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Technical note

Oxidation characteristics of airborne carbon nanoparticles by NO_2

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ABSTRACT

The oxidation characteristics of airborne carbon nanoparticles were investigated at various temperatures and NO_2 concentrations. Airborne carbon nanoparticles were generated by spark-discharging method using nitrogen as a carrier gas. Monodisperse carbon particles classified using a differential mobility analyzer were introduced into a tube furnace with NO_2 for oxidation reaction. The size distributions of oxidized carbon aerosol particles were measured using a scanning mobility particle spectrometer system which consisted of a differential mobility analyzer and a condensation particle counter. The result was that as NO_2 concentrations and reaction temperatures increased, the surface oxidation rate of carbon aerosol particles increased. For NO_2 gas, the activation energy of the oxidation reaction was approximately 76.3 kJ/mol. The activation energy of the oxidation reaction for the mixture of NO_2 and O_2 gases was 65.9 kJ/mol, which is smaller than that for only NO_2 gas.

1. Introduction

For over a century, diesel engines have proved reliable, economical and durable source of power for many applications. Currently, diesel vehicles are undergoing further development due to their higher fuel efficiency and lower CO_2 emissions as compared to gasoline vehicles because regulations concerning diesel vehicles are becoming more severe due to the harmful effects of diesel exhaust on human health and on the environment (Utell and Frampton, 2000; EPA600/8-90/057F, 2002). The particulate matter (PM) from diesel vehicles consists mostly of carbonaceous particle and a volatile organic fraction (VOF). The PM is often formed during combustion process in locally fuel-rich regions and exhausted in the form of solid agglomerates condensed with VOC (Walker, 2004).

For diesel engine, the lowering of PM emission is generally based on a filtration system in which trapped PM is periodically removed by means of controlled oxidation reaction with pure O_2 or air. Accordingly, many researchers have investigated the oxidation characteristics of PM exhausted from the diesel engine. Higgins et al. (2002, 2003) investigated the oxidation characteristics of size-selected soot nanoparticles in air with the temperature range of 800–1140 °C. However, according to recent research, NO_2 has been found to be a more efficient oxidant than O_2 at lower temperatures in the range of 200–500 °C (Dorai et al., 2000; Ehrburger et al., 2002; Jeguirim et al., 2005; Kamm et al., 2004).

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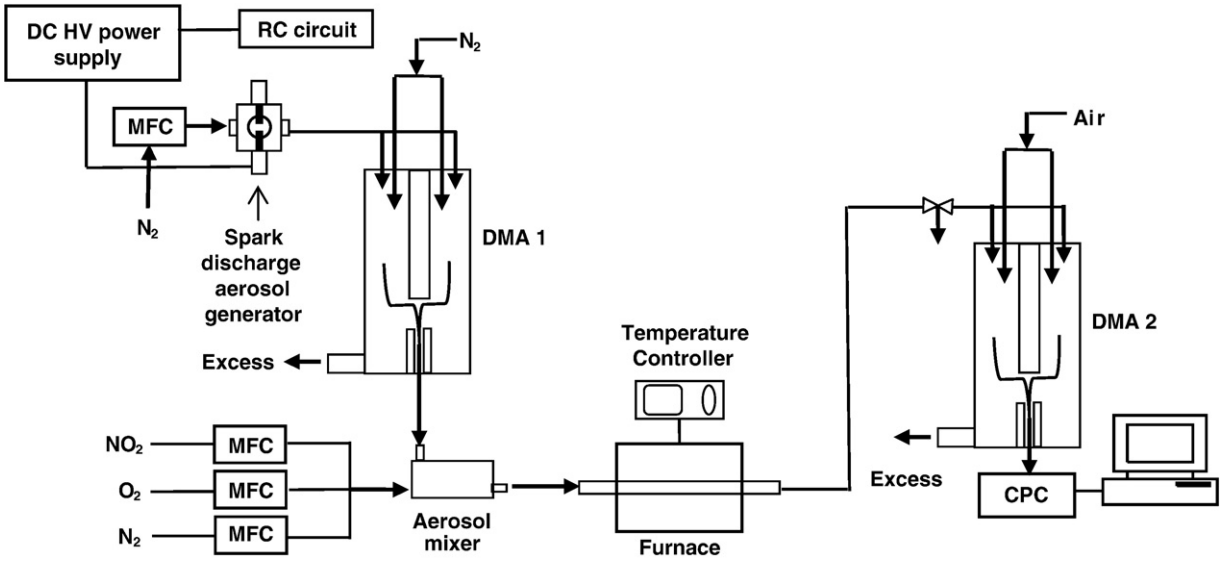

