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Chemically treated carbon black waste and its potential applications

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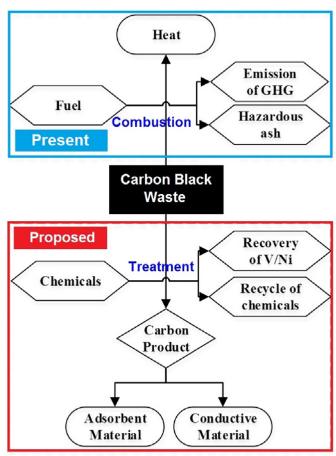
HIGHLIGHTS

- Hazardous impurities separated from carbon black waste with little damage to solid.
- Heavy metals were effectively removed from carbon black waste by HNO_3 leaching.
- Treated carbon black waste has high adsorption capacity (~356.4 mg_{dye}/g).
- Carbon black waste was also found to show high electrical conductivity (10 S/cm).

GRAPHICAL ABSTRACT



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ABSTRACT

In this work, carbon black waste – a hazardous solid residue generated from gasification of crude oil bottom in refineries – was successfully used for making an absorbent material. However, since the carbon black waste also contains significant amounts of heavy metals (especially nickel and vanadium), chemical leaching was first used to remove these hazardous impurities from the carbon black waste. Acid leaching with nitric acid was found to be a very effective method for removal of both nickel and vanadium from the carbon black waste (i.e. up to 95% nickel and 98% vanadium were removed via treatment with 2 M nitric acid for 1 h at 20 °C), whereas alkali leaching by using NaOH under the same condition was not effective for removal of nickel (less than 10% nickel was removed). Human lung cells (MRC-5) were then used to investigate the toxicity of the carbon black waste before and after leaching. Cell viability analysis showed that the leachate from the original carbon black waste has very high toxicity, whereas the leachate from the treated samples has no significant toxicity. Finally, the efficacy of the carbon black waste treated with HNO_3 as an absorbent for dye removal was investigated. This treated carbon black waste has high adsorption capacity (~361.2 $mg_{dye}/g_{carbonblack}$), which can be attributed to its high

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specific surface area (~559 m^2/g). The treated carbon black waste with its high adsorption capacity and lack of cytotoxicity is a promising adsorbent material. Moreover, the carbon black waste was found to show high electrical conductivity (ca. 10 S/cm), making it a potentially valuable source of conductive material.

1. Introduction

Oil refineries in Singapore generate carbonaceous solid residue of about 30 tons daily as an unavoidable by-product from gasification of crude oil bottoms. Although this carbonaceous solid residue has similar appearance to commercial carbon black and soot [1–3], it cannot be classified as either of them since carbon black is manufactured under controlled conditions for commercial use while soot is the by-product from combustion of carbon-based materials for energy and heat generation [4,5]. Therefore, this carbonaceous solid residue is denoted herein as a carbon black waste.

Carbon black is a carbonaceous material which has been commonly used as a reinforcing agent in rubber and tire industries [6,7]. Carbon black is also used as a coloring pigment, printing ink ingredient and adsorbent material [8,9]. With the wide applications of carbon black, there is strong demand for low cost and sustainable carbon sources for making carbon black [6,10]. The carbon black waste from oil refineries was found to have a carbon content of ~90 wt.%, which is slightly lower than the commercial carbon black (i.e. 97 wt.%) but higher than soot (i.e. 60 wt.%) [4,5]. Thus, carbon black waste is potentially a low cost and sustainable carbon source for making carbon black materials.

However, the carbon black waste obtained from oil refineries also contains high concentrations of heavy metals (especially nickel and vanadium) [11–13], which make it a highly hazardous material that can potentially cause environmental and human health problems if it is not properly disposed of or treated [2]. Currently, the carbon black waste produced from oil refineries in Singapore is disposed in incineration plants which are also known as waste-to-energy (WTE) plants. The heat generated from the combustion process is used to generate superheated steam in boilers and subsequently used to drive turbines to produce electricity. In addition to heat, incineration plants also produce (i) the normal primary products of combustion (CO_2 and water) as well as oxides of sulphur and nitrogen and other gaseous pollutants and (ii) nongaseous products include fly ash and bottom ash. As a consequence, incineration systems must have complex air pollution control systems in order to meet the required limits for protecting the quality of the ambient air and human health. Moreover, the produced ash must also be disposed properly as they usually contain high concentrations of heavy metals. Therefore, it is necessary to develop a new, effective and environmental friendly way for handling this carbon black waste.

