

Study of the behaviour of Cu-based oxygen carriers for understanding the behaviour of the redox kinetics in CLC

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STUDY OF THE BEHAVIOUR OF CU-BASED OXYGEN CARRIERS FOR UNDERSTANDING THE BEHAVIOUR OF THE REDOX KINETICS IN CLC

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Topic: Chemical reaction engineering

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Abstract:

Chemical-looping combustion (CLC) has proven to be a promising combustion technology for power production with integrated CO₂ capture. The selection of a suitable oxygen carrier is one of the most crucial steps in CLC.

In this work, the kinetics of CuO/Al₂O₃ have been measured with reduction oxidation cycles at different temperatures using TGA. For understanding the behavior of the CuO/Al₂O₃ oxygen carrier, different characterization techniques such as N₂ physisorption (BET) and SEM-EDX were used to study the morphological changes of the particles. A significant increase of the pore size and particle porosity with the number of cycles and an exponentially decrease in surface area has been observed as a function of the number of oxidation and reduction cycles. However, after only two redox cycles a cyclic steady state in the redox kinetics has already been obtained. In addition, it was observed with SEM-EDX, that copper agglomerates on the particle surface with increasing number of redox cycles. Therefore, the exact same experiment was carried out with particles of 1.1 mm diameter of CuO/Al₂O₃ and CuO/Al₂O₃ in powder. Both experiments showed the same conversion, confirming that internal mass transfer limitations do not influence the observed kinetics and that the drop in reaction rate is not due to inaccessible copper inside the particles. This is further confirmed by SEM/EDX of cut particles.

XRD analysis was carried out for the CuO/Al₂O₃ oxygen carrier after different numbers of redox cycles, observing that Cu₂O was present in both, oxidized and reduced state, owing to the possibility of oxygen uncoupling during the reduction cycle. An extensive study of different oxygen uncoupling reactions at 800°C was performed.

For all the experiments, TGA, XRD and SEM-EDX analyses were carried out. The experimental results have revealed that there is not influence of the number of redox and CLOU cycles on the oxygen uncoupling conversion.

It has also been observed that the oxygen uncoupling does not affect the structure of the oxygen carrier and that the redox cycles are the responsible for the increase in pore size and particle porosity. With the XRD analysis, the presence of Cu₂O in both, oxidized and reduced state after several number of cycles was observed. This suggest that the Cu₂O cannot be further reduced, thereby influencing the redox kinetics.

After this study, it can be concluded that gas phase diffusion in the pores of the particles does not play a role in the rate of particle conversion and that the oxygen uncoupling conversion is constant independent on previous cycles. It can also be concluded that, even after 100 redox cycles, still Cu₂O is present in reduced and oxidized state, indicating that the Cu₂O cannot be further reduced. Experiments starting from pure CuO and pure Cu₂O are carried out together with a kinetic study in the near future.

Reference 1:

Reference 2 :

Reference 3 :

Reference 4 :

Highlight 1: Gas phase diffusion in the pores does not play a role in the rate of particle conversion

Highlight 2: Cu₂O cannot be further reduced even after 100 redox cycles, influencing the redox kinetics

Highlight 3: Oxygen uncoupling does not affect the structure of the oxygen carrier