

DIRECT SOLAR THERMAL SPLITTING OF ZnO
FOLLOWED BY A QUENCH.
DETERMINATION OF THE RATE CONTROLLING STEPS

by

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Concentrated radiation provided by the sun or artificial sources, can be used to drive endothermic reactions at very high temperatures. These processes include the production of energy carriers or of chemical commodities. Many of them are performed for the chemical storage and/or transportation of solar energy. One of the most often mentioned option relies on the ZnO thermochemical cycle, leading to the production of solar H₂ or Zn. It relies on only two consecutive steps. In the first high temperature one, ZnO is dissociated into O₂ and storable Zn. In the second lower temperature one, Zn reacts with H₂O to produce H₂. The recovered ZnO is finally recycled towards the high temperature section.

The first step includes two parts in series. In the first high temperature solar reactor, the solid ZnO is heated and decomposes. In the second one, the evolved products are cooled down before the storage of zinc. One of the main difficulties to overcome is that the reaction of zinc reoxidation is very fast. The result is that the cooling section must be very efficient and one of the key condition of success is the optimisation of this quench reactor. The aim of the present paper is to report theoretical and experimental results allowing to determine the nature of the elementary rate controlling steps occurring in a quench reactor relying on the dilution of the reacting species with a cold gas and/or on efficient heat exchanges with a cold wall.

The paper describes experiments made in the 5 kW solar furnace simulator of Nancy (France). The process includes both the thermal dissociation of ZnO and the quench of the evolved species. Other experiments are also reported in which the condensation of Zn vapour (produced in an electric furnace) is studied inside a cooled tubular flow reactor, in the presence or not of added O₂ and in steady state conditions.

Microscopic observations show that the partial reoxidation of zinc may occur both on the cold walls and also inside the gas phase. In each case, the reactions probably pass through the intermediate of condensed zinc. Even if the reactions are studied in steady state conditions of flow, the depositions occur in transient, leading to structures that change with the time of reaction. A first simple model is proposed. It includes several chemical steps as well as other physical parameters such as the nature of the flow, the dilution and the heat and mass transfer processes. If these last factors can be approximatively calculated from the general litterature of reactive flows, it is not the case for the chemical steps for which no reliable kinetic data can be founded. The paper reports the first results on the nature of the rate controlling factors and proposes global kinetic expressions. The calculations rely on complete mass balances measured in the whole reactors. They include the mass of reactants and the masses and natures of the

products evolved in different locations of the quench sections (walls and filters intercepting the flow) and for given times of experiments. Finally the results derived from the experiments made from the ZnO dissociation process (solar simulator) and from the condensation of Zn (electric furnace) will be compared.