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,

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

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 mL·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).

$$\text{CuO+CO=CO}_2\text{+Cu} \quad \Delta H^{\circ}_{1200\text{K}}\text{=-133.1 kJ}\cdot\text{mol}^{-1}, \qquad \text{(A.2)}$$

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

In the hydrogen stage,

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

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

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

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 mL·min^{-1}.

In the oxidation stage,

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

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

where $V'_{CO,A}$ and $V'_{CO_2,A}$ are the amount of CO and CO_2 produced in the air oxidation stage, mL; $P_{O_2}(t)$ is the partial pressures of O_2 at t, %; $F_{N_2}$is the flow rate of N_2 in the air, 240 mL·min^{-1}.