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Effects of hydrothermal aging on SiC-DPF with metal oxide ash and alkali metals

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ABSTRACT

The silicon carbide used to make diesel particulate filters (SiC-DPF) has a maximum temperature of use, which is not the melting point of the filter material itself but rather the eutectic melting points of the ash materials and alkali metals deposited on the filter wall. Chemical reactions between the SiC filter and the other materials, i.e. ash materials and/or alkali metals, decrease the filtration efficiency and catalytic reactivity of engine out emission. The objective of this study is to understand the effect of hydrothermal aging on the SiC-DPF, and on the SiC-CDPF (catalyzed diesel particulate filter) deposited with ash materials and/or alkali metals. Hydrothermal aging simulated for the extreme condition of uncontrolled regeneration in DPF is carried out by using H_2O at high temperature. The surface change of the SiC filter was characterized in terms of the geometric microstructure and metal composites of the filter by using the SEM-EDS, BET and XRD. The accumulated ash materials and alkali metals in the SiC-DPF were an admixture, and the SiC-DPF after-treatment system always contained H_2O. According to the results, H_2O in the after-treatment system can be regarded as an influential factor of SiC-DPF durability even though the SiC itself has a very high melting point. The regeneration temperature has to be controlled under a critical value to ensure the durability of SiC-DPF in the after-treatment system, considering the fact that large quantities of ash materials, alkali metals and H_2O components are included in the exhaust gas.

1. Introduction

Diesel particulate matter (PM) discharged from diesel vehicles is harmful to both human health and the environment and has been an obstacle to the development of diesel vehicles. An effective technology against PM is the diesel particulate filter (DPF) in the exhaust after-treatment system [1,5].

The materials of the DPF have to have considerable endurance because the filter has to trap the PM from the diesel exhausts during severe operating conditions. At present, silicon carbide (SiC) is one of the most promising candidates as the base material of DPF or catalyzed diesel particulate filter (CDPF) because it has good mechanical performance, excellent heat-resistant properties, and wide applicability in diesel emission control. SiC also has high thermal conductivity, which allows the higher regeneration of the filter by both thermal oxidation and catalytic PM oxidation at the SiC-DPF or CDPF system. With these advantages, SiC-DPF or CDPF has been developed and used on diesel vehicles [2,4,6]. The PM accumulated in the filter system needs to be oxidized or removed physically from the system. In the oxidation method, inorganic oxide materials are added into the diesel fuel as additives admixture to remove the accumulated PM. On the other hand, the physical removal method is hardly used because of its ineffectiveness [3,7]. Ash materials of sulfates, phosphates, or other oxides of calcium and zinc are formed from the burned additives of the engine lubricant in the engine's combustion chamber. Some oxides of metals such as iron, manganese, and cerium, which are presented as additives of catalyst to CDPF to reduce the activation temperature of the catalytic reaction, are required to initiate the catalytic reaction of the soot in the CDPF. In addition, iron-containing oxides may result from the diesel engine friction, and from the erosion of the exhaust system [3,8].

However, the oxidation method may still have some disadvantages because it uses additives as admixtures (Fe, Ce, etc.) in the diesel fuel to decrease the temperature of PM oxidation. For example, the filter will become clogged with ash materials that result from the burning of inorganic materials. Furthermore, a second contamination or slip by the admixture of additives may occur. The accumulated carbon ingredients of the PM are removed during DPF's regeneration process, but the inorganic materials are not removed and can accumulate continuously [3,4,9]. As a result, the pressure drop can increase, gradually, and in turn the temperature of the DPF can increase greatly, and this would ultimately result in the reaction of materials with DPF. Finally, DPF can experience partial thermal aging or can malfunction [4,11,12]. Alkali metals such as sodium and cesium are used as a type of

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Table 1
Prepared sample matrix for inter-reaction between ash/alkali materials and SiC powder.

Material	Components (wt.%)							
	CeO_2	Fe_2O_3	SO_3	CaO	P_2O_5	MgO	Na	Cs
Ash material	100	100	100	100	100	100	–	–
Ash material mixture	58	12	12	8	8	2	–	–
Alkali metal	–	–	–	–	–	–	100	100
Alkali metal mixture	–	–	–	–	–	–	50	50
Ash and Alkali metal mixture	29	6	6	4	4	1	25	25

Specification of SiC substrate				
Melting point (°C)	Porosity (%)	Mean pore diameter (fan)	Purity (%)	Thermal conductivity (W/mK)
2400	55	12	99	60

catalytic washcoat for CDPF [5,13–15,17]. Therefore, when these ash materials or alkali metals join with the DPF, the SiC material of the DPF interacts with the ash materials or/and alkali metals. The effect of this interaction on the temperature at which sintering and adherence to the filter wall occur is studied because this effect can decrease the filtration efficiency and catalytic reactivity of the DPF and CDPF, respectively [1,3,10,16].

