



Research article

Volatile organic compounds (VOCs) in surface coating materials: Their compositions and potential as an alternative fuel



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ABSTRACT

A sampling system was designed to determine the composition ratios of VOCs emitted from 31 surface coating materials (SCMs). Representative architectural, automotive, and marine SCMs in Korea were investigated. Toluene, ethylbenzene, and xylene were the predominant VOCs. The VOC levels (wt%) from automotive SCMs were significantly higher than those from architectural and marine paints. It was found that target SCMs comprised mainly VOCs with 6–10 carbon atoms in molecules, which could be adsorbed by activated carbon. The saturated activated carbon which had already adsorbed toluene, ethylbenzene, and *m*-xylene was combusted. The saturated activated carbon was more combustible than new activated carbon because it comprised inflammable VOCs. Therefore, it could be an alternative fuel when using in a “fuelization system”. To use the activated carbon as a fuel, a control technology of VOCs from a coating process was also designed and introduced.

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1. Introduction

Volatile organic compounds (VOCs) are organic chemical compounds that have a boiling point less than or equal to 250 °C at 101.3 kPa of pressure (EUROPA, 2004). VOCs may also be defined as carbon compounds participating in atmospheric photochemical reactions except for several compounds listed by U.S. EPA (U.S.EPA, 2009). Some VOCs are toxic and affect human health (Guo et al., 2004; Massolo et al., 2010; Wolkoff and Nielsen, 2010; Wu et al., 2012). Benzene exposure causes a 40% increase in a lifetime cancer risk due to VOCs in the indoor environment. Thus, VOC exposure can contribute to an increased lifetime cancer risk (Guo et al., 2004). In particular, children living in industrial areas are exposed to VOCs (Massolo et al., 2010). In contrast, some VOCs do not affect cancer risk in humans, but they still cause health risks. A formaldehyde concentration of 0.3–0.5 mg/m³ may irritate the eyes (Wolkoff and Nielsen, 2010). Hexane and toluene are associated with health risks to the respiratory system. Toluene, hexane, xylene, and benzene increase health risks to the neurological

system (Wu et al., 2012). Furthermore, VOCs and NO_x are the main chemical precursors of ground-level ozone in the presence of sunlight (Chang et al., 2005; Duan et al., 2008; Latella et al., 2005; Ling et al., 2011; Qin et al., 2004). Especially, VOCs play a significant role forming ground-level ozone when the VOCs/NO_x ratio is high (Carter, 2013; Qin et al., 2004). VOCs also have indirect negative effects because VOCs are precursors of ground-level ozone, which increases risks to human health and welfare (Karaca and Ozturk, 2012; Li et al., 2011; Paoletti et al., 2014; Schlink et al., 2006; Sousa et al., 2011; Teixeira et al., 2011).

Surface coating materials (SCMs), as a source of VOCs, also contribute negatively to the environment. Automotive paint manufacturing releases 0.02–0.09 kg of VOCs per kg of paint (Papasavva et al., 2001), and 2–9% of household products are coated with paint (Zhang and Niu, 2003). It has been reported that 71 kilotons of architectural paint were used in Beijing, China, in 2007, of which toluene, xylene, and ethylbenzene are the most abundant VOCs (Yuan et al., 2010). Celebi and Vardar (2008) reported that 900 tons of total VOCs were emitted from the Tuzla shipyard area in 2006. Paint solvents in Seoul, Korea, account for about 46% of VOCs (Na et al., 2004). Many studies have been carried out on the emission characteristics of VOCs from SCMs (Anderson et al., 2006; Celebi and Vardar, 2008; Chung and Lee (2009);

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Katsoyiannis et al., 2012; Kim, 2011; Kim et al., 2000, 2011; Kwon et al., 2007; Yu and Crump, 2000; Yuan et al., 2010). However, there were few studies on the composition of VOCs from architectural paint (Kwon et al., 2007, 2008; Yu and Crump, 2000). No study has determined the VOCs composition ratios from automotive and marine paints. Particularly, no research has compared VOC characteristics among architectural, automotive, and marine paints.

There are many available techniques to control the emission of VOCs. They can be classified into process - equipment modification and add-on control techniques. The modification of process and equipment is more difficult than that of the add-on control system. Therefore, add-on control techniques have high applicability (Khan and Ghoshal, 2000). However, because the flue gas volume is large and the compositions and concentrations of VOCs from the painting process are very complicated, VOCs are not easy to control or remove (Alvarez-Hornos et al., 2011; Chang et al., 2002; Pierucci et al., 2005). Some popular control technologies were used and reported (Alvarez-Hornos et al., 2011; Hussey and Gupta, 1997; Stone, 1997). Thermal oxidizers such as regenerative thermal oxidizer and catalytic oxidizer are used to remove VOCs from coating processes. However, it is a big issue to remove paint particles during the coating process. On the other hand, adsorption using activated carbon (AC) has been widely used. Nevertheless, using this technology might lead to release a large amount of hazardous solid waste because of the paint particles. VOCs in coating materials such as toluene, xylene, and ethylbenzene are inflammable compounds with the high heat of combustion (40,940 kJ/kg, 41,214 kJ/kg, and 41,326 kJ/kg, (Perry et al., 1997) respectively). Consequently, AC containing flammable VOCs might be an alternative fuel if most of the paint particles could be removed before the adsorption process. There have not been any studies about this in the world.

