



Effect of blast furnace dust on the degradation of chlorinated organic and endocrine disrupting compounds

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ABSTRACT

This research was to study the effect of blast furnace dust (BFD) as a new reactive material for the degradation of chlorinated organic (COCs) and endocrine disrupting compounds (EDCs). When 100 g/L of BFD was used, the effective degradation of tetrachloroethylene (PCE) was obtained. The *cis*-DCE (0.93 mM) was dechlorinated to below detection limit within 120 h of reaction. Among various COCs and EDCs, they were degraded at least 90% except for sodium perchlorate. The metabolites of 4-*tert*-octylphenol (4-*t*-OP) were identified as 2,4,4-trimethyl-2-pentanol, hydroquinone and 2-*tert*-octylhydroquinone, respectively. The effective degradation of PCE and 4-*t*-OP was occurred at range of pH 4–7. A total of 100% of *cis*-DCE and 86% of 4-*t*-OP were degraded in fed-batch experiments after 264 h. A solution of highly enriched bacteria completely insolubilized 2.14 mM of Zn eluted from BFD within 168 h of culture. These researches will provide more information related to the application for other contaminated water and wastewater treatments.

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1. Introduction

The widespread contamination of drinking water sources with chlorinated organic compounds (COCs) and endocrine disrupting compounds (EDCs) has led to a concerted effort to find efficient and cost effective treatment methods for these compounds. Chlorinated organic compounds such as tetrachloroethylene (PCE), trichloroethylene (TCE), and *cis*-1,2-dichloroethylene (*cis*-DCE) are primarily used as industrial solvents, while EDCs such as nonylphenol and 4-*tert*-octylphenol are detergents, and bisphenol A is used in polycarbonate. These chemicals are prevalent and persistent groundwater contaminants, which are all known or suspected human carcinogens, mutagens or toxins [1–4]. The most cost effective method used to treat contamination by COCs and EDCs involves bioremediation. However, degradation using biological methods often takes longer than other processes, such as chemical reduction, and may produce toxic by-products resulting in incomplete

biodegradation [5–7]. Another dechlorination approach that has recently garnered substantial interest is the use of zero-valent metals, and iron, in particular, has been successful in the dechlorination of COCs [8–11]. Iron is relatively inexpensive but it is still a precious resource in the steel industry. Thus, a recycled material, slag, has been used as substitute for zero-valent iron for the treatment of COCs. Although slag is, in fact, classified as a waste material, it has been reused for various purposes for a long time. The European Waste Catalogue did not include slag, and in 1995 the council of the OECD (Organization for Economic Cooperation and Development) decided to exclude all slag materials that meet related standards, from the list of wastes [12,13]. Thus, slag is currently considered a by-product, rather than a waste.

Significant quantities of blast furnace dust (BFD) are generated as a solid waste material from integrated steel manufacturing plants. BFD is a mixture of oxides expelled from the top of the blast furnace, whose major components are iron oxides. Unfortunately, the direct recycling of BFD is not usually possible as it contains undesirable elements like Zn [14]. However, if it proves possible to remove COCs with BFD, and a cost-effective method for removing Zn from the BFD is established, then it will become possible to use this material in the treatment of COCs and EDCs. Many researchers have tried to utilize BFD in the wastewater treatment. For example, Li [15] reported a study on the degradation of 4-chlorophenol using BFD. BFD was also applied as biofilm support to investigate the removal of chemical oxygen demand and ammonia nitrogen from brewery wastewater in two lab-scale up-flow biological aerated

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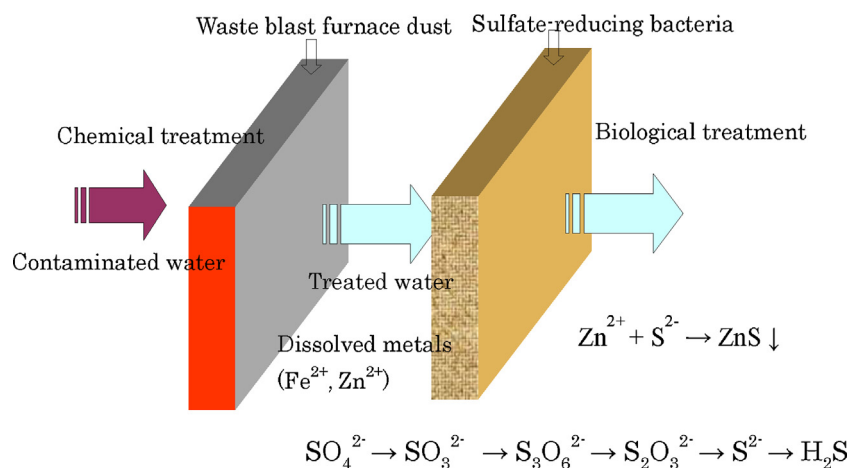