Fig. 1 – Schematic diagram of experimental setup.

Small amounts of NO_2 in the range of a few hundred ppm by volume can promote the continuous oxidation of carbon particulates. In most regenerative diesel particulate filters (DPF) using NO_2, NO_2 is produced by catalytic oxidation reactions or non-thermal plasma reactions of NO, which is previously present in the engine exhaust stream (Dorai et al., 2000; Ehrburger et al., 2002). Jeguirim et al. (2005) studied the oxidation of carbon black deposited on a quartz plate by NO_2 regarding the mass change of carbon black and the concentration of generated CO_2 and CO. Kamm et al. (2004) investigated the gasification of soot deposited on a quartz wool filter by O_3 and NO_2.

Most investigations of carbon oxidation by NO_2 have been on the carbon deposited on filters or plates. However, little is known about the oxidation characteristics of the carbon nanoparticles in aerosol state. In this study, we have focused on the oxidation of airborne carbon nanoparticles which are generated by a spark discharge generator. During the regeneration process of a DPF, the NO_2 can oxidize not only the soot collected on a DPF but also airborne PM emitted from the engine and redispersed from a DPF. This oxidation effect of PM by NO_2 in the airborne state could reduce the total PM emission as a concurring way for mitigation of PM emission.

In our experiment, the oxidation characteristics of nano-sized carbon aerosol particles in NO_2 gas and in the mixture of NO_2 and O_2 gases were investigated at various temperatures. Additionally, the enhancement of carbon oxidation by NO_2 in the presence of O_2 was investigated because in the exhaust stream of diesel engines, a large excess of O_2 is always present compared to NO_2.

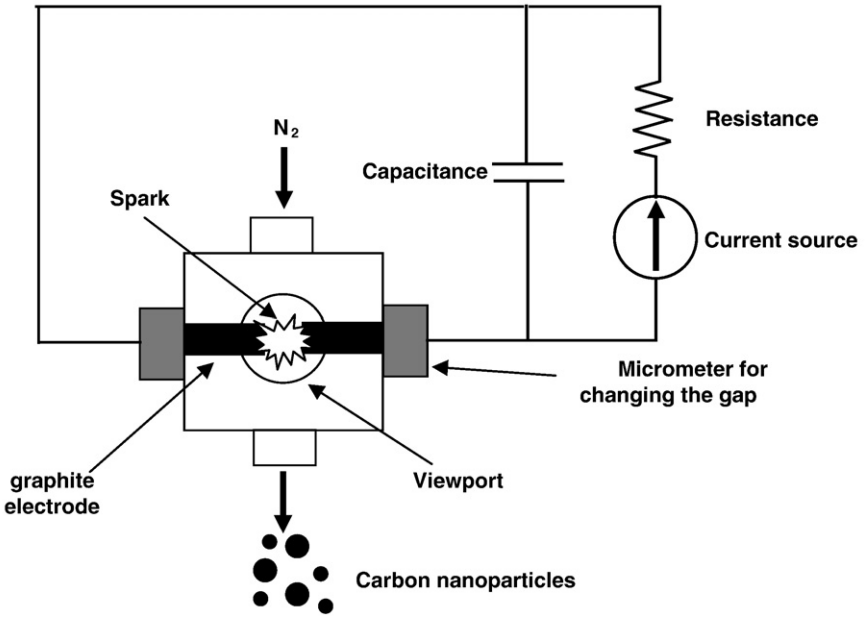

Fig. 2 – Schematic diagram of the spark discharge aerosol generator.