The present study aims to understand the behavior and the effect of ash material and alkali metal depositions on the damage of SiC-CDPF monolith during hydrothermal aging upon simulated uncontrolled regeneration. Moreover, hydrothermal aging behavior of catalyzed SiC-DPF with Pt/γ-Al_2O_3 was observed.

2. Experimental

2.1. Sample preparation

The SiC substrate used in this experiment is a commercial product from IBIDEN Co. Engine lubrication oil (Hyundai 5W30) includes compositions of Zn (950 ppm), P (768 ppm), Ca (1850 ppm), S (1700 ppm) and Mg (17 ppm). Al (600 ppm), Ca (400 ppm), Fe (600 ppm), Mg and Mn (50 ppm), Na (150 ppm), Zn (300 ppm) were detected by measurements of wear metals in used engine oil [18]. SiC-DPF system uses fuel-borne catalysts as CeO_2, Fe_2O_3 [4]. The composition of an actual ash of DPF consisted of CaO (29.6 wt.%), ZnO (9.9 wt%), MgO (5.5 wt.%), P_2O_5 (15.8 wt%) and Fe_2O_3 (0.41%) [3]. The mixing portion of ash materials and alkali metals considered this composition. The properties of a SiC substrate are shown in Table 1 [3,10]. An SiC sample extracted from SiC-DPF was fine-crushed into powder, and the catalyst (3 wt.% Pt/γ-Al_2O_3) was added to the SiC-DPF powder to produce a catalyzed DPF. A CDPF was obtained by gently shaking the both the catalyst and the DPF powder in a vessel and by sonification using an ultrasonic cleaner. Subsequently, to initiate the inter-reaction between the SiC substrate and ash materials and/or alkali metals, which was accumulated and deposited in performing the roles of DPF, the samples were pressed into a pellet of 2 mm in thickness and 13 mm in diameter at a special weight ratio (1:10) of ash material and/or alkali metal to SiC powder.

The simplified structure of the experiment sample is showed in Fig. 1(d). Fig. 1 shows the powder sample, the equipment used for pressing the sample (pressure is 400 kg/cm^2), the pellet sample, and the experiment sample. CeO_2, Fe_2O_3, $CaSO_4$, CaO, P_2O_5 and MgO powders were mixed with SiC powder at the weight ratio of 58:12:12:8:8:2. Also, the alkali metals, Cs and Na, were mixed at the weight rate of 1:1. The inter-reaction between SiC substrate and ash material mixture (or alkali metal mixture) was observed, and each mixture was prepared at 1:1 weight ratio. The sample components used in the experiment are shown in Table 1.

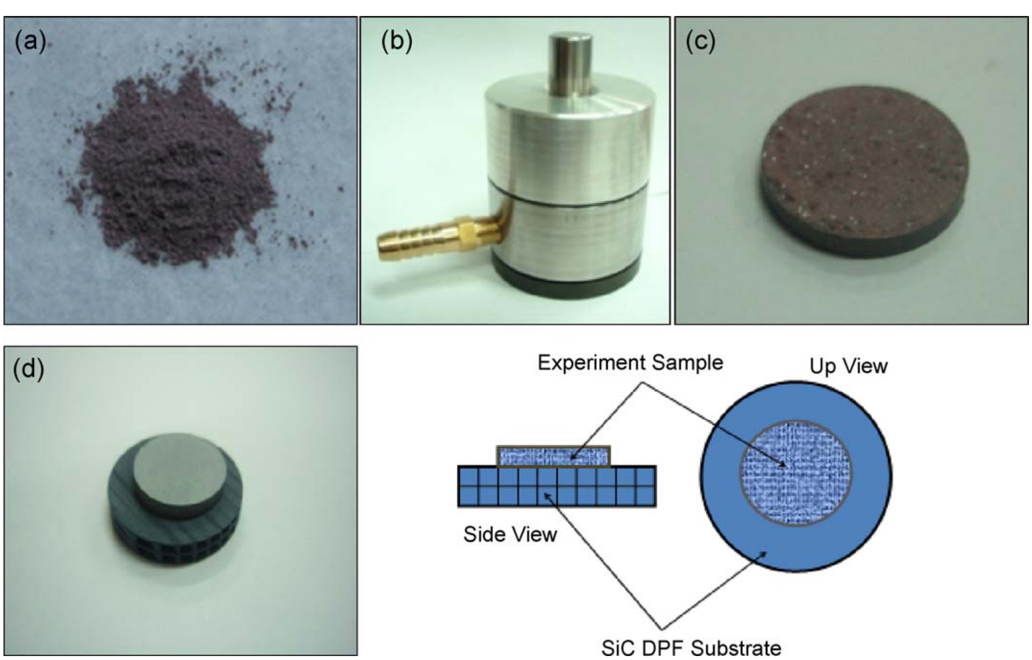

Fig. 1. Images of (a) mixture of Fe_2O_3/SiC powder, (b) equipment used for pressing sample, (c) the pellet shape sample and (d) experiment sample.