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Size distribution and oxidation rate of carbon nanoparticles

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Introduction

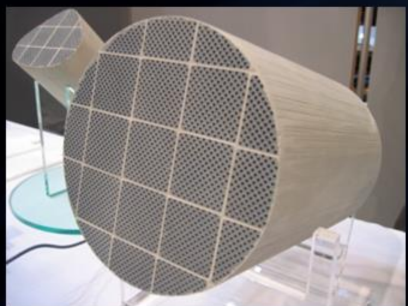
Diesel engine

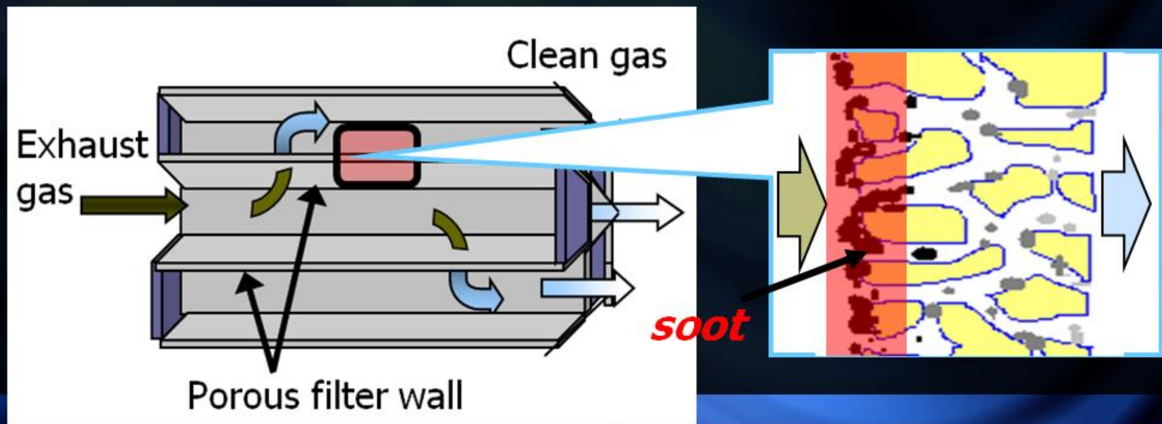
- Advantage of lower fuel consumption
- More particulate matters (PM), human carcinogen
- Stricter exhaust gas emission standards such as Euro VI

Diesel Particulate Filter (DPF)

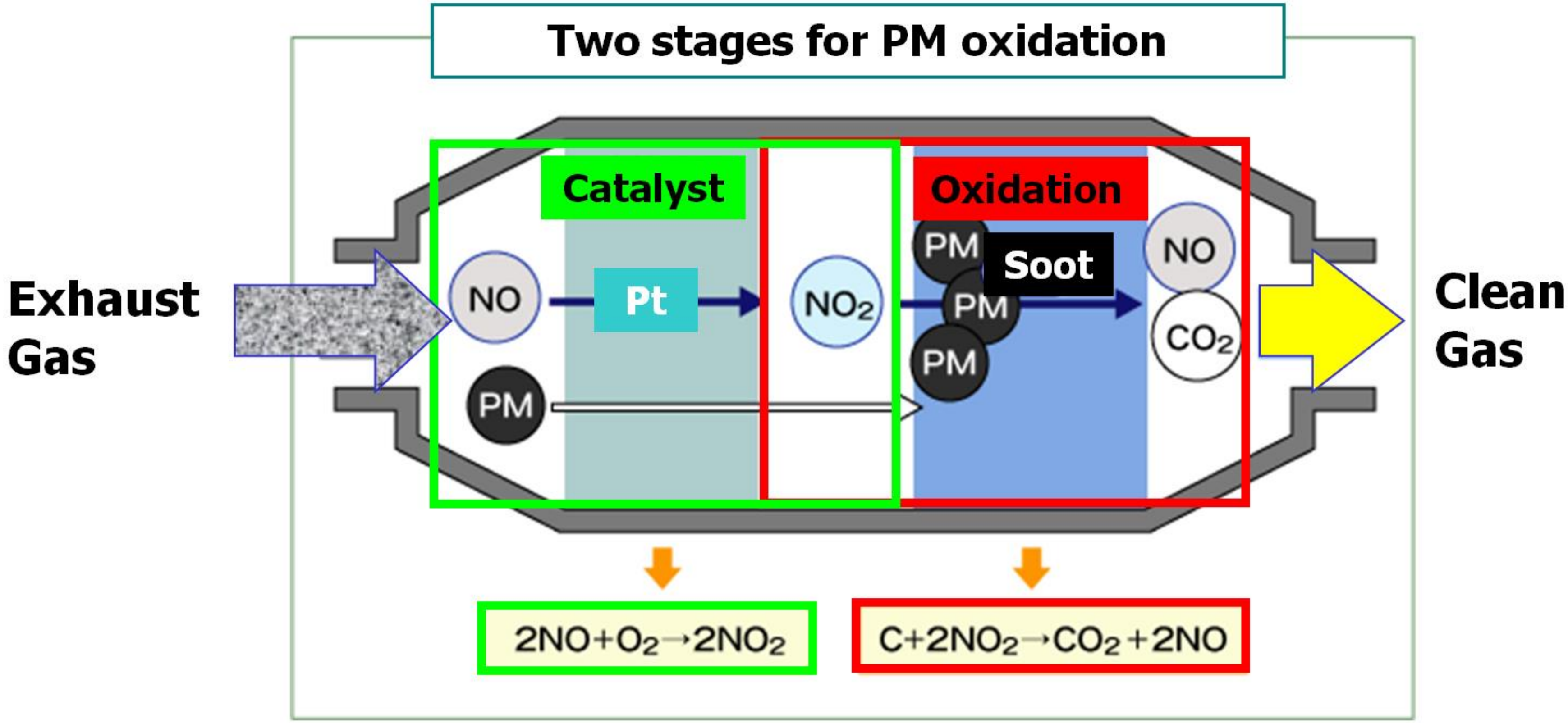
- Wall-flow ceramic filter to trap PM in exhaust after-treatment
- Easily Plugged, need to remove accumulated particles

