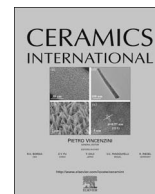




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Short communication

Carbon oxidation characteristics of yttrium manganate catalyst prepared via urea decomposition

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ABSTRACT

Y₂MnO₃ is a hexagonal crystal characterized by high carbon oxidation activity. In this study, carbon black powder has been directly oxidized at temperatures as low as 250 °C with the active oxygen species generated by Y₂MnO₃ catalyst. The activation energies measured for the non-catalyzed and Y₂MnO₃-catalyzed carbon oxidation reactions were 160 kJ mol⁻¹ and 131 kJ mol⁻¹, respectively. During combustion testing of particulate matter in a ceramic form coated with Y₂MnO₃, the captured soot was continuously purified at a temperature of 350 °C.

1. Introduction

Diesel engines exhibit high thermal efficiency and lower levels of CO₂ emission as compared to those of gasoline engines. On the other hand, diesel engines emit large quantities of particulate matter (PM), which are very hazardous for human health. Ceramic forms and diesel particulate filters (DPFs) are considered the most widely used techniques for removing PM from diesel emissions. The temperature of the exhaust gases emitted by the modern exhaust engines is relatively low, well below the ignition temperature of diesel particulates in air. As a result, a large amount of the collected PM does not burn immediately and remains on the surfaces of ceramic forms and DPFs. In particular, driving in urban areas increases the amount of deposited PM with distance, while the accumulated particulates cause significant pressure losses in the engine system and lower its fuel efficiency. Recently, several research groups have studied potential catalysts for the combustion (oxidation) of soot (carbon), which is the principal ingredient of PM [1–6]. In previous works, we reported that RE₂CuO₄ (RE=La, Pr) and La_{0.9}Ag_{0.1}FeO_x compounds exhibited very high carbon oxidation activity [7,8] since the mixed metal oxides containing rare earth ions were found to be effective combustion catalysts. In addition, various transition metal oxides (including MnO₂) demonstrated superior carbon oxidation performance [9]. It was reported that the presence of MnO_x–CeO₂ mixed oxide and La–K–Mn–O perovskite oxide catalysts decreased the combustion tempera-

ture of carbon black/soot [10,11]. If the ignition temperature of captured PM can be significantly lowered by coating catalyst species on the ceramic form and DPF surfaces, it will promote the carbon oxidation reaction and thus improve the fuel efficiency of the engine.

In the present study, the use of perovskite (ABO₃) catalysts for PM oxidation has been investigated. Perovskite oxides are good candidate catalysts for diesel soot combustion because of their good redox properties, thermochemical stability, and tunable catalytic performance [12]. The oxidation activity of perovskite oxide catalysts is comparable to that of noble metal catalysts under suitable conditions. Fino et al. have related the reactivity of perovskite oxides during soot combustion with the abundance of α-oxygen (O⁻) species that are weakly chemisorbed on the perovskite oxide surface and thus are able to enrich the surface of soot particles with oxygenated species [1,13]. According to the results of preliminary testing, the carbon oxidation activity of ABO₃ oxides increased remarkably when their B sites were occupied with Mn ions. On the other hand, rare earth (RE) elements were considered for the A sites of ABO₃ oxides. A series of screening tests were conducted to optimize the composition of REMnO₃ with respect to the carbon combustion efficiency. When REMnO₃ compounds (RE=La, Nd, Sm, Gd, and Dy) were prepared by a urea decomposition method, their crystal structures were orthorhombic (space group *Pbnm*), while those obtained for RE=Ho, Y, Er, and Yb were hexagonal (space group *P63cm*) [14,15]. The carbon oxidation activity of hexagonal YMnO₃, ErMnO₃, and YbMnO₃ catalysts was

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relatively high, while the catalytic activity of the oxides from the orthorhombic group was lower. In this work, we investigated the oxygen release and soot (carbon) oxidation characteristics of hexagonal YMnO_3 , which exhibited the highest carbon oxidation activity among various REMnO_3 compounds.

