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A novel reutilization method for automobile shredder residue as an adsorbent for the removal of methylene blue: Mechanisms and heavy metal recovery using an ultrasonically assisted acid

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ABSTRACT

This study investigates the recovery of heavy metals (Zn, Cu, Mn, Fe, Ni, Pb and Cr) from automobile shredder residue (ASR) using an ultrasound-assisted acid. The discovered recovery efficiencies of the metals were in the following order: Zn (98.1%) > Ni (92.8%) > Mn (87.4%) > Cu (84.1%) > Pb (80.2%) > Cr (80.1%) > Fe (19.9%). The residue after the extraction of heavy metals was used to remove methylene blue (MB) from an aqueous solution. The chemical characteristics of the surface of the adsorbent were examined through X-ray diffraction, Fourier transform infrared spectroscopy (FT-IR), and point-of-zero charge determination. The adsorption kinetics of MB on the ultrasonically treated ASR (UTASR) with respect to the initial dye concentration, pH, adsorbent dose and temperature were investigated. The adsorption of MB on UTASR was assessed on the basis of equilibrium and kinetic studies. The adsorption kinetic data was well fitted to a pseudo-second order kinetic model. Equilibrium isotherms were analyzed by Langmuir and Freundlich isotherms. The Langmuir isotherm was found to be a better fit of the MB adsorption data. Ultrasound with nitric acid had a synergistic effect when it was used to extract heavy metals from ASR.

1. Introduction

Vehicles manufacturing have increased rapidly worldwide and demand is continually increasing as well. However, the number of end-of-life vehicles (ELVs) has also increased rapidly, in keeping with the manufacturing of new cars. In South Korea, ELVs are also increasing, and they are demolished by approved centers. The first step of ELV management is to remove hazardous fluids (e.g., motor oil, gasoline and cooling liquids) and hazardous or reusable parts (e.g., batteries, catalysts and tires). In the second step, the dismantled ELVs are then shredded and ferrous metals are magnetically removed for recycling (Singh and Lee, 2015a). The shredder residue remaining after the further separation of valuable materials by flotation, eddy-current separation and hand-sorting into different non-ferrous fractions is termed ASR (Van Caneghem et al., 2010). It has been reported that ASR is a highly heterogeneous mixture of residual ferrous and non-ferrous metals (5–23%), plastics (20–49%), rubber (3–38%), textile and fibre material (4–45%), wood (2–5%), and glass (2–18%)

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(Granata et al., 2011; Vermeulen et al., 2011). ASR accounts for approximately 10–25% of the initial mass all ELVs, but it is in fact disposed of in landfills (Van Caneghem et al., 2010). At present, energy recovery and material recycling from ASR in Korea stand at approximately 85%, out of which the energy recovery rate is within 5%. However, by 2015 this percentage may increase to 95% for ASR (of which the energy recovery rate is within 10%) (Sakai et al., 2014). Recent studies have confirmed that the fine fraction of ASR (<250 μm) contains a high concentration of heavy metals (Singh and Lee, 2015a, 2015b). Therefore, the effective recovery of heavy metals from ASR using ultrasonic power and the reuse of ASR are studied in this work.

Recent studies have demonstrated that ultrasonic power can enhance either the extraction or leaching rates of metals (Arain et al., 2008; Kazi et al., 2009; Huang et al., 2011). When ultrasonic power is applied, it causes acoustic cavitation, bubble formation and collapses to occur in solutions (Ashley, 1998; Kyllonen et al., 2004). During the breakdown of the bubbles, extremely high temperatures and pressures are generated at the boundary of the collapsing bubbles. Furthermore, the oxidative energy of hydroxyl radicals and hydrogen peroxide produced during the sonolysis of water can be very effective when used for the extraction of heavy metals from mixtures of solids and liquids (Ashley, 1998).

Many synthetic dyes are considered to be weakly biodegradable (Karagöz et al., 2008). Dyes discharged from the textile, rubber, plastics and electroplating industries pose a risk to the environment. The release of colored wastewater from these industries into surface water can be toxic to living organisms (Yasemin and Haluk, 2006). Due to their complex aromatic molecular structures, dyes are stable and are not readily biodegradable (Hameed and Ahmad, 2009). Therefore, their removal from water systems by an ecological and cost-efficient method is imperative. Methylene blue (MB) is widely used for coloring paper, as a temporary hair colorant, and in the dyeing of cotton and wool. It can also cause an increased heart rate, vomiting, shock, cyanosis, jaundice, quadriplegia, and tissue necrosis in humans (Hameed and Ahmad, 2009). Therefore, significant effort is being made to remove dyes from textile wastewater via adsorption techniques with ultrasonically treated ASR (UTASR) as part of the effort to reduce environmental problems related to effluent materials from the textile industry.