⇒ Filter regeneration, catalyst to reduce oxidation temp.





PM Oxidation in CRT System



- PM is oxidized by catalyst indirectly
- Compared with O_2, NO_2 is much more reactive for soot oxidation
- Quantitative effect of NO_2 on soot oxidation is not clear

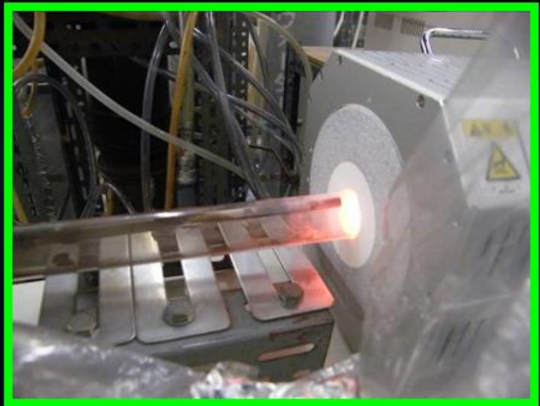
Objective

As for promotion of diesel soot oxidation, few data on particle size distribution and number is available

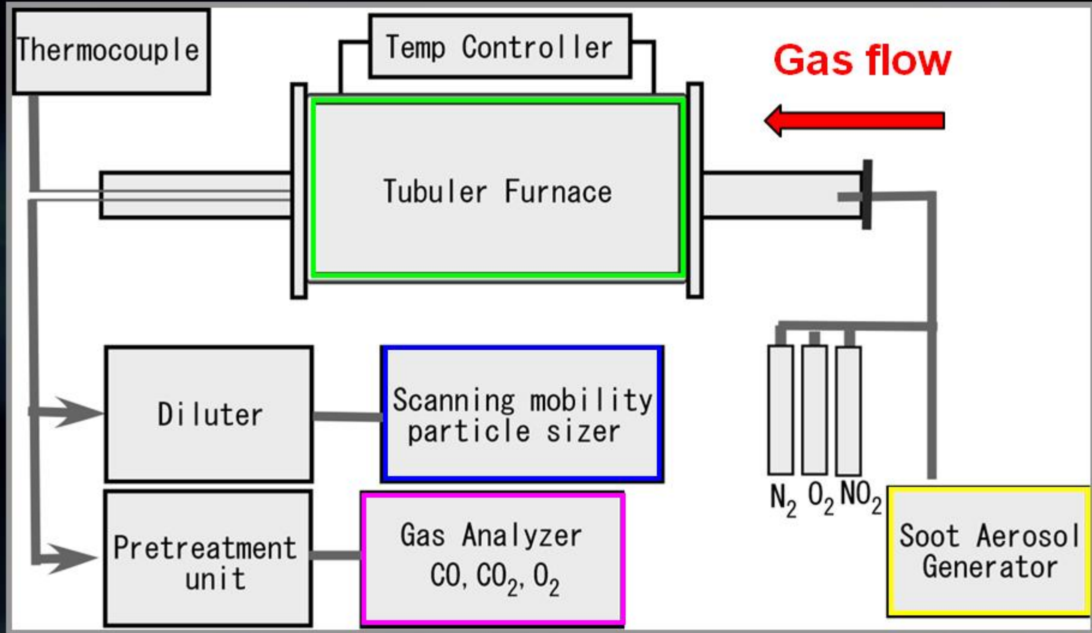
(1) Most of diesel soot is trapped by DPF, and we cannot evaluate variation of soot particle size during oxidation process in CRT system
(2) Characteristics of diesel soot depend on fuel properties, exhaust gas component, engine conditions

By using carbon particle generator, carbon particles are reacted in a temperature-controllable tubular furnace. Oxidation process is analyzed by monitoring particle size and its number concentration.