Accordingly, this study was conducted to assess VOCs from various kinds of SCMs. The combustible ability of used AC was also investigated. Additionally, a fuelization process was also suggested to depict reusing saturated AC as an alternative fuel.

2. Experimental methods and materials

2.1. VOCs composition of SCMs

2.1.1. Sampling system

A surface coating material comprises non-volatile compounds and volatile compounds. Hence, we designed a sampling system to determine the amount of these components. The sampling system consisted of a stainless steel chamber (25 mL), a zero air system, and a moisture drier (Fig. S1, supplementary information). On the basis of headspace method, a stainless steel chamber was designed to collect VOCs emitted from SCM. The chamber inlet and outlet were installed with two stainless steel valves. The chamber temperature was maintained at 40 °C with a water bath. A zero air generator (Model 701, API) was used to supply zero air to the chamber. Flow rate was controlled by a mass flow controller (model FC-280S, Tylan, Milford, MA, USA). The zero air flow rate was maintained at 100 mL/min. A moisture drier system was equipped with a Teflon coil surrounded by dry ice to capture water vapor emitted from VOC samples. The temperature inside the coil was approximately -5 °C. When the flue gas was flowed into the Teflon coil, only water vapor in the gas stream was changed from gas phase to solid phase and attached inside the coil because of the different boiling points of water and target gases.

2.1.2. Materials

Architectural (16 products: A1–A16), marine (10 products: S1–S10), and automotive (five products: C1–C5) SCMs were

carefully selected based on frequent use in Republic of Korea (Table 1). Korea had been the leadership in the global shipbuilding from mid-1990's to present. Korea has been had top five biggest shipbuilding companies and the fifth leading motor vehicle manufacturer in the world by 2014 (Mickeviciene, 2011; SAJ, 2014; Shin and Lim, 2014; Statista, 2015). Therefore, samples used in this study are highly representative. After picking up SCMs, chemical composition of SCMs had been instantly determined in the laboratory. After being opened, a container was carefully re-sealed by its cap.

2.1.3. Analytical method

A gas chromatography (GC) (Agilent 6890, Palo Alto, CA, USA)/mass selective detector (MSD, Agilent 5975) equipped with a thermal desorber (TD) (Unity 2, Markes International, USA) was used to analyze VOCs. A DB-624 GC column (L, 60 m × ID, 320 μm × 1.8 μm, Agilent J&W, USA) was used. Ozone precursor standard gas (56 different substances, Photochemical Assessment Monitoring Stations mix; Supelco, Bellefonte, PA, USA) was used to determine retention times and to identify the chemicals (Table S1, Supplementary information). The standard gas was also used to calibrate the response factors of MSD for the ozone precursors. A mass spectrometry library (WILEY 7.0) was also used to identify the other compounds.

The samples were collected on an adsorbent trap (Carbopack C, Carbopack B, and Carboxen 1000). The trap was desorbed using the TD. The breakthrough point of the adsorbent trap was observed approximately 70 mg of toluene at room condition (25 °C ± 2 °C, 1 atm). The trap was pre-purged with helium for 3 min at a split rate of 20 mL/min. Then, it was desorbed at 280 °C for 5 min. The desorbed gas was introduced into the TD cold trap. The temperature of the cold trap was -10 °C. Then, it was pre-purged for 1 min and desorbed for 5 min at 280 °C to introduce the desorbed gas into the GC through a heated (150 °C) fused silica line. The initial temperature of the GC oven was -30 °C. Temperature was increased at a rate of 3 °C/min to 120 °C and held for 10 min. Then, it was increased to 170 °C at a rate of 3 °C/min. At 170 °C, the temperature was increased continuously to 200 °C at a rate of 10 °C/min and held for 0.33 min. Total analytical time was 80 min. Helium (99.999%, Dong-A Gas Ltd., Seoul, Korea) was used as the carrier gas at a flow rate of 1.5 mL/min. The source temperature of the MSD was 230 °C, and the mass range was 35–350 amu.

2.1.4. Experimental procedure

The experimental process was separated into 3 steps. In step 1, we investigated the residence time of the coating material samples in the chamber. Two grams of each sample were placed in a glass Petri dish. The dish was put in the stainless steel chamber. Then, the chamber was placed in a water bath to maintain a temperature of 40 °C and to allow the VOCs to be emitted easily. Zero air was flowed into the chamber at a rate of 100 mL/min.

The mass of a sample was checked with a balance (Adventurer Pro AVG264C, Ohaus, Parsippany, NJ, USA) every hour. The process was repeated until the mass of the sample was constant. Total elapsed time (ET) was recorded. To prevent errors, total residence time (RT) of the sample in the chamber was calculated as total ET plus 1 h (RT = ET + 1).

The masses of the volatile and non-volatile compounds were calculated by equations (1) and (2), respectively.

$$M_{\text{volatile compounds}} = M_{\text{db}} - M_{\text{da}} \quad (1)$$

$$M_{\text{non-volatile}} = M_{\text{da}} - M_{\text{d}} \quad (2)$$

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