



# Removal of gaseous sulfur and phosphorus compounds by carbon-coated porous magnesium oxide composites

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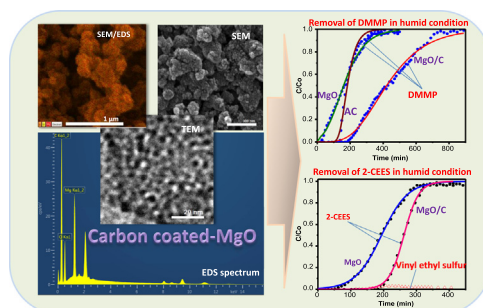
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## HIGHLIGHTS

- Carbon-coated MgO composites were synthesized using an aerogel method.
- MgO/C composites had a high surface area of 723 m<sup>2</sup>/g.
- The sorption capacity of DMMP and 2-CEES was higher than those of MgO and AC.
- The composite sorbed DMMP almost twice more than 2-CEES in dry condition.
- Carbon layer on MgO protected active catalytic and sorption sites from H<sub>2</sub>O molecules.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Carbon-coated porous magnesium oxide (MgO/C) composites were synthesized using an aerogel route for removal of dimethyl methylphosphonate (DMMP) and 2-chloroethyl ethyl sulfide (2-CEES) in dry and wet conditions. The sorption capacities of the as-prepared samples for DMMP (0.23 µg/mL) and 2-CEES (0.26 µg/mL) were evaluated by breakthrough experiments in nitrogen under ambient conditions. MgO/C composites exhibited a decrease in surface area with carbon content (648–723 m<sup>2</sup>/g), but had a higher surface area than MgO. Under dry conditions, the sorption capacities of the MgO/C composite with a low carbon content of 6.39 wt% (MgO/C-1; 67.8 mg/g for DMMP and 35.3 mg/g for 2-CEES) were higher than those of pure MgO and activated carbon (AC). The sorption capacity of MgO/C composites decreased with an increase in carbon content and became even lower than those of MgO and AC. Under humid conditions, the sorption capacities and breakthrough time of pure MgO decreased significantly and became lower than that of AC. In contrast, the sorption capacities of the MgO/C-1 composite for DMMP and 2-CEES under humid conditions remained at about 91 and 86% of those measured under dry conditions, and were higher than those of AC. In addition, the MgO/C composite remained reactive toward 2-CEES even under humid conditions. MgO/C composites were more stable than MgO under humid conditions because of the presence of carbon-coated shells.

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## 1. Introduction

Removal of hazardous chemicals from the environment is a critical issue from both biological and environmental standpoints

[1,2]. Environmental regulations for emissions from industries and acceptable levels of human exposure are continuously being adjusted and made more stringent.

The most abundant hazardous components can be classified into two categories based on their source: natural and anthropogenic hazardous materials. Anthropogenic pollutants generally originate from combustion, chemical reactions, or from the

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unsecured effluent of toxic materials. Furthermore, a considerable amount of anthropogenic chemicals containing sulfur and phosphorus such as mustard, sarin, and soman agents are produced for military purposes [3]. These anthropogenic chemicals are highly persistent in the environment and critically harm humans even at low concentrations.

Many efforts have been made both to reduce pollution and to eliminate toxic materials from the environment. Adsorptive removal of toxic components from the atmosphere is one of the most attractive technologies [4].

Among various candidate solid materials, metal oxides have been reported to be effective materials for adsorption and decomposition of toxic and persistent chemicals due to their high surface areas, large number of highly reactive edges, corner defect sites, unusual lattice planes, high surface-to-volume ratio, and reusability [5–11]. Particularly, magnesium oxide (MgO) materials have been the focus of attention for decontamination of toxic chemicals in recent years because well-designed MgO materials have a high sorption capacity as well as effective decomposition ability.

Many studies have focused on methods to synthesize MgOs with high surface area, pore volume, and small crystal size to improve sorption capacity and reactivity for toxic chemicals [12–15]. However, sorption capacity and reactivity of MgO are significantly reduced in the presence of water, because of the strong affinity of MgO for water [6]. Furthermore, the excellent removal performance of hazardous chemicals on adsorbents under dry conditions does not guarantee effectiveness in practice, because these chemicals exist in the environment with a certain level of humidity. Even though many solid materials have been reported to have higher efficiency under dry conditions than activated carbon, their performance is often equivalent or worse than that of activated carbon under humid conditions. Therefore, selection of solid materials for the removal of hazardous chemicals under humid conditions has been limited to strong hydrophilic adsorbents. It would be highly desirable to design MgO-based composites with hydrophobic surfaces that show high removal capacity for hazardous chemicals under humid conditions.

