17th ETH-Conference on Combustion Generated Nanoparticles
Zurich, Switzerland, June 23-26, 2013

Simulation on Soot Oxidation in Catalyzed DPF

Kazuhiro Yamamoto [*]

Faculty of Engineering, Nagoya University, Nagoya-shi, Aichi 464-8603, Japan
[*] *E-mail address*: kazuhiro@mech.nagoya-u.ac.jp

Introduction

Diesel passenger vehicles have been widely used especially in European countries. Compared with gasoline counterparts, they have good features of high torque at low speed, excellent durability and reliability, higher tolerance to fuel properties, and better fuel efficiency [1]. The main problem with diesel vehicles is that they produce particulate matters (PM) including soot in exhaust gas [2,3]. PM is a serious atmospheric pollutant and has also been linked to carcinogenicity [4], and diesel emissions and control are still very much in the forefront [3,5]. As one of the key technologies, a diesel particulate filter (DPF) for the after-treatment of the exhaust gas has been developed [5-7].

Figure 1 shows a schematic of DPF. Upper figure shows PM trap inside porous filter wall. Typically, the ceramic DPF has a honeycomb structure, with alternate closure of inlet and outlet channels. The mechanism for PM trap is simple: when the exhaust gas passes through the filter wall, PM is trapped, so that the soot cake is formed on the surface of filter substrate, which is orange area in this figure. Then, the filter would be plugged with PM to cause an increase of filter backpressure [8,9]. If the backpressure is high, the engine may stall or the fuel consumption increases. In order to prevent these disadvantages, the filter must be regenerated by oxidizing PM. However, the temperature of diesel exhaust gas is not high enough for soot oxidation [5,6,10]. In addition, DPF may be eroded due to the heat generated in PM oxidation process. Therefore, PM must be oxidized at low temperatures by keeping high filtration efficiency for PM trap.

The system where PM is trapped and oxidized simultaneously is called a continuously regenerating DPF [3,5]. A catalyst, which adheres to the surface of the ceramic filter substrate, is usually used to reduce the PM oxidation temperature. However, the thermal durability of existing platinum catalyst-supported DPF is inadequate, which means that the catalyst may be damaged by PM oxidation and the filter substrate may also be cracked. In addition, since platinum is a rare metal, the amount of catalyst must be suppressed. Then, a full investigation of exhaust gas flow, PM deposition, and PM oxidation in the continuously regenerating DPF is needed. Although a challenging to visualize the processes of PM oxidation and deposition has been conducted experimentally [11, 12], it is still difficult to observe the phenomena inside DPF. So far, we have proposed a numerical scheme for the simulation of DPF by the lattice Boltzmann method (LBM) [13-17].

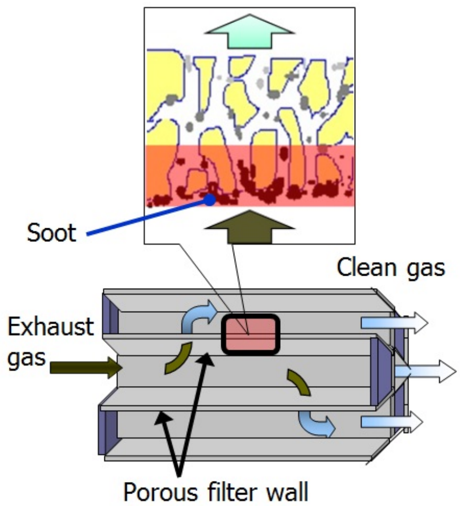


Fig. 1 A schematic of diesel particulate filter is shown. Upper figure shows PM trap inside porous filter wall.

To consider the catalyst, the simplified reaction model with the one-step reaction is applied. The reaction parameters such as activation energy are obtained by an engine test bench. Then, the catalyzed DPF is simulated to examine the effects of the catalyst on diesel soot oxidation. Based on the filter-back pressure, soot deposition and oxidation processes are discussed, which gives useful information to reduce the amount of catalysts.

Numerical Analysis

The lattice Boltzmann method (LBM) used in the simulation is the simplification of Boltzmann equation [18]. The model of D3Q15 using cubic lattices is usually used for 3D calculation [19]. As for the combustion simulation, we followed the same numerical scheme proposed in our previous study [20]. An overall one-step reaction of $C + O_2 \rightarrow CO_2$ was used [13,14,21]. Generally, the soot has complex geometry of nanoparticles [2,22,23]. Since the size of diesel soot has a nano-scale, it is difficult to realize the deposition phenomenon of soot particles precisely. Then, the soot deposition is described by the modified particle deposition model [24]. Different from Lagrangian approach through the equation of motion, individual particles were not considered. Instead, the soot concentration was monitored.

In the numerical simulation, the catalyzed cordierite DPF was considered. We used the internal structure of DPF obtained by the X-ray CT technique. The spatial resolution was 1.15 μm/pix, which was the grid size in the simulation. Figure 2 shows the numerical domain. The filter structure obtained by the X-ray CT is placed at the center of the numerical domain. The width of the numerical domain is W. In the preliminary simulation, W was varied to determine the proper numerical domain. In the present paper, W is 40 μm. The total size is 330 μm (x) × 46 μm (y) × 46 μm (z). The exhaust gas passes through the filter wall in x-direction.

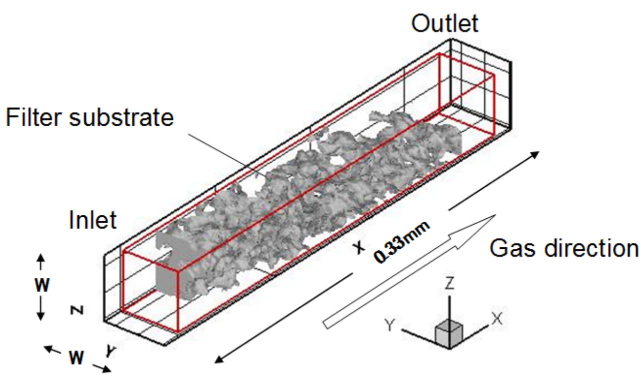


Fig. 2 Coordinate and calculation region are shown. Exhaust gas passes through the filter in x-direction. Gray region corresponds to the filter substrate.

