

Adsorption and desorption characteristics of hydrocarbons in multi-layered hydrocarbon traps

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ABSTRACT

In this study, a hydrocarbon (HC) trap was developed using a multi-layered zeolite, in which zeolite A and zeolite beta was selected for the water trap and the hydrocarbon trap, respectively, with the goal of reducing hydrocarbon emissions during a cold-start. The adsorption and desorption of hydrocarbons was examined in the monolithic adsorber during a vehicle test as well as in a lab-scale micro-reactor as a powder form. The competitive adsorption of water vapor and hydrocarbons in the exhaust gas from a gasoline vehicle on the hydrocarbon adsorption sites at low temperatures caused the decrease in the HC removal efficiency during a cold-start. In a multi-layered adsorption system composed of the water trap and the hydrocarbon trap, the hydrocarbon removal efficiency was greatly improved owing to the elimination of water with the hydrophilic zeolite A in the upper layer. Furthermore, the hydrocarbon desorption can be delayed because of the latent heat of desorption of adsorbed water in the upper layer. Therefore, this double-layered zeolite-based hydrocarbon trap showed the better performance compared with the conventional hydrocarbon trap during a cold-start.

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1. Introduction

In order to protect global environment, the efficiency of the automobile fuel consumption must be improved and the exhaust gas must be purified as much as possible. One of concerns in the automotive emission control is the hydrocarbons (HC) emitted from gasoline automobiles during a cold-start. Typically, 70–80% of the total hydrocarbon can be emitted at this stage [1–3]. Therefore, this cold-start HC must be reduced to meet the strict emissions regulations required for a super-ultra low emission vehicle (SULEV) or a EURO V. Until now, two different main approaches have been tried to solve this problem [4]. The first concept is to heat the catalyst quickly to its working temperature. The second approach is to trap hydrocarbons during a cold-start for release after the catalyst has reached the operating temperature. Although various detailed techniques have been disclosed, each has been reported to have its own strong and weak points [4]. Among them, the idea of using hydrocarbon traps has been widely examined in order to reduce the amount of hydrocarbons emitted during a vehicle's cold-start [4–13]. The hydrocarbon trap adsorbs hydrocarbon during start-up when the gas is cold. As the exhaust gas heats up, the trap temperature increases, causing the hydrocarbon to be des-

orbed. The hydrocarbon released from the trap material may be converted by a traditional three-way catalyst located either downstream or on the same monolith where the hydrocarbon trap is located. One of important issues on the hydrocarbon trap is the selection of adsorbents in the hydrocarbon trap. The adsorbent in the hydrocarbon traps should be designed to have good selectivity for hydrocarbons. Until now, zeolites are the most commonly-used hydrocarbon trap material [4–25]. Some lab-scale experiments have been reported on various zeolites such as H-ZSM-5 [4,18,21], H-Beta [4,21], La-(or Cu)-ZSM-5 [14], Ag-ZSM-5 [15], EUO [16], mordenite [16], Na-BEA [17], SSZ-33 [19,22], silicoaluminophosphate molecular sieves [20], MCM-68 [22], ZSM-12 [23], Ferrierite [24], heteroatom-substituted microporous AFI and ATS structured materials [25], and Li, K or Ag ion-exchanged Beta [26]. The other important issue on the hydrocarbon trap is the design of the overall system containing the water trap, the hydrocarbon trap and the three-way catalyst. In order to effectively remove adsorbed hydrocarbons, the hydrocarbon trap should be able to release the trapped hydrocarbons after the catalyst has reached the operating temperature. To achieve this goal, the combined hydrocarbon trap in which the HC adsorbent and the three-way catalyst are coated on the same monolith has been proposed. For this system, the HC adsorbent stable even at severe hydrothermal condition is required. On the other hand, the different concept in the HC trap, in which the HC trap is working only at low temperatures

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and the exhaust gas at high temperatures is bypassed the HC trap and directed into the three-way catalyst, can be proposed because the adsorption capacity of zeolite is generally decreased after the hydrothermal aging. For this system, it is quite required to attain a proper overlap between the hydrocarbon desorption temperature and the catalyst light-off temperature in the three-way catalyst. Therefore, the amount of adsorbed HC as well as the desorption temperature should be increased. Because the competitive adsorption can be occurred between water and the hydrocarbons on zeolite, the addition of the water adsorbent into the HC trap can increase the amount of HC adsorbed. Furthermore, the latent heat of water desorption can delay the desorption of hydrocarbons adsorbed on the HC trap during the desorption stage. In this study, a double-layered zeolite-based hydrocarbon trap was prepared to remove hydrocarbon effectively under a cold-start condition. A mass spectrometer utilizing chemical ionization was used in a vehicle test to examine the adsorption–desorption characteristics of hydrocarbons in HC traps.

