one-hour isothermal cycles. Physically meaningful rate constants and diffusivities are obtained by an adaptation of an analytical solution to the diffusion-reaction model, confirming faster reaction and diffusion kinetics for these high-performing CO_2 sorbents. These sorbents should find application not only in SE-SMR for hydrogen production, but also in integrated gasification combined cycle (IGCC) processes for power production, and blast-furnace top gas valorization in steel manufacture.