Results and discussion

Firstly, the soot oxidation rate with catalyst was evaluated. The catalyst was uniformly adhered to the surface of the filter substrate. The soot reaction rate was obtained based on data in engine test bench equipped with the catalyzed DPF [9,17]. Table 1 shows the engine specifications. Here, the experimental procedure was explained. Initially, some amount of diesel soot was trapped inside DPF. Then, the temperature was increased for the filter regeneration. By measuring the emissions of CO and CO_2, the mass of reacted soot was calculated. The soot oxidation rate was assumed as the first-order reaction of the soot concentration (soot mass) with the following Arrhenius type equation [14]:

$$-\frac{dm}{dt} = km, \qquad k = A\exp\left(-\frac{E}{RT}\right)$$

where *m* represents the mass of unreacted soot inside DPF, *k* represents a reaction rate constant, A represents a pre-exponential factor, *E* represents activation energy, and *R* represents the gas constant. Based on the Arrhenius plots, the pre-exponential factor was 5.92 × 10^9 1/s, and activation energy was 184 kJ/mol.

Table 1 Engine specifications

Model	NISSAN YD252
Engine Type	Inline 4-cylinder, DOHC 4 valves
Cylinder head port	Tandem port
Displacement	2488 cc
Maximum power	126 kW @ 4000 rpm
Maximum Torque	403 Nm @ 2000 rpm
EGR System	EGR cooler
Turbocharger system	Variable Geometry Turbo with mechanical control

References

[1] A. Yezerets, N. W. Currier, et al., *Applied Catalysis* B 61 (2005) 120-129.
[2] D. B. Kittelson, *Journal of Aerosol Science* 29 (1998), 575-588.
[3] T. V. Johnson, *SAE Technical Paper* 2010-01-0301 (2010) 16-29.
[4] I. M. Kennedy, *Proc. Combust. Inst.* 31 (2007) 2757-2770.
[5] T. Tzamkiozis, et al., *Atmospheric Environment* 44 (2010), 909-916.
[6] G. C. Koltsakis, A. M. Stamatelos, *Prog. Energy Combust. Sci.* 23 (1997) 1-39.
[7] J. Adler, *International Journal of Applied Ceramic Technology* 2 (2005), 429-439.
[8] E. Wirojsakunchai, et al., *SAE Technical Paper* 2007-01-0320 (2007).
[9] K. Tsuneyoshi, et al., *SAE Technical Paper* 2011-01-0817 (2011) 297-305.
[10] H. J. Stein, *Applied Catalysis B* 10 (1996) 9-17.
[11] P. Karin, et al, *SAE Technical Paper* 2009-01-1476 (2009) 725-733.
[12] G. D. Harvel, et al., *SAE Technical Paper* 2011-01-0599 (2011) 147-157.
[13] K. Yamamoto, N. Takada, and M. Misawa, *Proc. Combust. Inst.* 30 (2005) 1509-1515.
[14] K. Yamamoto, et al., *Proc. Combust. Inst.* 32 (2009) 1965-1972.
[15] K. Yamamoto, et al., *the European Physical Journal* 171 (2009) 205-212.
[16] K. Yamamoto, et al., *Catalysis Today* 153 (2010) 118-124.
[17] K. Yamamoto, et al., *Philosophical Transactions A*, The Royal Society, (2011) 2584-2591.
[18] S. Chen, and G. D. Doolen, *Annual Reviews of Fluid Mech*. 30 (1998) 329-364.
[19] Q. Zou, and X. He, *Physics of Fluids* 9 (1997) 1591-1598.
[20] K. Yamamoto, He, X. and G. D. Doolen, *Journal of Statistical Physics* 107 (2002) 367-383.
[21] K. B. Lee , M. W. Thring, and J. M. Beer, *Combust. Flame* 6 (1962) 137-145.
[22] J. Song, M. Alam, A. L. Boehman, and U. Kim, *Combust. Flame* 146 (2006) 589-604.
[23] K. C. Oh, H. D. Shin, *Fuel* 85 (2006) 615-624.
[24] B. Chopard, et al., *Computer Physics Communications* 129 (2000) 167-176.

17th ETH Conference, June 23th – 26th (2013)

Simulation on Soot Oxidation in Catalyzed DPF

Kazuhiro YAMAMOTO
Nagoya University, Japan

Introduction

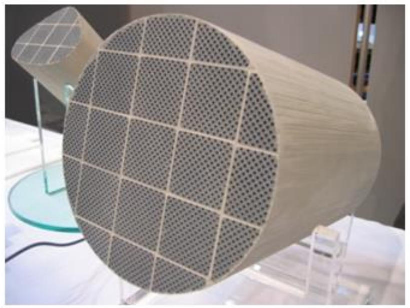
Diesel Engine

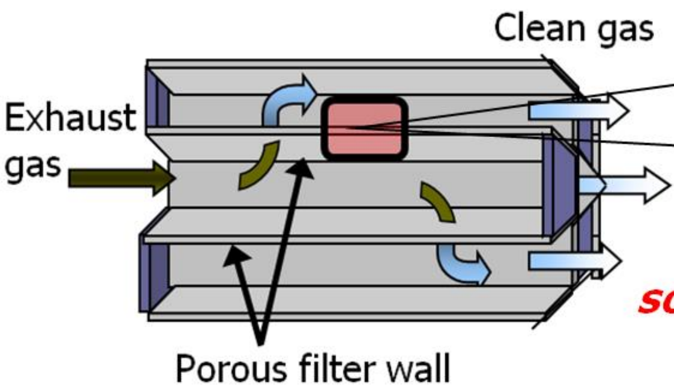
- Advantage of lower fuel consumption
- More particulate matters (PM), human carcinogen
- Stricter exhaust gas emission standards such as Euro V

