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Modeling of CO Oxidation Controlled by Oxygen Diffusion in a Spherical Three-way Catalyst Particle

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In this article, isothermal CO oxidation at 225 °C was conducted through Three-way Catalyst (TWC)-particles membrane filter under a superficial velocity of 20 mm/s as shown in Fig. 1 (left). The TWC-particles membrane was composed of agglomerated TWC-particles sizing in the range of 0.8 to 1.6 µm. The agglomerated particles were composed of sintered primary particles sizing about 200 nm. The primary particles sizes were around 30-50 nm as expressed in the cross-section of TWC-particle in Fig. 1 (right). The chemical composition of TWC-particle were much the same as commercial three-way catalyst monolith converters. CO (44 ppm) with N_2 balance was introduced into the membrane filter without any oxygen gas supply. According to the experimental result as described in Fig. 2 (a), the supplied CO (44 ppm) was completely oxidized since there was no CO emission at the beginning of the experiment. However, the attenuation of CO_2 emission was gradually decreased with time while CO was increased accordingly. Considering CO oxidation reaction as a surface chemical reaction occurred at the surface of TWC-particles, a one-dimensional numerical simulation model was constructed. The concept of the simulation model was based on transport of oxygen atoms from the inside of a TWC-particle to the surface through diffusion. Therefore, a one-dimensional radial diffusion in a spherical coordinate was applied by solving the governing equation as shown in Eq. 1. Due to the spherical geometry, symmetric boundary condition was applied at the center of the particle (at $r = 0$) as shown in Eq. 2. The mass flux occurred by the surface chemical reaction was applied at the surface of the particle (at $r = a$) as described in Eq. 3. Surface chemical reaction of the particle was considered as a concentration of one lattice distance of the surface of the spherical TWC-particle. Therefore, surface boundary condition can be expressed as a product of collision frequency (A), surface concentration of the particle ($C_{r=a}$), lattice spacing of nanostructured Ceria (Ce) particle ($d_{Lattice}$), concentration of supplied CO (C_{CO}), and the kernel of exponential function for the temperature dependent chemical reaction. The model was constructed based on single TWC-particle and the concentration of all of the particles from the membrane were assumed as a uniform concentration throughout the reaction. Fig. 2 (b) displays the simulation result of a concentration profile of oxygen atoms in a TWC-particle at 225 degrees C. Considering CO oxidation reaction occurred by the oxygen

$$\frac{\partial C}{\partial t} = D\left[\frac{\partial^2 C}{\partial r^2} + \left\{\frac{2}{r}\ \frac{\partial C}{\partial r}\right\}\right] \quad \textbf{(1)}$$

$$\text{B.C. at r = 0} \Rightarrow \quad \frac{\partial C}{\partial r} = 0 \quad \textbf{(2)}$$

$$\text{B.C. at r = a} \Rightarrow \quad -D\frac{\partial C}{\partial r} = A\, C_{(r=a)}\, d_{Lattice}\, C_{CO}\, e^{\frac{-E_C}{RT}} \quad \textbf{(3)}$$

transport from the inside of the sphere, the oxygen concentration of the particle was radially decreased with time whereas the concentration gradient at the surface becomes greater. Simulation result reveals that the transport of oxygen atom is limited within a narrow range around 100 nm from the surface of the TWC-particle. The concentration profile attained by the simulation was relevant according to the structure of the spherical TWC-particle in which the sintered primary particle sizes were around 200 nm. Simulation result revealed that the oxygen atoms located around 100 nm region from the surface of the spherical TWC-particle might be potentially involved in transport of oxygen atoms required for CO oxidation. The oxygen atoms flux attenuation measured by CO_2 emission and the flux obtained by the numerical simulation were fitted when the diffusion coefficient of oxygen atoms was assumed as 6.26 x10^{-18} m^2/s.

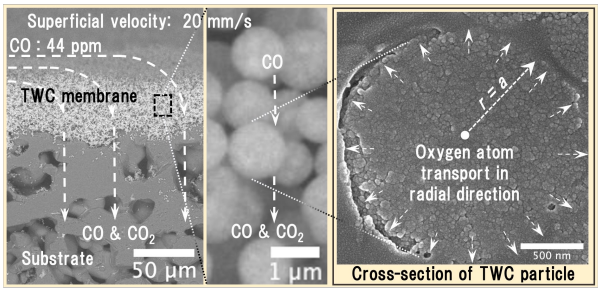

Fig. 1 Oxidation of CO by TWC-particles membrane

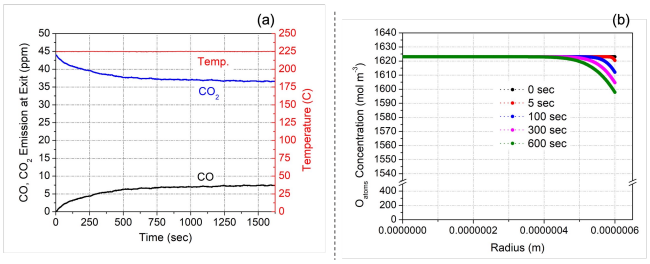

Fig. 2 Emission of CO_2 and CO by CO oxidation under isothermal condition at 225 °C (a) and Concentration profile of oxygen atoms in a TWC-particle (b)