In view of these considerations, re-utilization of the carbon black waste was investigated in this work. Instead of disposal by incineration, the carbon black waste was first treated to remove the hazardous impurities (i.e. heavy metals) producing non-toxic carbon material which was further used as an adsorbent material. The processes for recovery of the heavy metals (i.e. nickel and vanadium) in the leachate will be developed in our future work.

Several processes have been developed and used for removal or extraction of heavy metals from different types of solid waste, such as subcritical water treatment, leaching [14–17], chlorination [18,19] and electro-dialytic remediation [20]. Although the chlorination and electro-dialytic remediation methods show high extraction performance, these processes have high operating costs and their reaction conditions also constrain the recovery of the remaining solid. For instance, the high temperature (about 1000 °C) used in chlorination method [19] damages the structure of the remaining solid. Consequently, leaching process was considered the most suitable technique for carbon black waste as it can effectively remove heavy metals with minimum change in the physical properties of the carbon material [15,16]. There are two different types of leaching processes, which are (i) chemical leaching and (ii) biological or microbial leaching [21,22]. For instance, Mazurek [14] used citric acid to extract metals from spent vanadium catalyst and found that ~90% of vanadium and more than 60% of iron were extracted within 3 h at 50 °C. On the other hand, Bharadwaj and Ting [23] used bacterium (*Acidianus brierleyi*) to remove metals from spent catalyst. Nearly 100% extraction of Fe, Ni and Mo was achieved after 30 days, which is higher than chemical leaching using the same concentration of sulfuric acid as that produced by the bacteria. Although biological leaching showed high heavy metals extraction efficiency, these processes require long leaching times. Moreover, there is also difficulty in separating microorganisms from the remaining solid. Therefore, chemical leaching should be preferentially considered for extraction of heavy metals from the carbon black waste, and the selection of a suitable leaching agent is highly essential in achieving high extraction efficiency with minimum change in physical properties of the carbon material.

Numerous leaching agents, such as inorganic mineral acids (e.g. sulfuric acid, hydrochloric acid, and nitric acid [24–28]), organic acids (e.g. acetic acid and citric acid [29]), chelating reagents (e.g. ethylendiaminetetraacetate, EDTA [30]) and also alkaline solutions (e.g. ammonium and sodium hydroxide [31]), have been successfully used for extraction of heavy metals from the solid wastes like fly ash and slag. Alkaline leaching using ammonium solutions has the main disadvantage that only the amphoteric metals (e.g. Pd and Zn) can be dissolved in alkaline solution, whereas other impurities remain in the solid residue [27]. On the other hand, sulfuric and hydrochloric acids generally appear to be the most suitable leaching agents due to their low cost and high leaching efficiency although lead which may exist in the carbon black waste cannot be leached because of the formation of water-insoluble species, i.e. $PbSO_4$ or $PbCl_2$ [28].

Therefore, in this work, strong acid and alkali solutions (i.e. nitric acid and sodium hydroxide, respectively) were used as leaching agents to extract heavy metals, especially nickel and vanadium from the carbon black waste. The effect of leaching parameters on extraction efficiency and properties of the remaining solid carbon were discussed. As mentioned previously, the solid carbon remained after leaching was further investigated as an adsorbent for dye removal. The effects of adsorption parameters, i.e. adsorbent amount, pH solution, initial concentration and contact time, on the adsorption capability of the treated carbon black waste were studied. Moreover, other potential applications were also discussed.

2. Experimental section

2.1. Sample characterization

Carbon black waste (carbonaceous solid residue from gasification of crude oil bottoms) was collected from oil refineries in Singapore. A commercial activated carbon (Darco G 60) which