

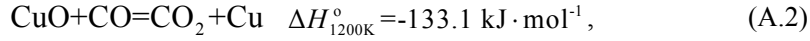
Appendix A: Calculation methods for gases volumes in three stages

The exit gas concentration files detected by the online analyzer were utilized for calculation. In the reduction stage,

$$V'_{\text{CO,R}} = \int_{t-t_{\text{red}}}^t (P_{\text{CO}}(t) \times F_{\text{N}_2}) dt, \quad (\text{A.1})$$

where $V'_{\text{CO,R}}$ is the amount of CO detected in the N_2 sweeping period $((t-t_{\text{red}})-t)$, mL; $P_{\text{CO}}(t)$ is the partial pressure of CO at t , %; F_{N_2} is the flow rate of sweeping N_2 , $500 \text{ mL} \cdot \text{min}^{-1}$.

Moreover, when Fe55Cu5Al5 served as the oxygen carriers, the reaction below also need be considered to calculate $V^*_{\text{CO}_2}$ besides Eqs. (1)–(3).



It was proved by XRD that CuO was completely converted to Cu after the reduction stage.

In the hydrogen stage,

$$V_{\text{H}_2} = \sum_0^t V_{\text{H}_2}(t) = F_{\text{N}_2} \int_0^t \left(\frac{P_{\text{H}_2}(t)}{100 - P_{\text{H}_2}(t) - P_{\text{CO}}(t) - P_{\text{CO}_2}(t)} \right) dt, \quad (\text{A.3})$$

$$V'_{\text{CO,H}} = \sum_0^t V'_{\text{CO,H}}(t) = F_{\text{N}_2} \int_0^t \left(\frac{P_{\text{CO}}(t)}{100 - P_{\text{H}_2}(t) - P_{\text{CO}}(t) - P_{\text{CO}_2}(t)} \right) dt, \quad (\text{A.4})$$

$$V'_{\text{CO}_2,\text{H}} = \sum_0^t V'_{\text{CO}_2,\text{H}}(t) = F_{\text{N}_2} \int_0^t \left(\frac{P_{\text{CO}_2}(t)}{100 - P_{\text{H}_2}(t) - P_{\text{CO}}(t) - P_{\text{CO}_2}(t)} \right) dt, \quad (\text{A.5})$$

where V_{H_2} is the amount of hydrogen produced in the hydrogen stage, mL; $V'_{\text{CO,H}}$ and $V'_{\text{CO}_2,\text{H}}$ are the amount of CO and CO_2 produced in the hydrogen stage, mL; $P_{\text{H}_2}(t)$, $P_{\text{CO}}(t)$ and $P_{\text{CO}_2}(t)$ are the partial pressures of H_2 , CO and CO_2 at t , %; F_{N_2} is the flow rate of N_2 , $1000 \text{ mL} \cdot \text{min}^{-1}$.

In the oxidation stage,

$$V'_{\text{CO,A}} = \sum_0^t V'_{\text{CO,A}}(t) = F_{\text{N}_2} \int_0^t \left(\frac{P_{\text{CO}}(t)}{100 - P_{\text{O}_2}(t) - P_{\text{CO}}(t) - P_{\text{CO}_2}(t)} \right) dt, \quad (\text{A.6})$$

$$V'_{\text{CO}_2,\text{A}} = \sum_0^t V'_{\text{CO}_2,\text{A}}(t) = F_{\text{N}_2} \int_0^t \left(\frac{P_{\text{CO}_2}(t)}{100 - P_{\text{O}_2}(t) - P_{\text{CO}}(t) - P_{\text{CO}_2}(t)} \right) dt, \quad (\text{A.7})$$

where $V'_{\text{CO,A}}$ and $V'_{\text{CO}_2,\text{A}}$ are the amount of CO and CO₂ produced in the air oxidation stage, mL;
 $P_{\text{O}_2}(t)$ is the partial pressures of O₂ at t , %; F_{N_2} is the flow rate of N₂ in the air, 240 mL·min⁻¹.