Experimental Setup



Temperature-controllable tubular furnace



Scanning mobility particle sizer (TSI,SMPS3034)

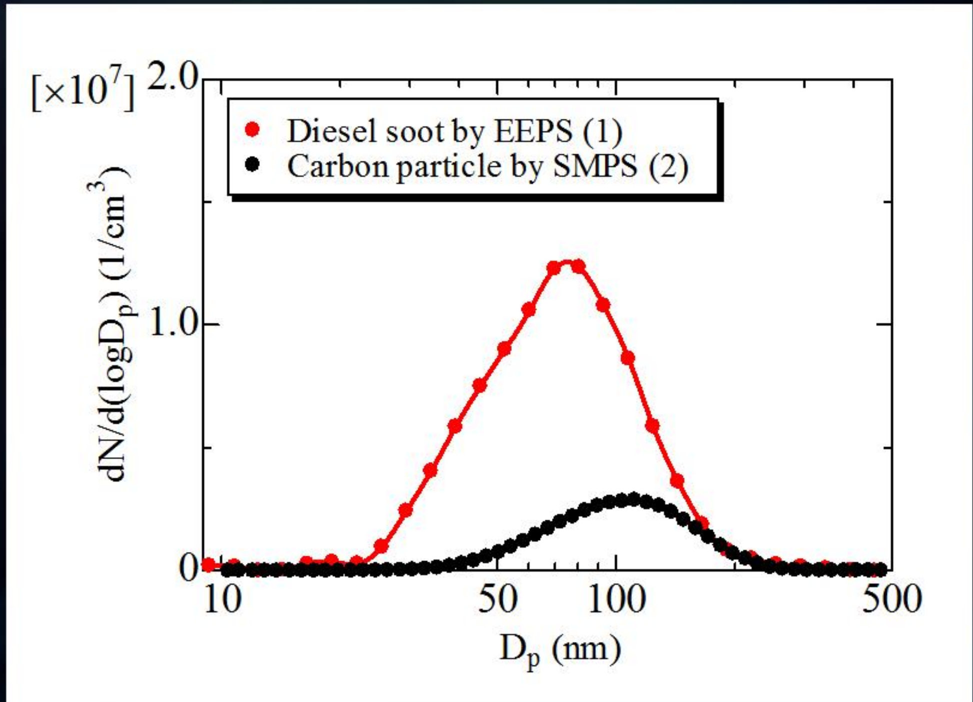
Gas analyzer (CO, CO2, NOx)

Carbon particle generator (Palas, GFG-1000)

Size Distribution of Carbon Particle

(1) Diesel soot by EEPS (SAE Paper 2011-01-0817, 2011)

(2) Carbon particle by SMPS (present study)



- Both distributions has single peak, not double peak
- Size distribution is in the range of 20 nm to 300 nm
- Particle number of diesel soot is slightly larger than that of carbon particle

Experimental Conditions

Experimental conditions

- Temperature：
  200～1100℃

- Gas component:
  Oxygen concentration (X_{O2}): 0～20 %
  NO_2 concentration (X_{NO2}): 0～2000 ppm