2. Experimental

YMnO_3 was prepared by the urea decomposition method. $\text{Y}(\text{NO}_3)_3$, $\text{Mn}(\text{NO}_3)_2$, and $\text{CO}(\text{NH}_4)_2$ were selected as starting materials, which were mixed at a molar ratio of 1:1:4. The mixture was calcined at 350 °C for 1 h and then milled inside a ball mill (Fritsch Co., Pulverisette 6). The obtained powder was thermally treated at 1000 °C for 1 h. The specific surface area of YMnO_3 determined using a three-point Brunauer–Emmett–Teller method (Quantachrome Co., NOVA3200) was equal to 4.0 $\text{m}^2 \text{g}^{-1}$.

The carbon oxidation (combustion) activity of the prepared YMnO_3 catalyst was examined by heating its mixture (10 mg) with carbon black from room temperature to 700 °C at a heating rate of 10 °C min^{-1} and air flow of 20 mL min^{-1} inside a thermo-gravimetric differential thermal analyzer (TG–DTA, Rigaku Co., TG8120). Carbon black (Tokai Carbon Co., Ltd., #8500/F, specific surface area: 290 $\text{m}^2 \text{g}^{-1}$) was used for the evaluation of the carbon oxidation activity of YMnO_3 instead of soot. YMnO_3 and carbon black were thoroughly mixed at a weight ratio of 19:1. The apparent activation energy of the carbon oxidation process was determined by the Ozawa method [16].

Next, ^{18}O species were introduced into the YMnO_3 lattice by treating YMnO_3 with $^{18}\text{O}_2$ gas (99% purity) at a temperature of 400 °C for 1 h in order to investigate the contribution of the lattice YMnO_3 oxygens to the oxidation (combustion) of carbon black. The obtained ^{18}O -substituted YMnO_3 sample was heated from room temperature to 800 °C at a heating rate of 10 °C min^{-1} in a mixed stream of Ar (79 vol%) and $^{16}\text{O}_2$ (21 vol%) gases. The generated $^{16}\text{O}^{18}\text{O}$ molecules (molecular weight: 34) were detected using a gas chromatography/mass spectrometry (GC/MS) system (Clarus 560 GC/MS, PerkinElmer Co., Ltd.). $^{12}\text{C}^{18}\text{O}$ (molecular weight: 30) species generated by the mixture of carbon black with ^{18}O -substituted YMnO_3 were also detected under the same experimental conditions.

In situ X-ray diffraction (XRD) analysis of the YMnO_3 sample heated to different temperatures between 30 and 700 °C was performed using a Rigaku Ultima IV diffractometer with $\text{CuK}\alpha$ radiation in the 2θ range of 10–90°.

3. Results and discussion

Fig. 1(a) shows the TG–DTA curves recorded for the YMnO_3 /carbon black mixture, which exhibited gradual weight decrease during heating. An abrupt weight loss was observed between 300 and 450 °C due to the combustion of carbon black, as indicated by the exothermic DTA peak with a maximum at 395 °C. On the other hand, Fig. 1(b) shows the TG–DTA curves obtained for carbon black; the corresponding DTA peak temperature was equal to 665 °C. Hence, the DTA peak temperature of carbon black drastically decreased after the addition of YMnO_3 catalyst.

The apparent activation energy of the carbon oxidation process was determined by the Ozawa method using the following expression:

$$\{d \log(\Phi)/d(1/T_x)\} = 0.4567(E_a/R)$$

where Φ is the heating rate, T_x is the temperature corresponding to the conversion of $x\%$ of the original carbon amount, and E_a is the apparent activation energy [16]. The magnitude of E_a can be estimated from the slope of the straight line fit obtained for the plot of $\log(\Phi)$ versus $1/T_x$ using the least squares method. In this work, the related E_a measurements were conducted by heating 10 mg of the studied mixture from room temperature to 700 °C at heating rates of 2, 5, 10, and 20 °C min^{-1} and air flow of 200 mL min^{-1} followed by the construction

