

U.S. DEPARTMENT OF ENERGY  
OFFICE OF FOSSIL ENERGY  
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## OXYGEN CARRIER DEVELOPMENT FOR CHEMICAL LOOPING COMBUSTION

### Background

Chemical looping combustion (CLC) is a flameless combustion technology where there is no direct contact between air and fuel. The CLC process utilizes oxygen from metal oxide oxygen carrier for fuel combustion. The products of CLC are  $\text{CO}_2$  and  $\text{H}_2\text{O}$ . Thus, once the steam is condensed, a relatively pure stream of  $\text{CO}_2$  is produced ready for sequestration. The many benefits of this combustion process are that it does not produce  $\text{NO}_x$ , its production of a sequestration ready  $\text{CO}_2$  stream does not require additional separation units, and there is no energy penalty or reduction in power plant efficiency.

The majority of the work performed to date on CLC has been performed using gaseous fuels. There are only limited studies with oxygen carriers used to combust solid fuels such as coal. Those few studies that have been performed using coal suggest that more experimental information is needed. Development of oxygen carriers that have stable performance during multiple cycles and understanding the reaction mechanism and other process parameters are critical for the success of the CLC process.

### Accomplishments

The combustion and re-oxidation properties of direct coal chemical-looping combustion (CLC) over  $\text{CuO}$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{Co}_3\text{O}_4$ ,  $\text{NiO}$ , and  $\text{Mn}_2\text{O}_3$  were investigated by using thermogravimetric analysis (TGA) and bench-scale fixed-bed flow reactor studies. When coal/metal oxide was heated either in nitrogen or carbon dioxide ( $\text{CO}_2$ ), weight losses were observed due to partial pyrolysis and coal combustion by metal oxides. Among various metal oxides evaluated,  $\text{CuO}$  showed the best reaction properties:  $\text{CuO}$  can initiate the reduction reaction as low as  $500^\circ\text{C}$  and complete the full combustion at  $700^\circ\text{C}$ . In addition, the reduced copper can be fully re-oxidized by air at  $700^\circ\text{C}$ . The combustion products formed during the CLC reaction of coal/metal oxide mixture are  $\text{CO}_2$  and water, while no carbon monoxide was observed. Multi-cycle TGA tests and bench-scale fixed-bed flow reactor tests strongly supported the feasibility of chemical-looping combustion of coal by using  $\text{CuO}$  as an oxygen carrier. The interactions of flyash with metal oxides were investigated by X-ray diffraction and thermodynamic analysis. Overall, the results indicated that it is feasible to develop chemical-looping combustion with coal by metal oxides as oxygen carriers. Reaction mechanism studies and measurement of rate parameters necessary for reactor design work have also been completed.

### Benefits

Chemical looping combustion can lead to increased power efficiency. CLC produces a stream of combustion products that primarily consist of  $\text{CO}_2$  and steam. A relatively pure stream of  $\text{CO}_2$  that is sequestration ready can be produced by simply

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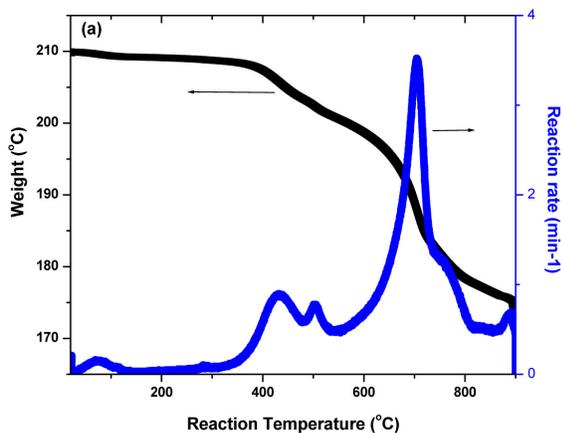
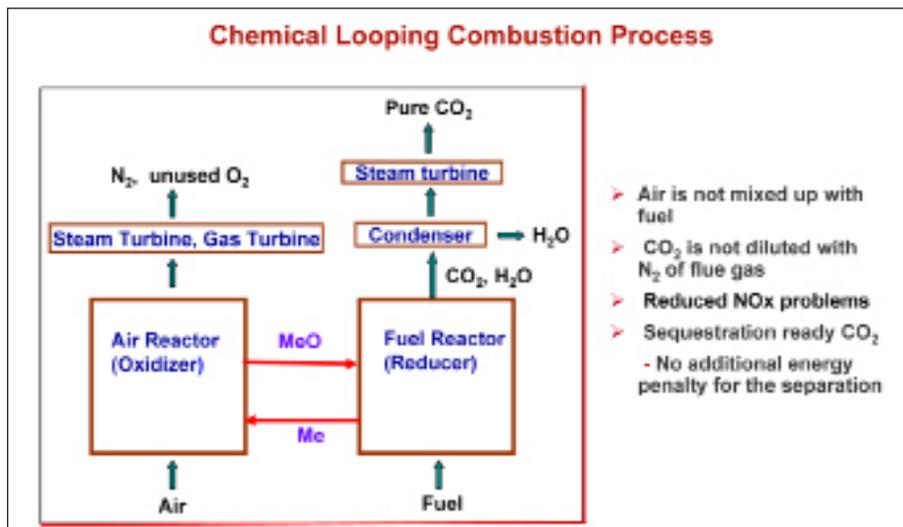
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condensing the steam. This avoids the energy penalty that traditional fossil fuel fired combustors must pay to produce a pure stream of CO<sub>2</sub>. Additionally, CLC avoids production of NO<sub>x</sub> that is produced in almost all other combustion processes.

Development of improved oxygen carriers could increase their mechanical strength leading to increased service life, improve the rate of oxidation and reduction steps, and improve the stability during multi cycle tests. These improvements could significantly improve the economics of the CLC process as an effective carbon management tool in electric power generation from coal.



*Chemical looping combustion of coal with CuO under CO<sub>2</sub> environment  
(a) combustion segment and  
(b) oxidation segment*