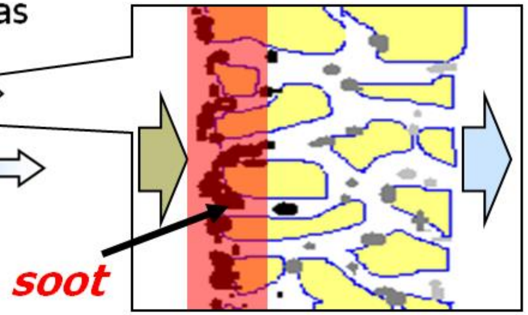
Diesel Particulate Filter (DPF)

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

⇒ Filter regeneration, catalyst to reduce oxidation temperature

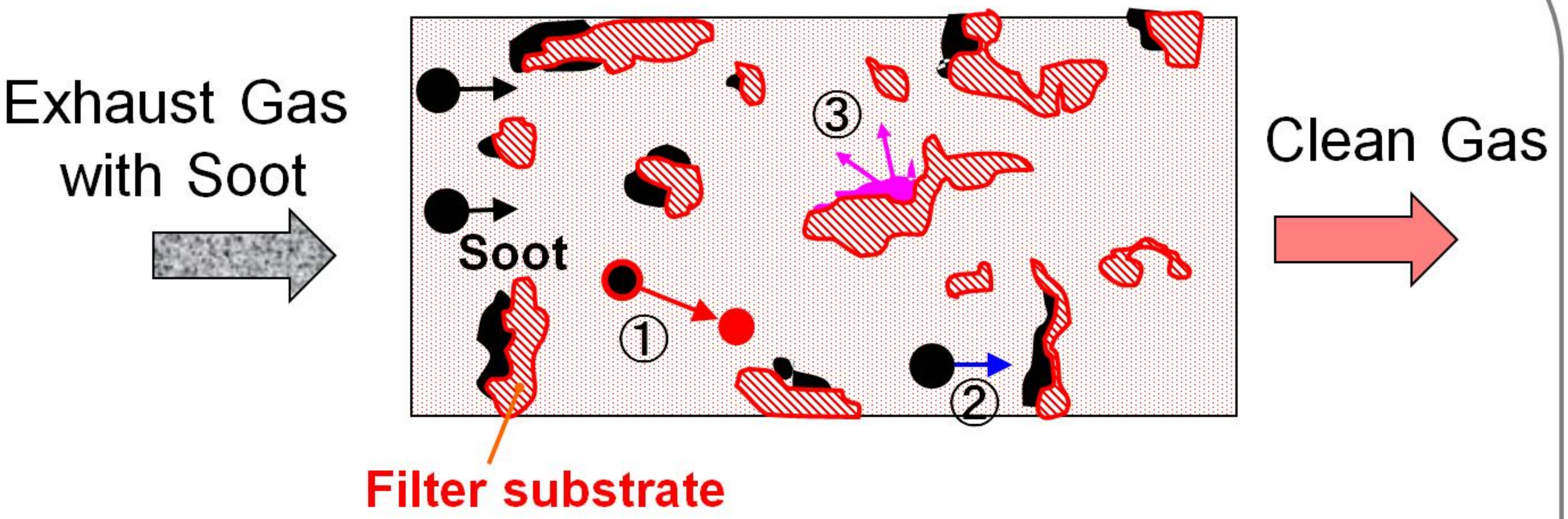






Continuously Regenerating Trap (CRT)

Both soot trap and oxidation occur simultaneously



① **Soot oxidation in gas phase occurs**
② **Soot which is not oxidized in gas phase deposits on filter substrate**
③ **Deposited soot is oxidized by catalyst at filter surface**