Fig. 1. Conceptual processing scheme for the treatment of COCs and EDCs using BFD and sulfate reducing bacteria.

filters [16]. Jain et al. [17] investigated the utilization of BFD as adsorbents for the removal of anionic dyes such as ethyl orange, metanil yellow and acid blue from aqueous solutions. However, BFD was not suitable for the wastewater treatment. A comparative study of the adsorbents prepared from BFD for the removal of 2-bromophenol, 4-bromophenol and 2,4-dibromophenol from wastewater was also carried out. As compared to carbonaceous adsorbent, the BFD adsorb bromophenols to a much smaller extent [18]. The removal of 2,4-dichlorophenoxyacetic acid and carbofuran from aqueous solution was also compared by using carbon slurry and steel industry wastes (blast furnace slag, dust, and sludge) as adsorbents in batch. Adsorption was found to be in decreasing order: carbon slurry, blast furnace sludge, dust, and slag, respectively [19]. Although many researchers have attempted to apply the utilization of BFD for the wastewater treatment, there have been no reports of the degradation of COCs or EDCs with BFD as reactive material. Recently, we found that the BFD was the effective material for the degradation of COCs or EDCs compared to different steel industry wastes and designed a new conceptual processing scheme for the chemical and biological treatment of COCs and EDCs using BFD and sulfate reducing bacteria shown in Fig. 1.

On the other hand, recently, the Ministry of the Environment in Japan reported that COCs contamination was conspicuous and PCE contamination was the most serious among COCs according to the total number of wells exceeding environmental quality standards identified from the general monitoring survey in 2011.

Thus, in this study we preferentially studied the feasibility of using BFD as a new reactive material for the degradation of a variety of COCs involving PCE. In addition, degradation of EDCs was investigated. Furthermore, the removal of Zn eluted from BFD was tested using *Desulfomaculum nigrificans* (ATCC® 19998™) and enriched bacteria such as *Desulfovibrio* sp. NC-1, *Desulfomonas* sp. NC-1, and *Clostridium* sp. NC-1 isolated from a closed mine in Hokkaido, Japan.

2. Materials and methods

2.1. Chemicals

All chemicals used were of analytical grade. The following chemicals: *cis*-DCE, 4-chlorophenol (4-CP), 2,4-dichlorophenol (2,4-DCP), 2,4,5-trichlorophenol (2,4,5-TCP), 2,4,6-trichlorophenol (2,4,6-TCP), *p*-chlorophenol (CP), 4-*n*-propylphenol (4-PPP), 4-*n*-butylphenol (4-BP), 4-*n*-hexylphenol (4-HP), 4-*n*-nonylphenol (4-NP), 2-*tert*-amylphenol (2-AP), 2-bromophenol (2-BP), 2-*sec*-butylphenol (2-*s*-BP), 3-ethylphenol (3-EP), 4-ethylphenol (4-EP), 4-*tert*-butylphenol (4-*t*-BP), 4-*tert*-octylphenol (4-*t*-OP), bisphenol A (BPA), bisphenol F (BPF), bisphenol S (BPS), chlorobenzene, 1,3,5-trichlorobenzene, 1,2,3,5-tetrachlorobenzene, and sodium perchlorate, were purchased from Tokyo Chemical Industry (Tokyo, Japan). The remaining chlorinated chemicals: PCE, TCE, *trans*-1,2-dichloroethylene (*trans*-DCE), 1,1-dichloroethylene (1,1-DCE), 1,1-dichloroethane (1,1-DCA), 1,2-dichloroethane

(1,2-DCE), 1,2-dichloropropane (1,2-DCP), 1,1,2-trichloroethane (1,1,2-TCE), and vinyl chloride (VC), were purchased from GL Science (Tokyo, Japan). The BFD was kindly provided by Nippon Steel Corporation, Japan. Iron powder (99.99% in purity) was purchased from Wako Pure Chemical Industries, Japan.