2. Experimental

2.1. Preparation of the hydrocarbon traps

The monolithic hydrocarbon traps used in this study were prepared by applying a wash-coat to a cordierite honeycomb ceramic substrate (cell density, 93 cells/cm² or 600 cells/in²; wall thickness, 0.102 mm; size, 84 mm × 148 mm × 86.5 mm, or 0.9 L). The wash-coat was composed of zeolite A (COSMO), zeolite beta (Si/Al = 300; Zeolyst) and zeolite binder. In the lower layer, the hydrocarbon adsorbent (zeolite beta) with a hydrophobic property was coated. In the upper layer, the water adsorbent (zeolite A) with a hydrophilic property was incorporated. After the zeolites were applied to the monolith, all samples were dried in air at 150 °C and calcined in air at 530 °C. Multi-layered hydrocarbon traps contain only a water adsorbent and hydrocarbon adsorbent without precious metals. This was done to eliminate the possibility of precious metal acting as a catalyst to convert hydrocarbons, which could complicate the data interpretation. Table 1 lists the compositions of the hydrocarbon traps prepared in this work. To confirm the hydrocarbon adsorption–desorption characteristics, the powder samples listed in Table 2 were prepared from the water adsorbent and the hydrocarbon adsorbent. They were slowly mixed in a mortar for 20 min.

The scanning electron microscopy (SEM) image of the cross-section of the double-layered HC trap was obtained in JEOL; JSM 5610.

Table 1
Preparation of the hydrocarbon traps (monolith type) (unit:g/L).

Catalysts	Amounts of hydrocarbon adsorbent	Amounts of water adsorbent	Remarks
Sample 1	50	–	a
Sample 2	70	–	a
Sample 3	90	–	a
Sample 4	70	5	b
Sample 5	70	15	b
Sample 6	70	5	c

^a Hydrocarbon trap.

^b Layered-type trap.

^c Mixed-type trap.

Table 2
Preparation of the hydrocarbon traps (powder type) (unit:g).

	Amounts of hydrocarbon adsorbent	Amounts of water adsorbent
Case 1	100	0
Case 2	98	2
Case 3	95	5
Case 4	90	10
Case 5	80	20
Case 6	70	30
Case 7	50	50
Case 8	30	70
Case 9	0	100

2.2. Temperature-programmed hydrocarbons desorption (TPD) test

To determine the effect of the water removal on the performance of hydrocarbon trap materials, hydrocarbon-trapping experiments were also performed on powder samples in a micro-reactor. The sample was pre-activated at 520 °C with a N₂ flow of 1000 cc/min. Subsequently, the powder sample was cooled to 50 °C in a N₂ flow of 40 cc/min. After a cleaning procedure, a test gas of 2.4% C₃H₆, 0.6% C₃H₈ and 10% H₂O in N₂ was introduced into the test sample for 2 min at 50 °C. After 2 min, the gas was changed to pure N₂ (1000 cc/min) for desorption of hydrocarbons from the sample and the N₂ purging was confirmed until the concentration of desorbed hydrocarbon from the sample was below 5 ppm at the ambient temperature. The test sample was heated to 500 °C at a heating rate of 10 °C/min. The hydrocarbons were measured using a standard quadrupole mass spectrometer (V&F; Austria), monitoring masses of 42 (propene) and 46 (propane).

2.3. A vehicle test

In this study, all vehicle tests were performed on the exhaust of a 2000 Optima with a 2.0 L engine, during Phase 1 of the federal test procedure (FTP). This vehicle was fueled with a low-sulfur (below 10 ppm) gasoline. The catalytic converter of hydrocarbon trap was placed in the exhaust system's under-floor, 29.5 in. from the engine manifold. Fig. 1 shows the inlet exhaust gas temperature for the catalytic converters, measured at 1 inch in front of the catalytic converter for the first 500 s of the test. Sample ports were inserted in the exhaust pipe in order to collect emissions at inlet and outlet of the hydrocarbon trap. Total hydrocarbon emissions were

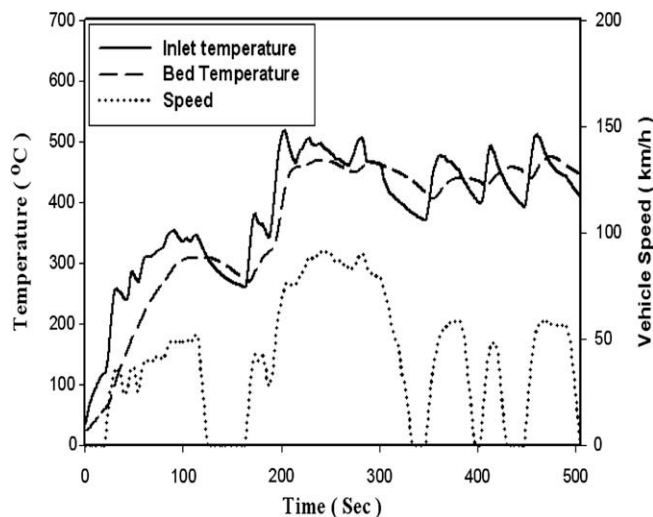


Fig. 1. The temperature and the vehicle speed profiles during the federal test procedure (FTP).

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