Catalyst is expensive, need to reduce amount of catalyst

Objective and Three Approaches

(1) Simulation code
- Lattice Boltzmann method
- Soot deposition and oxidation

CRT system is simulated for the reduction of catalyst

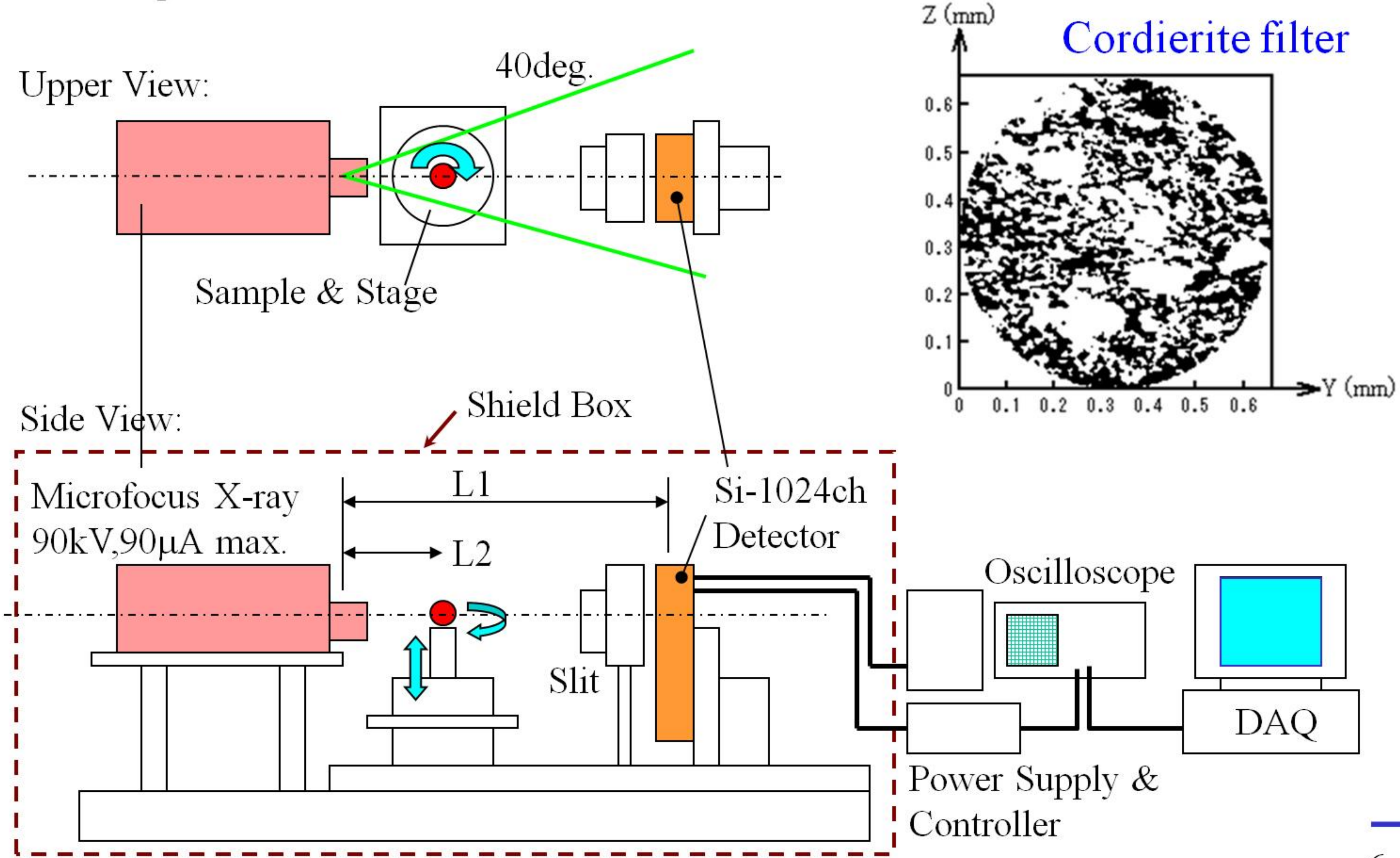
(2) X-ray CT
- Real cordierite filter
- Inner structure

(3) Experiments
- Engine test bench
- Reaction with Pt-catalyst

1. X-ray CT Technique

2. Numerical Method

Experimental Setup for X-ray CT



Lattice Boltzmann Equations

- Flow field (distribution function for P)

$$p_\alpha(\boldsymbol{x}+\boldsymbol{e}_\alpha, t+\delta_t) = p_\alpha(\boldsymbol{x}, t) - \frac{p_\alpha - p_\alpha^{eq}}{\tau}$$

BGK Model

τ: relaxation time related with viscosity

$$p_\alpha^{eq} = w_\alpha \left\{ p + p_0 \left[3\frac{(\boldsymbol{e}_\alpha \cdot \boldsymbol{u})}{c^2} + \frac{9}{2}\frac{(\boldsymbol{e}_\alpha \cdot \boldsymbol{u})^2}{c^4} - \frac{3}{2}\frac{\boldsymbol{u}^2}{c^2} \right] \right\}$$

$$p = \sum_\alpha p_\alpha, \quad \boldsymbol{u} = \frac{\rho_0}{\rho}\frac{1}{p_0}\sum_\alpha \boldsymbol{e}_\alpha p_\alpha, \quad \nu = \frac{2\tau - 1}{6}\frac{\delta_x^2}{\delta_t}$$

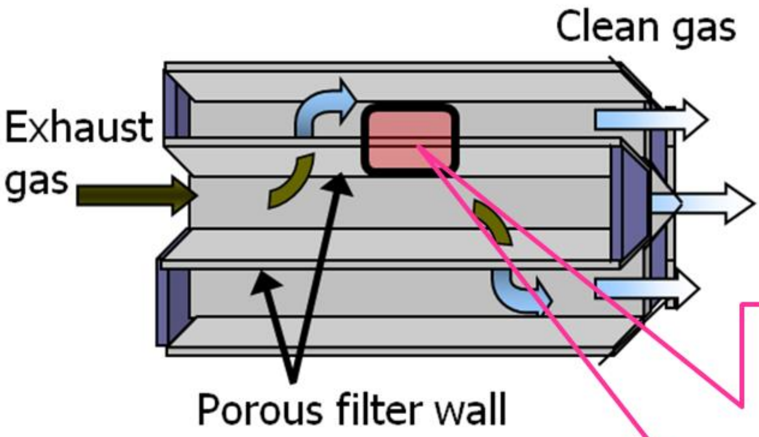
- Scalar Field (distribution functions for Y_i, T)

$$F_{s,\alpha}(\boldsymbol{x}+\boldsymbol{e}_\alpha, t+\delta_t) = F_{s,\alpha}(\boldsymbol{x}, t) - \frac{F_{s,\alpha} - F_{s,\alpha}^{eq}}{\tau_s} + w_\alpha Q_s$$

