Encapsulated iron oxide nanoparticles for chemical looping based hydrogen production

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Chemical looping (CL) based hydrogen production involves a two-step process in which in the first step a synthesis gas reduces a metal oxide (Me_xO_y) oxygen carrier (Eqs. 1,2). Subsequently, the reduced metal oxide is re-oxidized using steam to produce a pure stream of H_2 (Eq. 3).

$$H_2 + Me_xO_y \leftrightarrow H_2O + Me_xO_{y-1} \quad (1)$$

$$CO + Me_xO_y \leftrightarrow CO_2 + Me_xO_{y-1} \quad (2)$$

$$H_2O + Me_xO_{y-1} \leftrightarrow H_2 + Me_xO_y \quad (3)$$

An appropriate oxygen carrier for the CL-based H_2 production must possess the following characteristics[1]:

(i) Thermodynamics that favor the complete conversion of synthesis gas to CO_2 and H_2O.

(ii) Regenerable with steam with a high 'hydrogen to steam' ratio.

(iii) Fast and stable kinetics over multiple cycles.

In this regard iron oxide is arguably the most promising material due to favorable thermodynamics, wide availability and being environmentally benign[1,2]. However, the lack of sufficient redox stability of pure Fe_2O_3 at high temperatures (700-1000 ˚C) is a major concern. Bohn et al.[3] demonstrated that unsupported Fe_2O_3 deactivates after only a few redox cycle. A possible solution to stabilize the redox performance of iron oxide is by supporting it on a high Tammann temperature material, such as SiO_2, TiO_2, ZrO_2, or CeO_2[4]. Within this strategy fall core-shell nanomaterials which have shown promising results in stabilizing the activity of various catalysts.[5]

Here we report the development of iron oxide nanoparticles encapsulated by SiO_2, CeO_2 and ZrO_2 for the chemical looping-based production of hydrogen. The materials were characterized by high resolution transmission electron microscope (HR-TEM), powder X-ray diffraction and N_2 adsorption. The redox performance of the synthesized oxygen carriers were assessed in a thermo-gravimetric analyser. The shell thickness is optimized to ensure a good compromise between redox stability and mass transfer characteristics. The newly developed materials show an improved stability against sintering when compared to conventionally supported oxygen carriers.

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