Results

① Size distribution of carbon particles

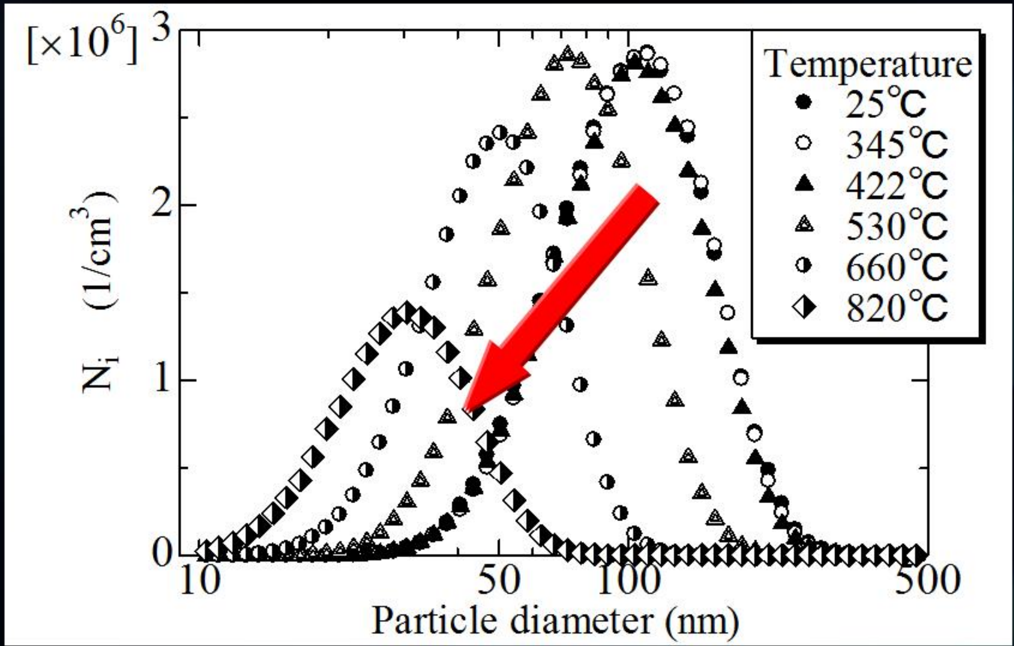
② Particle diameter

③ Total particle number and volume fraction

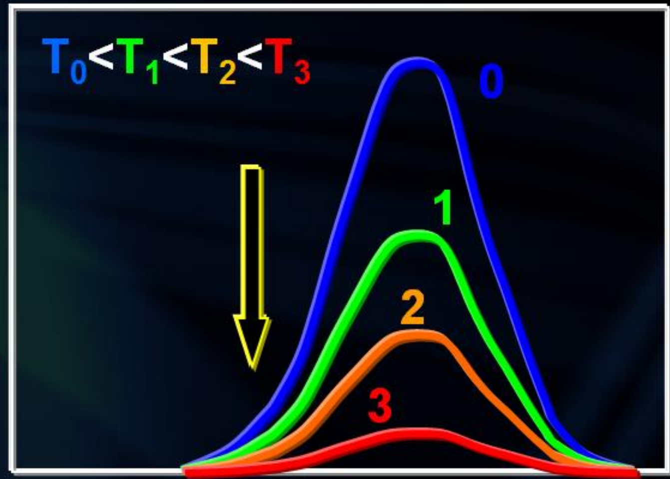
④ Reaction rate constant by Arrhenius plot

①-1 Distribution of Particle Size (w/o NO_2)