Source Term

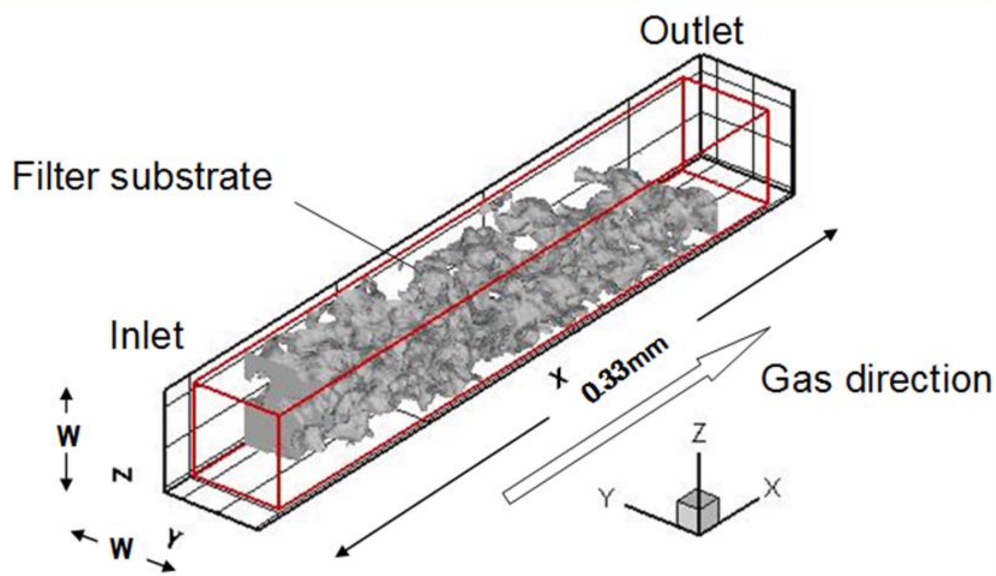
$$F_{s,\alpha}^{eq} = w_\alpha s \left[1 + 3\frac{(\boldsymbol{e}_\alpha \cdot \boldsymbol{u})}{c^2} + \frac{9}{2}\frac{(\boldsymbol{e}_\alpha \cdot \boldsymbol{u})^2}{c^4} - \frac{3}{2}\frac{\boldsymbol{u}^2}{c^2} \right] \qquad s = \sum_\alpha F_{s,\alpha}$$

Numerical Domain



Numerical domain:

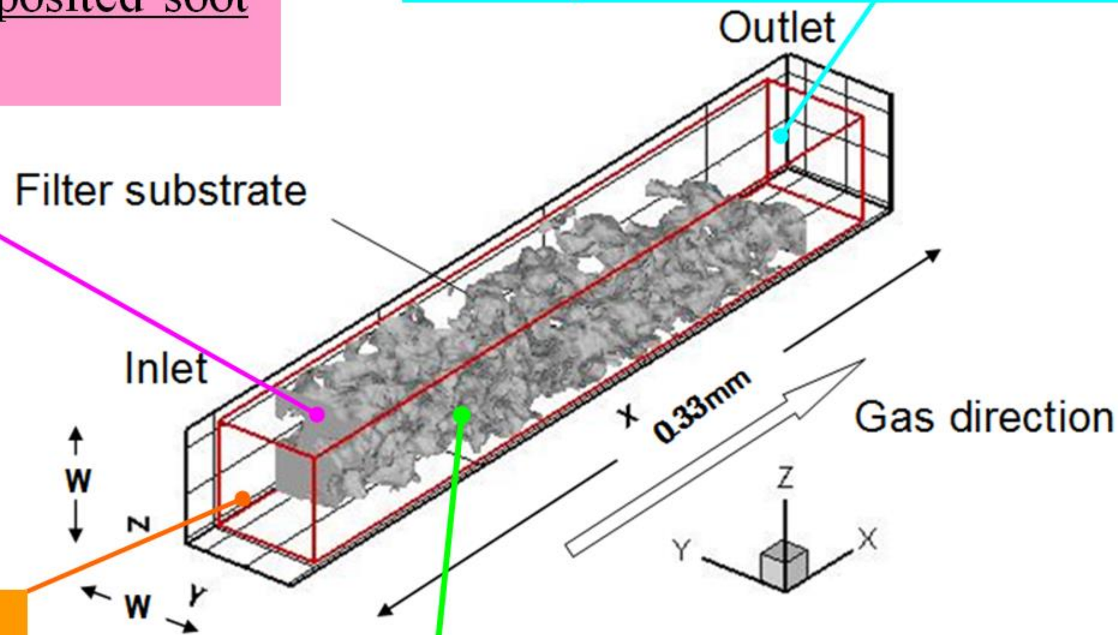
330 μm × W × W (W = 40 ~ 200 μm)
(grid size 1μm = CT resolution)



