UNIVERSITY of ROCHESTER

Department of Chemical Engineering presents

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"Oxidative Coupling of Methane over Doped Rare-Earth Oxides"

The oxidative coupling of methane (OCM) is a potential pathway to directly convert methane to highly valuable ethylene, and other hydrocarbon products. However, it is very difficult to obtain high yields of C_{2+} product (i.e. products such as ethane, ethylene and higher hydrocarbons), due to the formation of more thermodynamically favored higher oxidation products, such as CO and CO₂ (CO_x). Li/MgO is a simple and active OCM catalyst with a relatively high C₂ product selectivity, but it suffers from severe deactivation due to the volatility of lithium under OCM reaction conditions. Sesquioxides of rare earth oxides, such as samaria (Sm_2O_3) and lanthana (La_2O_3), are amongst the most efficient single component oxide catalysts in the methane coupling reaction. In our laboratory we are interested in how doping affects rare earth oxides and their activity, selectivity, and long-term stability in the oxidative coupling of methane. We have investigated the effects of doping on samarium, terbium, praseodymium and cerium oxides, as they represent a "non-reducible" sesquioxide (samaria) and reducible rare earth oxides (terbia, praseodymia and ceria). The reducible oxides are expected to be more selective to Cox products than C_{2+} products. However, we have shown that addition of lithium and sodium to these oxides can increase the activity and selectivity toward OCM significantly. In fact, Li-doped terbium oxide supported on an MgO support is not only a more active and selective OCM catalyst compared with the reference Li/MgO, Sm₂O₃/MgO and Li-Sm₂O₃/MgO catalysts, it is also more stable with time on stream. The higher stability appears to be due to a stronger interaction between lithium and terbia compared with lithium and samaria, as evidenced by both X-ray diffraction and X-ray photoelectron spectroscopy measurements. We are currently investigating the effects of transition metal dopants on rare earth oxides in the oxidative coupling of methane.