NO_2 0 ppm



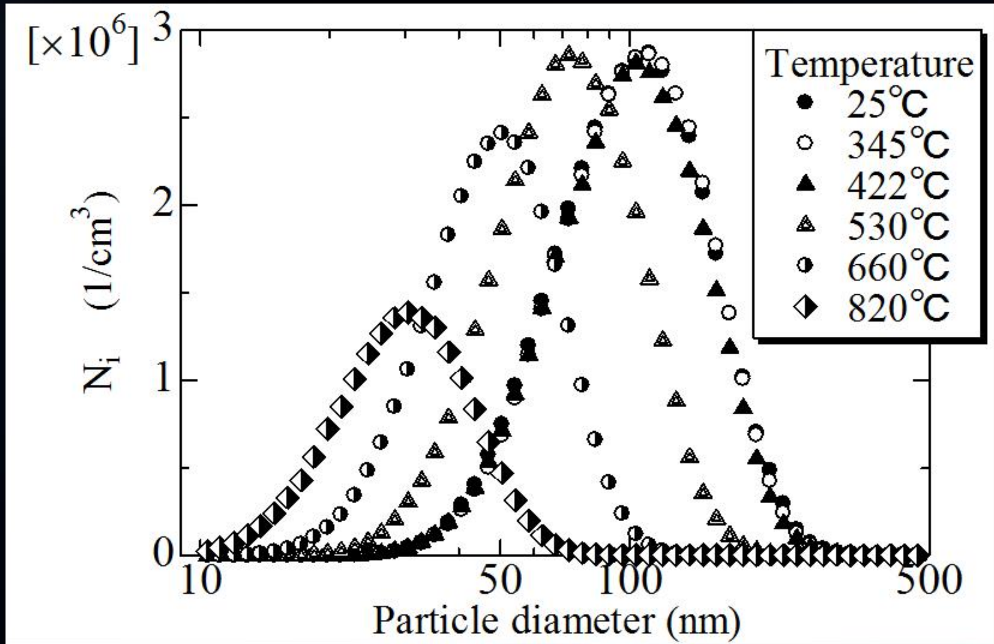
Particle number concentration



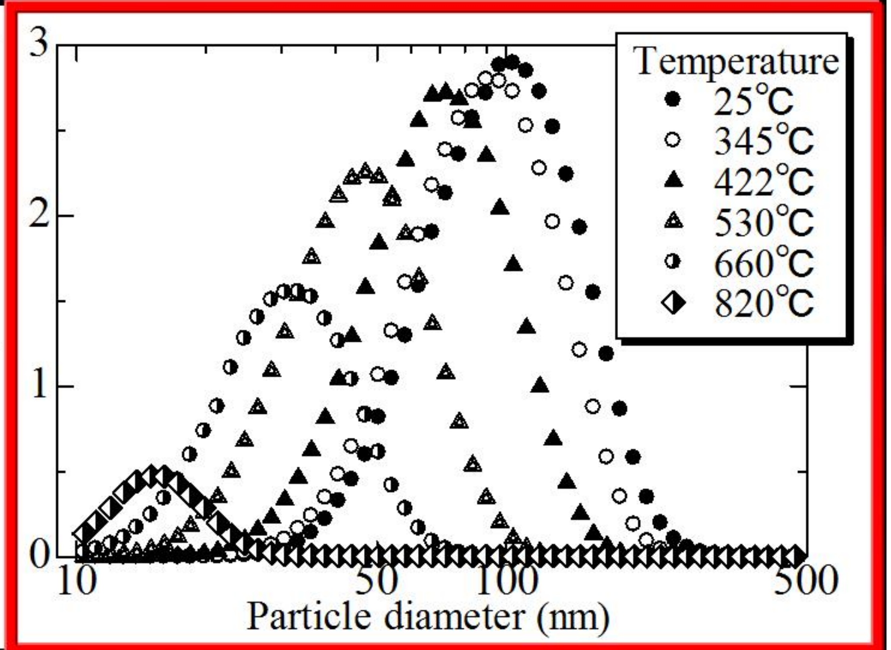
- Little change is observed in size distribution until temperature is 345 ºC.
- At 530 ºC, size distribution is only shifted to smaller particle size. No substantial change in number concentration is confirmed.
- At 820 ºC, size distribution is largely shifted and mode particle size is reduced by 50%, compared to the original value.

①-2 Distribution of Particle Size (with NO_2)

NO_2 0 ppm

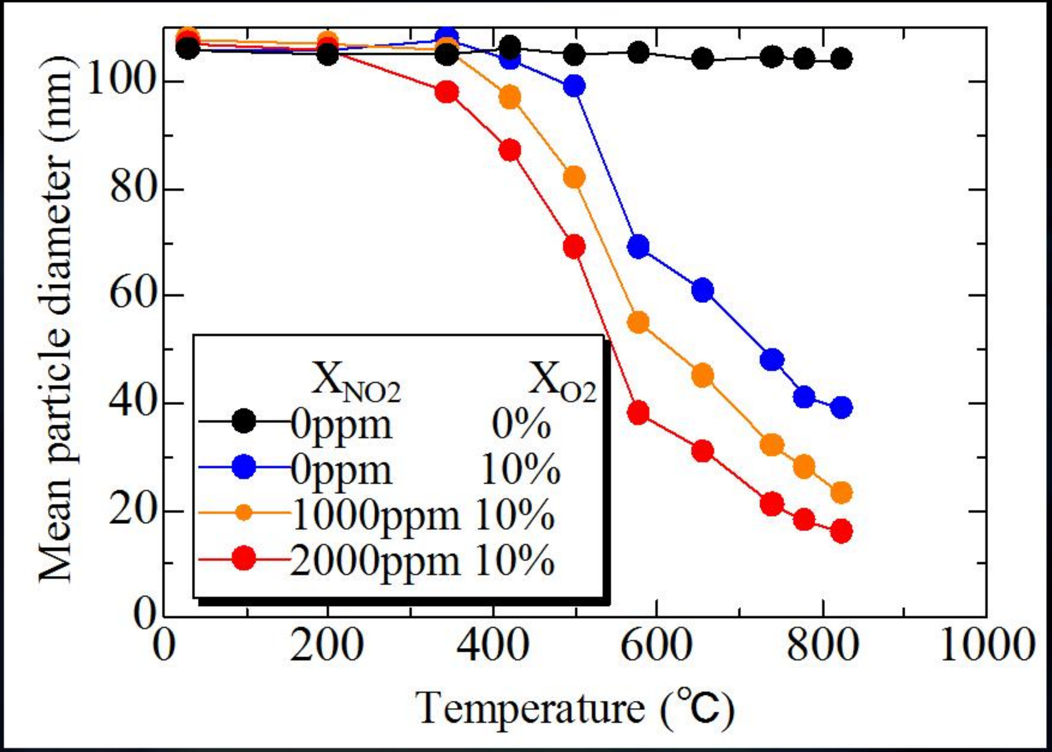


NO_2 2000 ppm



- Even when temperature is 345 ºC, a clear difference from the original size distribution is observed.
- Oxidation temperature is reduced in presence of NO_2.
- At 660 ºC, size distribution is shifted to a greater extent. At 820 ºC, most of particles are oxidized.