Boundary Conditions

3. At outlet
- $P = P_{out}$
- Developed boundary condition for temperature, concentration

4. On the filter substrate or deposited soot
Non-slip boundary condition



1. At inlet
- Inflow boundary condition
- Properties of exhaust gas
$U_{in}=0.03$ m/s, $Y_{c,in}=1.6\times10^{-4}$

2. At four side walls
Periodic boundary condition

Reaction Model

First order reaction

$$\frac{dC_{soot}}{dt} = -kC_{soot}, \quad k = A\exp\left(-\frac{E}{RT}\right)$$

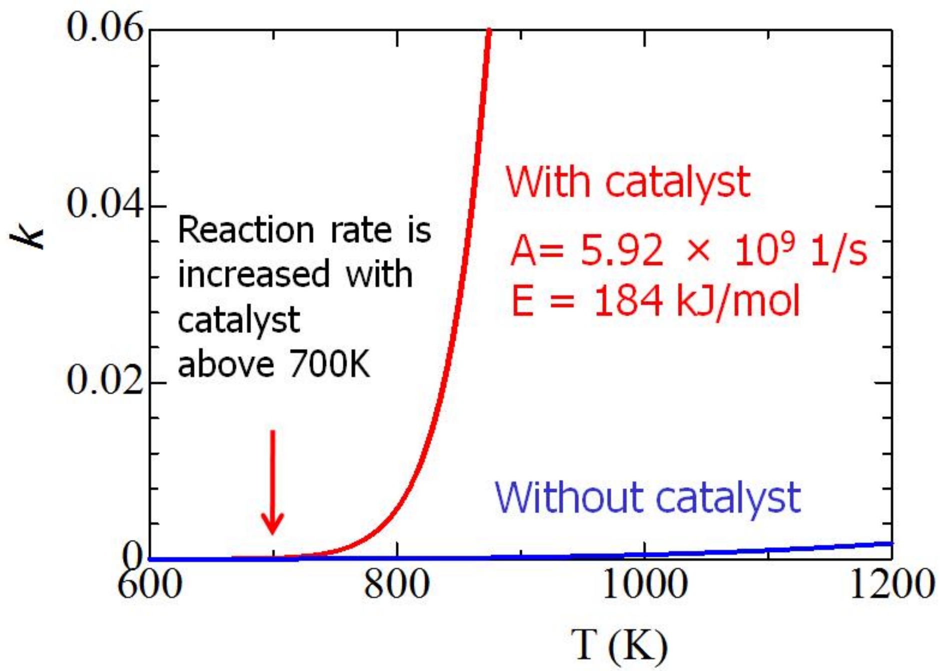


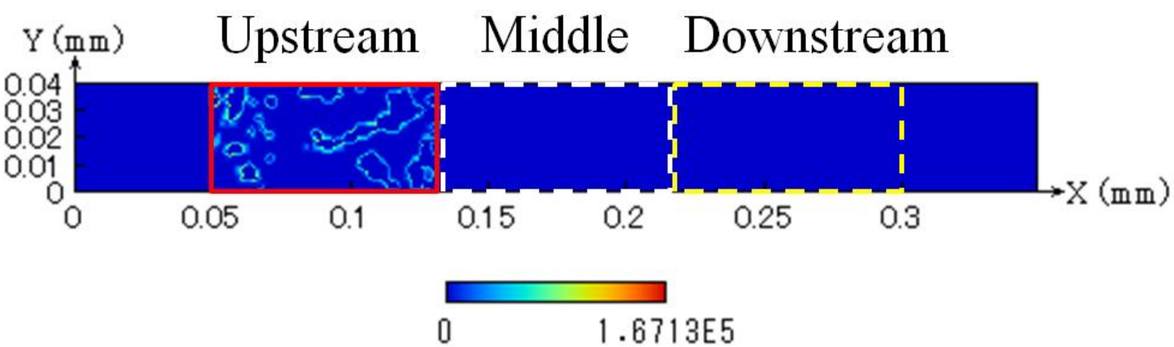
Table 1 Engine specifications

Model	NISSAN YD252
Engine Type	Inline 4-cylinder, DOHC 4 valves
Cylinder head port	Tandem port
Displacement	2488 cc
Maximum power	126 kW @ 4000 rpm
Maximum Torque	403 Nm @ 2000 rpm
EGR System	EGR cooler
Turbocharger system	Variable Geometry Turbo with mechanical control

Results

Simulation in Cases 1-3

Case	Upstream	Middle	Downstream
1	No	No	No
2	Catalyzed	Catalyzed	Catalyzed
3A	Catalyzed	No	No
3B	No	Catalyzed	No
3C	No	No	Catalyzed