The objectives of this study are to recover heavy metals (Zn, Cu, Mn, Fe, Ni, Pb, Cd, and Cr) from the fine fraction of ASR with an ultrasonically assisted acid and to reuse the remaining residue for the removal of MB from aqueous solutions.

2. Materials and methods

Fig. 1 shows a schematic view of the process of the recovery of heavy metals from ASR and the reuse of residue for the removal of MB.

2.1. Sample collection and initial characterization

An ASR sample of approximately 7.0 kg was collected from an automobile shredder plant (Wonchang Junkyard Co., Gyeonggi-Do, South Korea). The ASR sample was sieved using a 250-μm sieve. The moisture content of the sieved sample was measured after drying it in an oven (Vision Scientific Co., Ltd., South Korea) at 105 °C for 24 h. The pH of the extraction solution for the fine fraction of ASR was measured by a digital

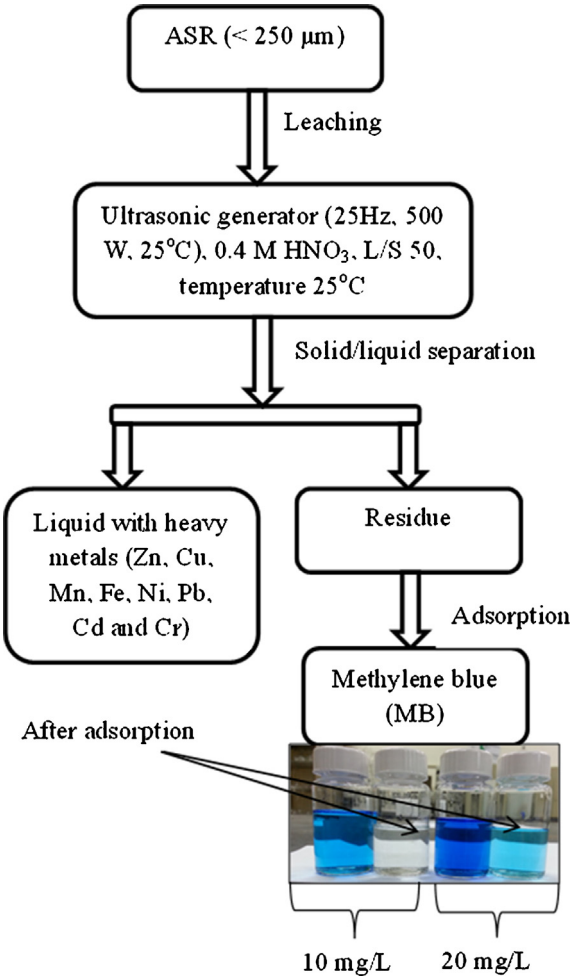

Fig. 1 – Schematic view of the recovery of heavy metals from ASR and the reuse of residue for dye removal.

pH meter (Orion 3-Star, Thermo Scientific) at a w/v ratio of the ASR sample to distilled water of 1:10). 10 g of the ASR sample was put into a 250 mL conical flask with 100 mL of distilled water and shaken for 2 h at 150 rpm in a water bath (SWB-35, Hanyang Scientific Equipment Co., Ltd., South Korea) at room temperature. An electronic muffle furnace (Jeio Tech Co., Ltd., South Korea) was used for the determination of the volatile solid and ash contents by ignition of the sample at 550 °C (Singh and Lee, 2015a). Inductively coupled plasma atomic emission spectroscopy (ICP–AES) (Perkin Elmer Inc., Optima 2000 DV) was used to measure the concentrations of Zn, Cu, Mn, Fe, Ni, Pb, Cd, Cr and Co after the digestion of 1.0 g of dried ASR sample with 20 mL of aqua regia (a mixture of HNO_3 and HCl at a ratio of 1:3) in a Teflon beaker using a hot plate. The digestion of ASR was performed in triplicate and the average values were determined. Before the ICP-MS analysis, all samples (50 mL) were boiled with 1 mL conc. HNO_3 at 200 °C for 3 h to remove organic-matter impurities from the samples.

2.2. Leaching experiment

The leaching of heavy metals from ASR was performed with 0.4 M of HNO_3 (liquid-to-solid (L/S) ratio—50 mL/g, and temperature—303 K). An ultrasonic generator (VCX-500, Sonics & Materials, Inc., USA) was used for the leaching of metals from the ASR. Ultrasound was applied to the ASR sample in a 1000 mL beaker through an immersed titanium probe. The ultrasound device used in this study had a frequency and