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Supplement of

NO_2-initiated multiphase oxidation of SO_2 by O_2 on $CaCO_3$ particles

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S1. Influence of gas phase diffusion on reactive uptake coefficients

The Influence of the gas phase diffusion on reactive uptake coefficients was analyzed using the resistor model described by Davidovits et al. (2006) and the references therein.

$$\frac{1}{\gamma} = \frac{1}{\Gamma_{diff}} + \frac{1}{\alpha} + \frac{1}{\Gamma_{sat}+\Gamma_{rxn}} \quad (1)$$

where Γ_{diff} is the transport coefficient in the gas phase, $1/\Gamma_{diff}$ is the resistance due to the diffusion in the gas phase. Similarly, $1/\Gamma_{sat}$ and $1/\Gamma_{rxn}$ are the resistance due to liquid phase saturation and liquid phase reaction, respectively. α is the mass accommodation coefficient of SO_2.

$1/\Gamma_{diff}$ can be determined using the following equation:

$$\frac{1}{\Gamma_{diff}} = \frac{0.75+0.238\mathrm{Kn}}{\mathrm{Kn}(1+\mathrm{Kn})} \quad . \quad (2)$$

where Kn is Knudsen number. Knudsen number is defined as

$$\mathrm{Kn} = \frac{\lambda}{a} \quad , \quad (3)$$

where λ is the mean free path of a molecule in the gas phase and a is the radius of the particle.

λ can be derived from

$$\lambda = \frac{3D_g}{c}, \quad (4)$$

where D_g is the diffusion coefficient in the gas phase and c is the mean molecular velocity.

c is determined by

$$\mathrm{c} = \sqrt{\frac{8RT}{\pi M}} \quad (5)$$

where R is the gas constant, T is temperature, and M is the molecular mass of SO_2.

$1/\Gamma_{diff}$ was calculated to be 78 and $1/\gamma$ was calculated to be ~8.3×10^4. $1/\Gamma_{diff}$ only accounted for <0.1% of $1/\gamma$. Therefore, the reactive uptake of SO_2 in this study was not limited by gas phase diffusion.

The same conclusion can also be drawn by calculating the gas phase diffusion correction factor for a reactive uptake coefficient according to the method in Pöschl et al. (2007) (Equation 20 in their study, also shown as equation 6 below).

$$\mathrm{C}_g = \frac{1}{1+\gamma\frac{0.75}{Kn}} \quad (6)$$

where C_g is the gas phase diffusion correction factor for a reactive uptake coefficient.

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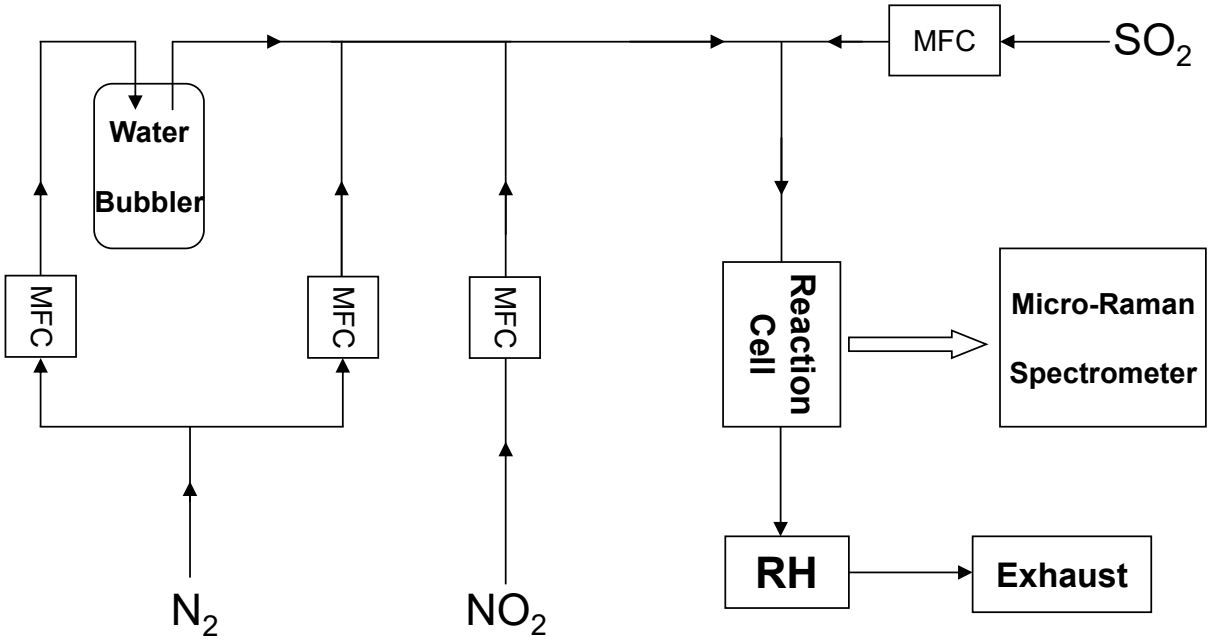

Figure S1. Schematic diagram of the experimental setup (Zhao et al., 2017). MFC: mass flow controller.

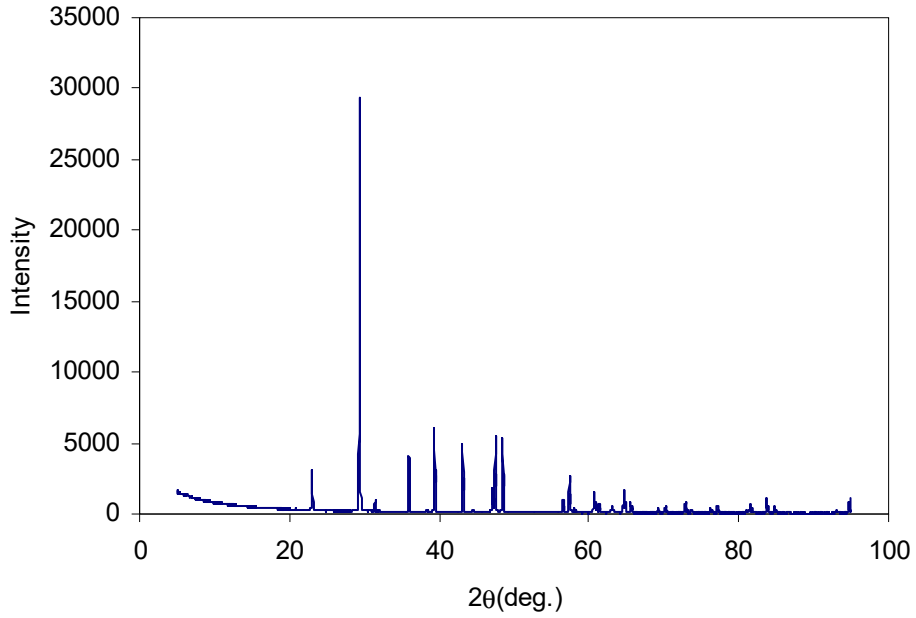

Figure S2. X-ray diffraction spectra of $CaCO_3$ particles.