Reaction rate of case 3A

Soot in Gas Phase in Case 2

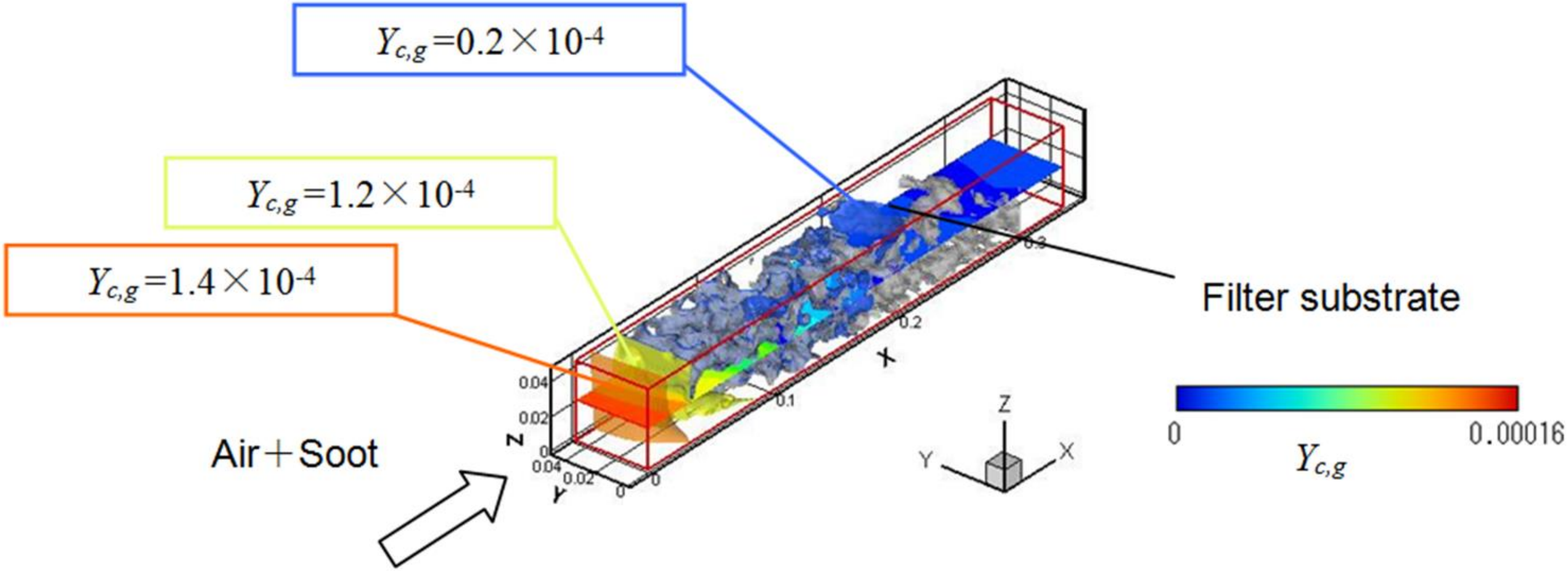
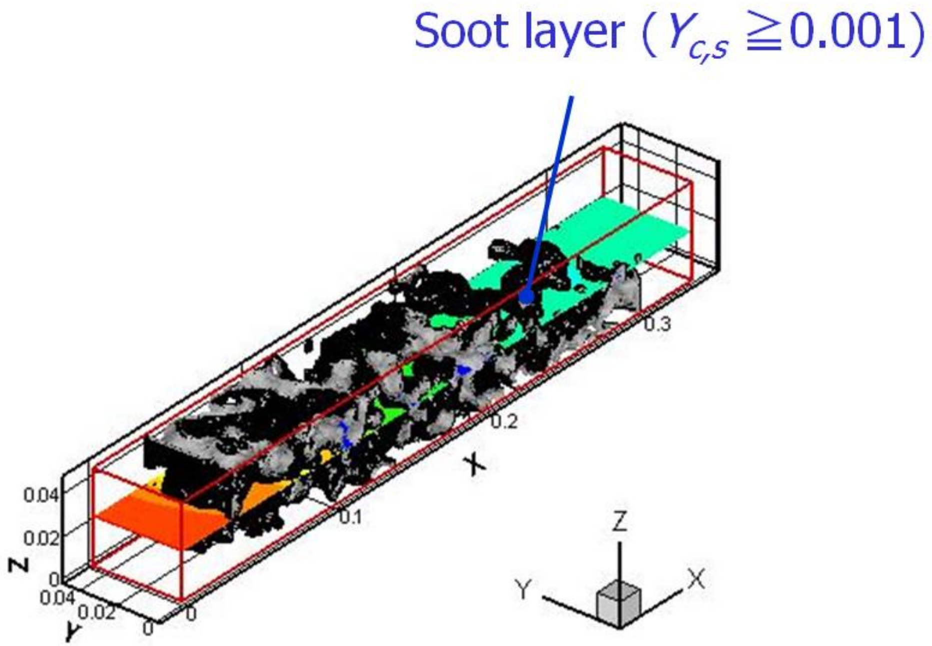
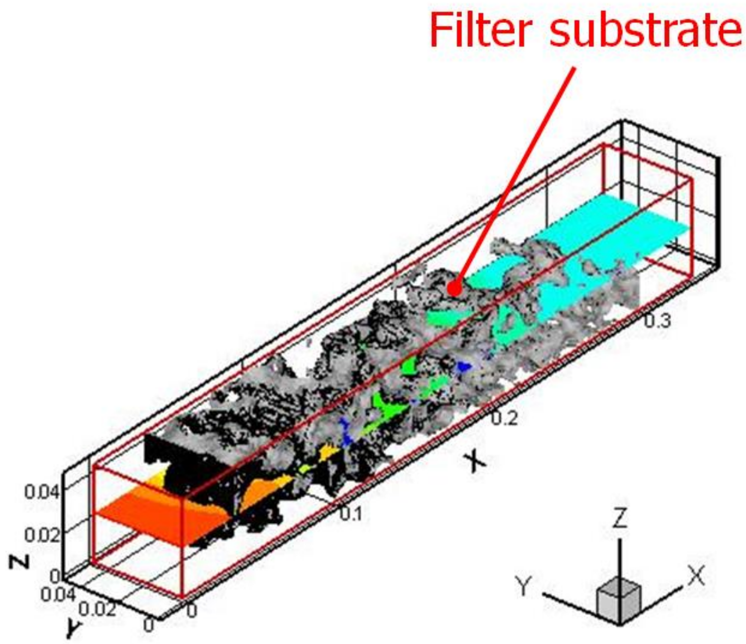


Fig. 3D distribution of soot mass fraction is shown. Three color surfaces correspond to the contour of different soot concentration in gas phase, and gray region corresponds to the filter substrate.