② Mean Particle Diameter (D_p)



- For comparison, experiment of no oxygen was conducted. Reasonably, when there is no oxygen, diameter is not changed.
- Original particle diameter is 105 nm. When oxygen is 10 %, D_p starts to decrease around 400 ºC.
- When more NO_2 is added, D_p is smaller.

③-1 Particle Volume Fraction (f_v)

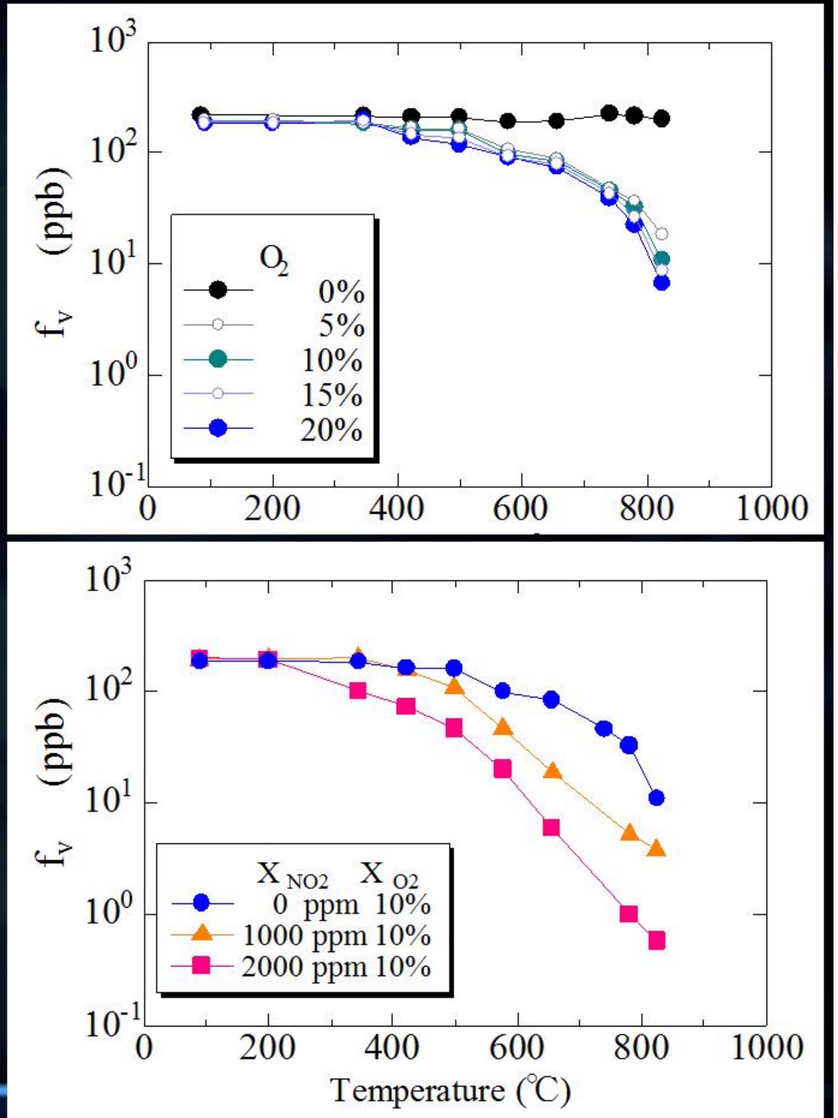
When particles are oxidized, particle size is firstly smaller, with same number