Carbon-coated metal oxides have been produced using various synthesis methods for various applications. A carbon-coated metal oxide was developed to improve the stability, electric conductivity, and electrochemical performance of batteries [16]. Coating of a metal oxide with hydrophobic carbon could minimize the problem of water adsorption because carbon has a non-polar surface [17]. In addition, various carbon-coated metal oxides have been developed, such as carbon-coated ZnO and CaO prepared by poly vinyl alcohol pyrolysis [18], ferrite nanoparticles coated with carbon prepared using a hydrothermal method [19], and carbon-coated Ni/TiO<sub>2</sub> prepared via a hydrothermal method [20]. To create MgO/carbon composites, various synthesis methods have been suggested, including chemical vapor deposition (CVD), pyrolysis of a magnesium hydroxide aerogel modified with resorcinol, precipitation with the assistance of sucrose, and one-pot assembly [17,21–23].

A chemical weapon agent (CWA) is a chemical substance whose toxic properties are used to kill, injure, or incapacitate soldiers (and sometimes civilians). Therefore, the decontamination of chemical warfare agents is arguably the most challenging issue facing militaries around the world. Furthermore, due to possible terror threats, protecting civilians from CWAs has become increasingly important for many governments.

Sulfur mustard (SM), commonly known as mustard gas, is a class of related cytotoxic and vesicant CWAs, which can cause large blistering on any exposed skin and in the lungs [24]. There is no known antidote or specific treatment against SM exposure, and the current therapy is largely supportive. Like some other nerve CWAs, sarin attacks the nervous system by interfering with the degradation of the neurotransmitter acetylcholine at

neuromuscular junctions, and death will usually occur [25]. In various studies, dimethyl methylphosphonate (DMMP; CH<sub>3</sub>PO(OCH<sub>3</sub>)<sub>2</sub>) is used as a simulant for toxic phosphorus compounds such as sarin [26] and 2-chloroethyl ethyl sulfide (2-CEES; CH<sub>3</sub>CH<sub>2</sub>SCH<sub>2</sub>CH<sub>2</sub>Cl) is often considered as a surrogate compound for SM [27,28]. Therefore, development of effective materials for removing DMMP and 2-CEES could help protect the environment and humans from refractory hazardous chemicals as well as CWAs.

In this study, carbon-coated magnesium oxide (MgO/C) composites were synthesized via an aerogel route to remove efficiently CWAs. As representative surrogates of CWA, dimethyl methylphosphonate and 2-chloroethyl ethyl sulfide were selected. The sorption capacities of the MgO/C composites were measured by breakthrough experiments in gas phase under ambient dry and wet conditions (0.23 and 0.26 μg/mL DMMP and 2-CEES in N<sub>2</sub> flow, respectively). The removal efficiency of the MgO/C composites were evaluated and compared with those of pure MgO and activated carbon (AC).

## 2. Experimental section

### 2.1. Materials

The following materials were used in this study: toluene (Aldrich, USA, 99.9%), a magnesium methoxide solution in methanol (Aldrich, 7.82%), 2-CEES (Aldrich, 98%), DMMP (Aldrich, 97%), glucose (Aldrich, 99%), activated carbon (Calgon Filtrasorb 300, coal-derived AC), N<sub>2</sub> gas (Dae-Deok Gas, Korea, 99.999%), H<sub>2</sub> (Dae-Deok Gas, 99.999%), and air (Dae-Deok Gas, 99.999%). All chemicals and solvents were used without further purification.

### 2.2. Preparation of composites

MgO/C composites were developed using an aerogel procedure. In a typical experiment, which was conducted at room temperature, a mixture of toluene (100 mL) and magnesium methoxide solution (20 mL) was placed in a glass reactor with a stirrer. Another solution was prepared by dissolving the desired amount of glucose in 2.0 mL of distilled water and this solution was slowly added using a syringe to prepare the mixture. The addition of the glucose solution led to a white cloudy precipitate, but the solution became clear after a few minutes. To minimize evaporation of organic solvent, the glass reactor top was covered with aluminum foil. The mixture was stirred vigorously overnight at room temperature to allow completion of hydrolysis.

Subsequently, the hydrolysis gel was put into a high-pressure autoclave reactor. The gel was first flushed and then pressurized up to 100 psi with N<sub>2</sub> gas. The autoclave reactor was gradually heated from room temperature to 265 °C at a rate of 1.0 °C/min and this temperature was remained for 10 min. Solvent vapors in the reactor were quickly vented to the atmosphere and flushed with N<sub>2</sub> again to remove any remaining solvent vapors. The obtained powder in the reactor was dried in an oven at 120 °C for 12 h to remove residual organic solvents. Hydrated powder (before calcination) was obtained and denoted HY-MgO/C.

The final step was calcination. Calcination has been shown to improve the textural properties and sorption capacity of MgO [12,29]. In this study, the hydrated powder was calcined in a furnace under vacuum using the following steps to produce mesoporous MgO with a high surface area: (1) ramping from room temperature to 220 °C at 1 °C/min and soaking at 220 °C for 5 h, (2) ramping from 220 to 280 °C at 0.8 °C/min and soaking at 280 °C for 1 h, (3) ramping from 280 to 350 °C at 0.8 °C/min and soaking at 350 °C for 2 h, and (4) ramping from 350 to 500 °C at 0.8 °C/min

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