Soot Deposition Region



(a) Case 1



(b) Case 2

Soot Accumulation and Pressure Drop

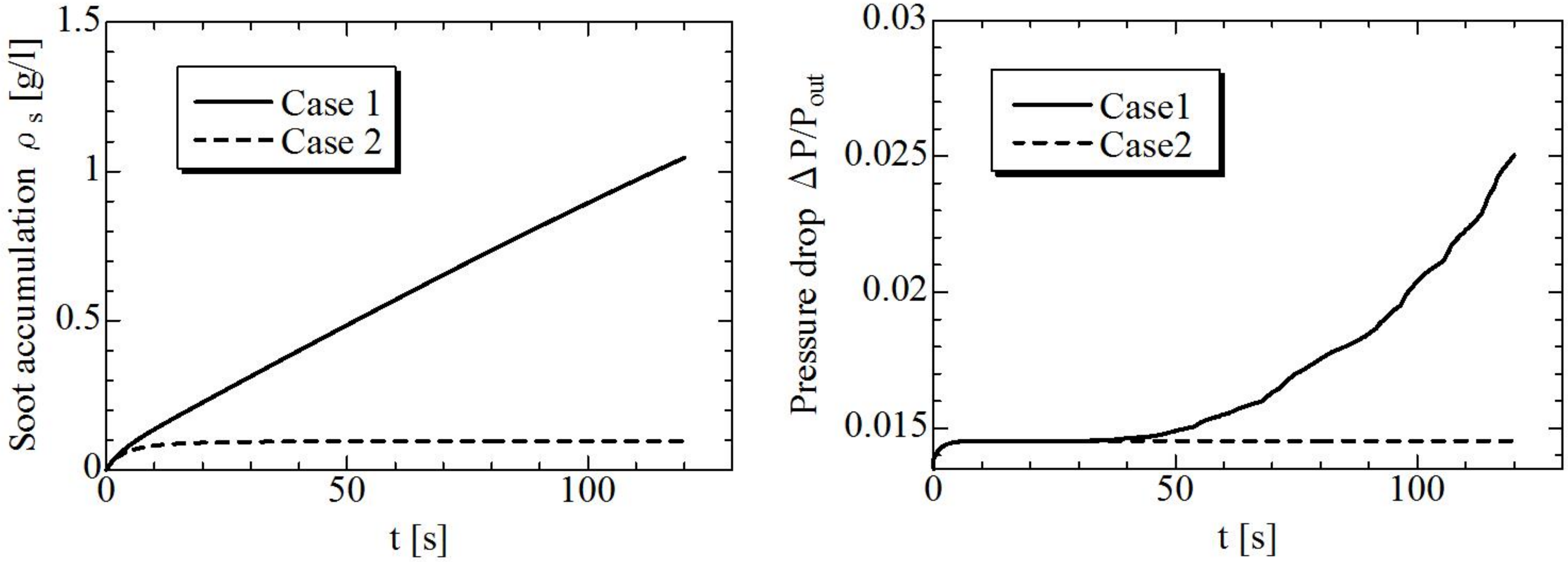
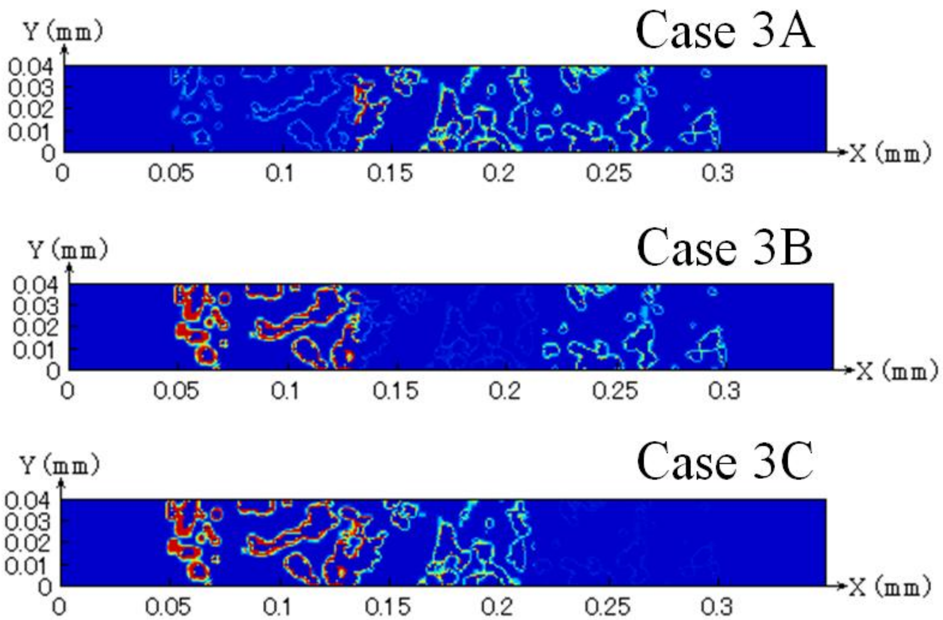
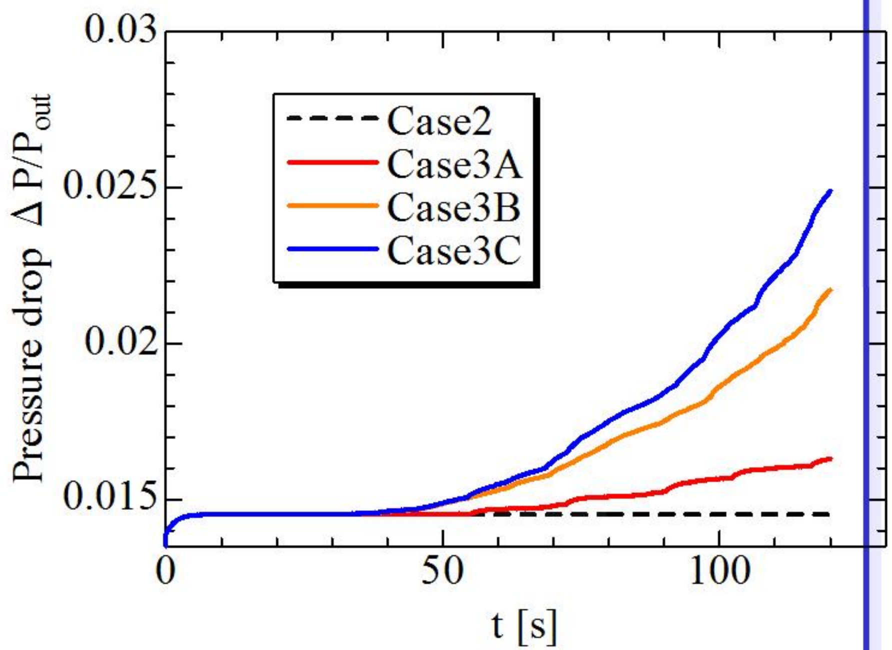


Fig. Mass of soot accumulation and pressure drop across the filter of cases 1 and 2 are shown to observe the effect of catalysts

Soot Layer and Pressure Drop



Region of soot accumulation



Time-variation of pressure drop

Summary

In this study, by the lattice Boltzmann method (LBM), we simulated the soot oxidation in the catalyzed DPF. The simplified model with first order reaction was applied. The reaction parameters such as activation energy were evaluated by using the engine test bench. Based on the mass of deposited soot and filter-back pressure, the effect of catalyst on the soot oxidation was discussed, which gave us useful information to reduce the amount of the catalyst. Numerical simulation is a good tool for the efficient development of DPF technology.

Acknowledgements

This work was partially supported by the Rare Metal Substitute Materials Development Project of New Energy and Industry Technology Development Organization (NEDO) in Japan.