Supplemental Information

Chemically and Physically Robust, Commercially-viable Iron-based Composite Oxygen Carrier sustainable over 3000 Redox Cycles at High Temperatures for Chemical Looping Applications

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Experimental section

Reactivity Measurement. The oxygen carrier samples (around 25 mg for each sample) were mounted in an alumina crucible and run through 3000 reduction-oxidation cycles at high temperatures using a Setaram SETSYS Evolution Thermogravimetric Analyzer (TGA). The reduction step used a 250 mL/min flow of gas containing 20% H_2, 20% CO, 40% CO_2 balanced with N_2 at 1000°C. Oxidation step was performed using 200 mL/min flow of air at 1000 °C. The reduction and oxidation steps lasted for 1.5 minutes and 5 minutes each. One redox cycle is consisted of one reduction step followed by a 3 minutes flushing step with N_2 (50 mL/min) and one oxidation step with a 2.5-minute flushing step with N_2 (50 mL/min) as well.

Crushing Strength Measurement. Crushing strength measurement was carried out by placing one spherical oxygen carrier pellet on a testing stand consist of a smooth steel surface and a vertically mounted force gauge (SHIMPO FGE-200XY). A PC was used to record the force data, in Newton, at a sampling rate of 100 Hz while the gauge moved downward to crush the oxygen carrier. Crushing strength (MPa) was obtained by dividing the first recorded peak of the gauge by the cross-section area of the sample.

Attrition Measurement. The jet cup used in this study was a modified version of the original design published by Particulate Solid Research, Inc. (PSRI). [1] A disengagement zone with a designed cut-off size of 53 microns was attached above the jet-cup to ensure that only fine particles underwent elutriation

while majority of the particles remain. An additional particulate filter was installed at the gas outlet to assist in closing the mass balance. Sieve analysis was carried out for 100 g of oxygen carrier sample prior to being placed in the jet-cup. For this study, jet velocity of 144 m/s of air was injected tangentially at the bottom of the jet-cup. Air flowrate was stopped periodically over the course of one hour to perform particle size distribution analysis with both sieves and the recorded weight change in the particulate filter. The attrition index presented is defined as the accumulative weight fractions of particles below the cut-off size in the disengagement zone.

Reference

[1] Cocco, R., Arrington, Y., Hays, R., Findlay, J., Karri, S. B. R., & Knowlton, T. M. (2010). Jet cup attrition testing. *Powder Technology*, *200*(3), 224-233.