2.2. Pretreatment of the BFD by washing

Quantitative analysis of the COCs and EDCs was difficult because of the impurities (such as oils) in the original BFD, so the BFD was pretreated prior to use. Twenty grams of BFD and 20 mL of acetone were added to a 50 mL centrifuge tube and centrifuged for 5 min at 2500 × g. The supernatant was then discarded after further centrifugation (8000 × g, 15 min). The wet BFD was then air-dried at room temperature for 24 h in a drier and this material was used in each degradation experiment. This process was repeated until total organic compound (TOC) in the BFD was less than 3 mg/L. The concentration of TOC was analyzed using a TOC-5000A (Shimadzu, Kyoto, Japan).

2.3. Dechlorination of PCE and *cis*-DCE

Dechlorination of chlorinated aliphatic compounds was performed in a 50 mL vial containing 30 mL of de-aerated deionized water (prepared by boiling the water and purging with nitrogen, 99.99%) and BFD (3 g). The vial was sealed with a Teflon-lined rubber septa and aluminum crimp caps. PCE (0.5 mM) or *cis*-DCE (10.3, 30.9, 309, and 928 μM) was then injected into the vial, the vial was wrapped in foil to avoid light interference, and the reaction was conducted by shaking (120 rpm) at 30 °C. The degradation of *cis*-DCE in the vials was estimated by subtracting the abiotic loss (leak) from the final concentration of *cis*-DCE. During the incubation, the loss of *cis*-DCE in the abiotic reaction was less than 2.0%. For the analysis of PCE and *cis*-DCE, headspace samples (250 μL) were directly taken from each vial. Before analysis, the bottles were shaken well by hand and allowed to stand for 15 min at 30 °C to reach equilibrium. The concentration of each compound in the vial was calculated using the appropriate Henry's law constants [20]. The experiment was conducted in triplicate. A control experiment had no added BFD.

In determining the optimal concentration of BFD for the PCE dechlorination, different concentrations of BFD, 0, 0.3, 0.5, 1, 3, 5, and 10 g were added to 50 mL vial containing 30 mL of working volume comprising 0.5 mM of PCE. Batch experiments were carried out at 120 rpm under anaerobic conditions by purging with nitrogen at 30 °C for 72 h.

To study changes in PCE concentrations, and degradation products with reaction time, batch experiment in 50 mL vial containing 30 mL of working volume comprising 3 g of BFD and 0.5 mM of PCE was carried out at 120 rpm under anaerobic conditions by purging with nitrogen at 30 °C for 192 h.

In addition to investigate effect of initial *cis*-DCE concentration on dechlorination, batch experiments in the 50 mL vial containing 30 mL of working volume comprising 10.3, 30.9, 309, and 928 μM of *cis*-DCE with 3 g of BFD were carried out at 120 rpm under anaerobic conditions by purging with nitrogen at 30 °C for 96 h.

2.4. Degradation of other chlorinated aliphatics and various EDCs

Thirty milliliters of de-aerated deionized water (prepared by boiling then purging with nitrogen) and 3 g of the BFD were added to a 50 mL vial, which was sealed with a Teflon-lined rubber septa, and aluminum crimp cap. The chlorinated aliphatic compounds (0.3 mM), TCE, *trans*-DCE, 1,1-DCE, 1,1-DCA, 1,2-DCE, 1,2-DCP, 1,1,2-TCE, and VC, were then injected to separate vials. The degradation experiments for a range of EDCs (4-CP, 2,4-DCP, 2,4,5-TCP, 2,4,6-TCP, CP, 4-PPP, 4-BP, 4-HP, 4-NP, 2-AP, 2-BP, 2-*s*-BP, 3-EP, 4-EP, 4-*t*-BP, 4-*t*-OP, BPA, BPF, BPS, chlorobenzene,

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