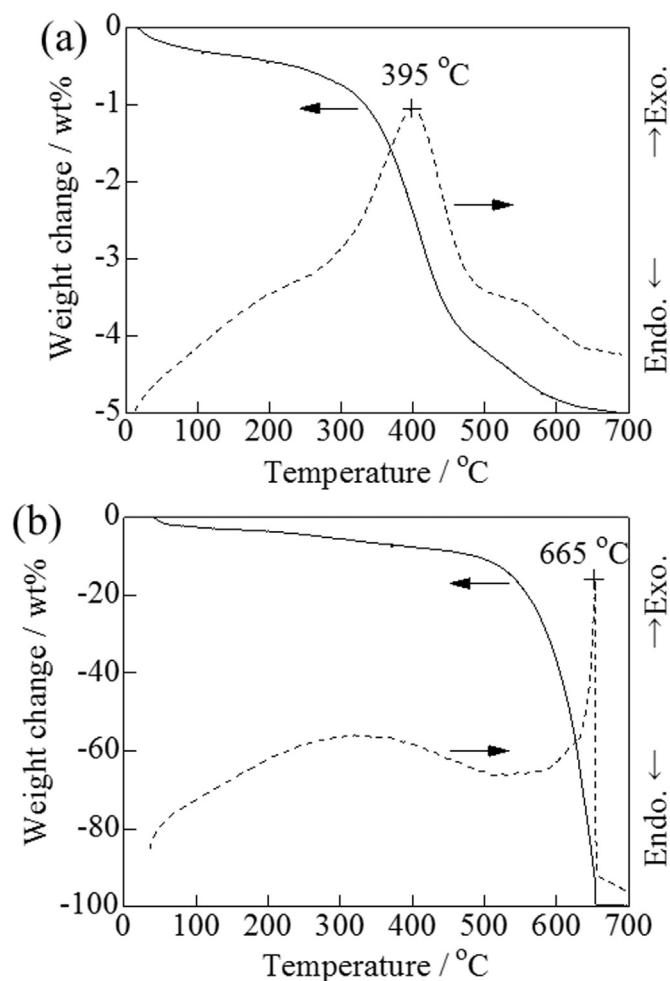


Fig. 1. TG–DTA curves recorded for the (a) 5 wt% carbon black+ YMnO_3 mixture and (b) pure carbon black powder.

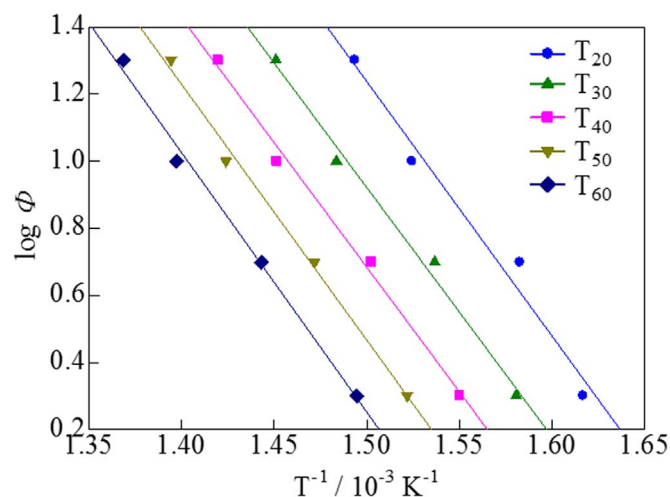


Fig. 2. Ozawa plots obtained for YMnO_3 at different conversion levels of carbon black. Various heating rates (5, 10, 15, and 20 °C min^{-1}) were utilized during carbon black oxidation.

of Ozawa plots at various carbon conversion levels (see Fig. 2). The resulting graphs of the logarithmic heating rate versus inverse temperature obtained at different carbon conversion levels exhibited good linear fits. The magnitudes of E_a determined for the non-catalyzed and catalyzed reactions were 160 and 131 kJ mol^{-1} , respectively, indicating a significant effect produced by the presence of a catalyst on the

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