Volume fraction of particles is calculated

$$f_V = \sum_{i=l}^{u} \frac{\pi}{6} D_{p,i}^3 \cdot N_i$$

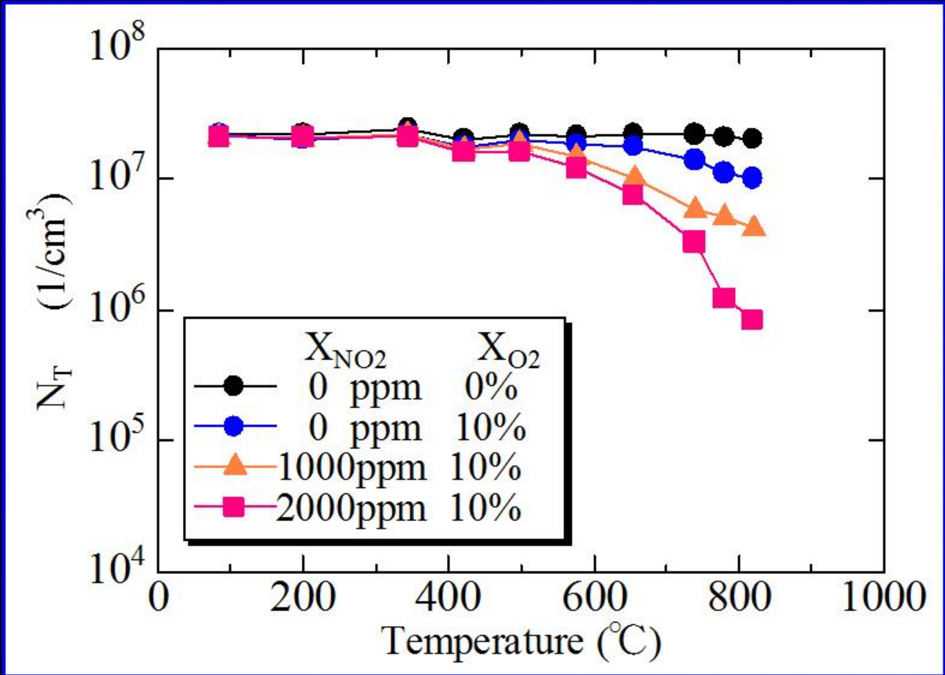
- When X_{O2} is over 5 %, no large difference is observed.
- It implies that, within present conditions, carbon oxidation process would depend only on amount of particle when oxygen is 5 % or more.
- Similarly, f_v decreases as T is increased, but f_v is smaller as more NO_2 is added.

Particle volume fraction, f_v

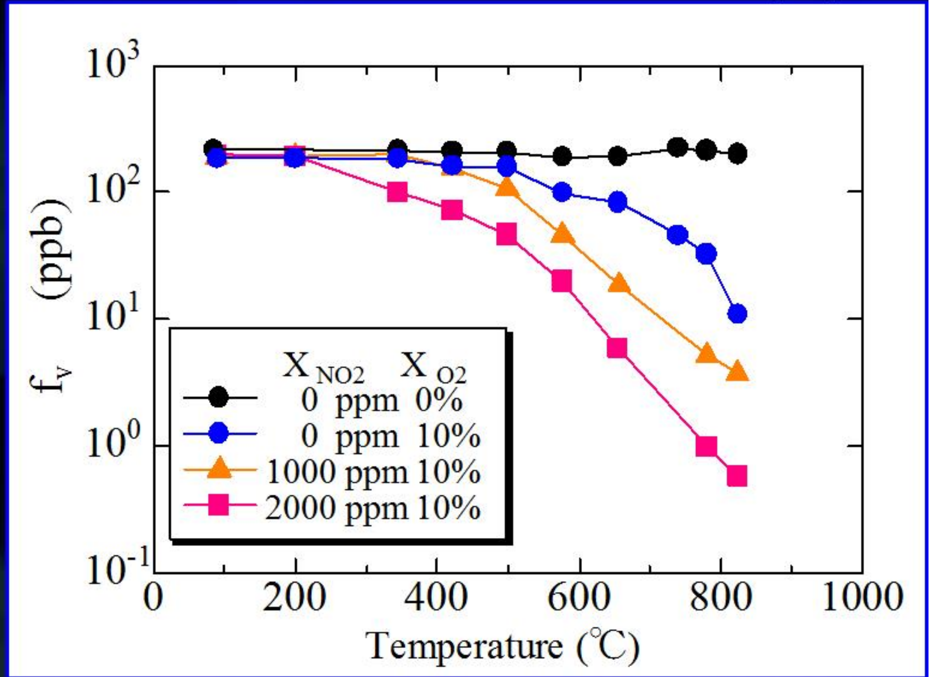


③-2 Total Number Concentration and f_v

Total number of particles, N_T

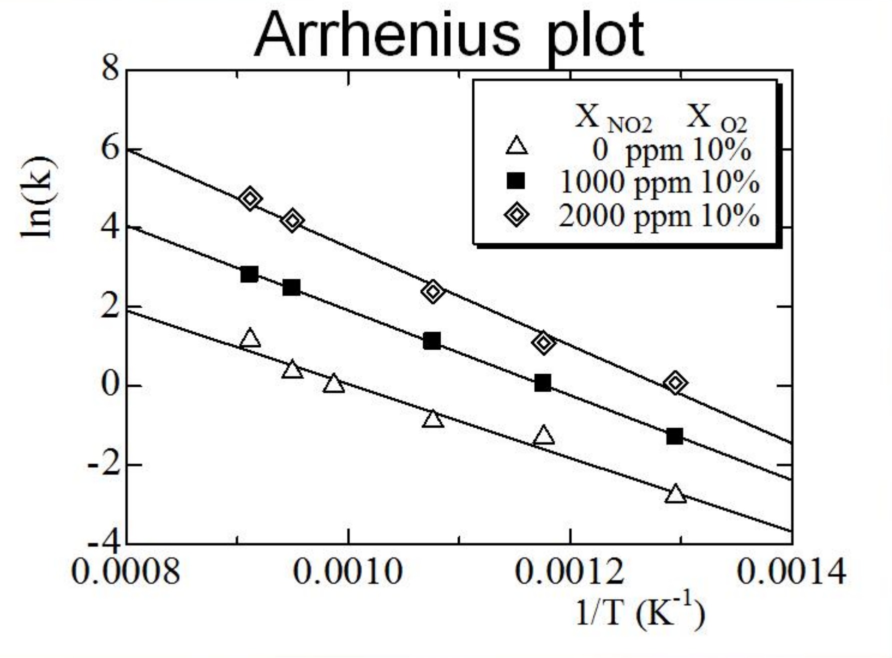


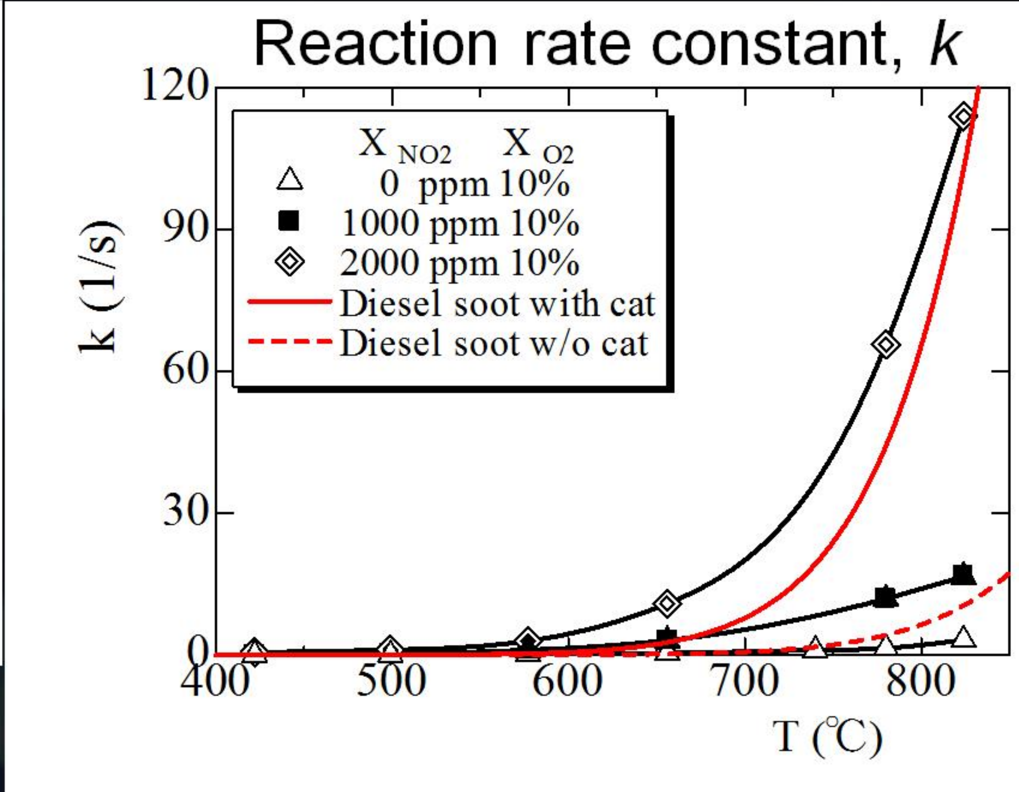
Particle volume fraction, f_v



- Total number of particles (N_T) before oxidation is 2×10^7 1/cm^3.
- N_T decreases as T is increased. In particular, at 820 ºC for X_{NO2} = 2000 ppm, N_T is reduced to 9.6×10^5 1/cm^3 (3 % of original value).
- f_v decreases at a lower T, suggesting that, particle size firstly decreases, and then, particle number decreases due to complete burnout of individual particle.

④ Reaction Rate Constant (k)





- For X_{NO2} = 0 pm, k is close to that of diesel soot without catalyst.

- When NO_2 is added, at temperature below 650 ºC, k for X_{NO2} = 1000 pm is close to the value of diesel soot without catalysis. At high temperature of 700 to 800 ºC, k for X_{NO2} = 2000 pm is close to the value with catalyst.

Summary

We used carbon particles as model soot, and carbon particle size and its number concentration were experimentally measured. To realize oxidation at uniform temperature, a tubular furnace was used. Following results were obtained.

(1) Carbon particle size and number concentration decrease as furnace temperature is increased. When only oxygen is an oxidizer, little change is observed in particle size distribution until temperature is 345 ºC. Carbon particle starts to be oxidized at 420 ºC. Hence, the bulk particle size firstly decreases, and then, the particle number becomes smaller.

(2) When NO_2 is added, oxidation temperature is reduced. Roughly, in the presence of NO_2, carbon oxidation rate is close to the value of diesel soot with catalyst.