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Reduced zinc leaching from scrap tire during pavement applications

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ABSTRACT

Large quantities of scrap tires have been generated and accumulated over the years. However, a significant amount of them are stocked due to the lack of environmentally-friendly methods for disposing of, or reusing them. Because tires contain approximately 1–2% zinc by weight, leaching of zinc from scrap tires could be an environmental concern. In this research, we investigated the leaching of zinc from tire particles that are used with asphalt for pavement applications. The effects of tire particle size and pH on zinc leaching were also examined. Our results indicated that asphalt treatment significantly reduced zinc leaching from tires, and that it was also reduced by increasing the tire particle size and pH. The leaching of zinc was quantified by using a speciation-based modeling approach. The model parameters, namely, the total leachable zinc mass and the adsorption constant, can be used to predict the leaching of zinc under different conditions. The reduction in zinc leaching from asphalt-treated tire particles was due to the physical blocking of the tire surface by the asphalt. Results also indicated that, while the leaching of zinc and other selected toxic elements from untreated tire particles using simulated acid rain was not significant compared to the drinking water regulations, asphalt treatment during the pavement application further improved the environmental performance of the tire particles.

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1. Introduction

According to the U.S. Tire Manufacturers Association (TMA), approximately 300 million scrap tires are generated annually in the United States (U.S.) (TMA, 2017). Due to the durability, large quantity, and lack of effective mass recycling methods, scrap tires have become one of the most problematic wastes in the world (Adhikari et al., 2000). In the U.S., approximately 11.2% (about 27.5 million) of the scrap tires were disposed of in landfills in 2015 (TMA, 2017). Scrap tires not only occupy land space, but also impact environmental quality through leaching of organic and inorganic contaminants (Evans, 1997). Scrap tires could float to the top of landfills (U.S. EPA, 1991), provide habitat for mosquitos (Yee, 2008), and are nearly impossible to extinguish after being set on fire (IWMB, 2002).

Two major recycling approaches are used for scrap tires: material reuse and fuel derivation. The first approach includes using scrap tires on playgrounds and for artificial turf fields, or as an artificial reef or replacement for the aggregate in concretes or pavements, etc. (Llompart et al., 2013; Li et al., 2010; Collins et al., 2002; Youssf et al., 2014). For the second approach, the tires go through a pyrolysis process to produce diesel, and the residue from this process can be used as high performance coating, toner, printing inks, and activated carbon, etc. (Roy et al., 1999; Ucar et al., 2005; Pilusa and Muzenda, 2013; Gupta et al., 2011). In 2015, 25.8% of the total scrap tires produced in the U.S. were used as ground rubber, 7.0% were used for civil engineering applications, and 48.6% were used to produce fuel; the rest 11.4% were land disposed, and 7.1% were used for reclamation projects, exported, etc. (TMA, 2017). Although producing fuel appears to be a more attractive recycling option, the high temperature pyrolysis process also needs energy and produces carbon dioxide (CO_2). It was reported that the material recycling approach offers a greater lifecycle benefit than the fuel derivation approach does, especially for CO_2 reduction (Feraldi et al., 2013). Furthermore, it is a better option in terms of waste management (EPA, 2014).

While the use of scrap tires for ground rubber in playgrounds is one of the material recycling applications, the leaching of toxic organic and inorganic compounds from the tires could be a concern (Pierce and Blackwell, 2003; Selbes et al., 2015). According to the California's Office of Environmental Health Hazard Assessment (OEHHA), the tire rubber releases various types of toxic inorganic elements, including arsenic (As), barium (Ba), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), mercury (Hg), nickel (Ni), selenium (Se), antimony (Sb), lead (Pb), and zinc (Zn) (OEHHA, 2007). Scrap tire rubber also releases organic compounds such as PAHs, antioxidants, benzothiazole and derivatives (Llompart et al., 2013).

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The hydrophobic and non-polar properties of asphalt, which is extensively used in road construction (Roberts, 1991), make it an ideal waterproofing material for retarding water diffusion (Fang et al., 2009). These properties can be utilized to isolate scrap tires from water and, therefore, control the release of toxic compounds into the environment.

Zn accounts for 1–2% of the total weight of a tire, which can be leached into the environment (Rhodes et al., 2012). Although Zn is not listed in the National Primary Drinking Water Regulations (U.S. EPA, 2016a), it has a secondary standard of 5 mg/L (U.S. EPA, 2016b), and the excess consumption of Zn is still harmful because it suppresses the uptake of Cu and Fe (Kelleher and Lönnerdal, 2006). Zn also damages the stomach line (Bothwell et al., 2003), and is highly toxic to plants, invertebrates, and fish (Fosmire, 1990; Eisler, 1993), with a freshwater Criterion Maximum Concentration (CMC) of 0.12 mg/L (U.S. EPA, 1995).

Many factors, such as pH, particle size, and solid to liquid (S/L) ratio, could impact Zn leaching from tires (Gualtieri et al., 2005; Kanematsu et al., 2009; Hartwell et al., 1998; Rhodes et al., 2012; Wik et al., 2009). However, impacts of these factors on Zn leaching have not been well quantified. The objectives of this research were to understand the effect of asphalt on Zn leaching from tire particles during pavement applications, and to develop models to quantify Zn leaching from scrap tires resulting from the effects of asphalt treatment, particle size, and pH.

2. Theoretical aspects

2.1. Tire surface site acidity

We hypothesize that the surface of a tire particle contains multiple weak monoprotic acid sites, denoted as $\underline{S_i}OH$ (i = 1 − n). The deprotonation of $\underline{S_i}OH$ produces $\underline{S_i}O^-$. Based on our previous research, different surface site species are responsible for adsorbing differently charged metal species (Wang et al., 2004; Wang et al., 2007; Wang et al., 2008; Su et al., 2008; Su and Wang, 2011). Thus, the total surface site concentration, the acidity constant, and the solution pH are crucial in metal adsorption or the leaching process. A batch equilibrium acidimetric-alkalimetric titration method can be used to determine the surface site concentration and acidity constant of a sorbent particle (Wang et al., 2004).

The titration method assumes that the surface contains multiple (n) types of mono-protonic acid sites. The following equation can be used to express the relationship between acid/base consumption and a corresponding equilibrium pH (Wang et al., 2004):

$$\Delta V_{ss} = \sum_{i=1}^{n} \frac{V_0 S_{Ti} K_{Hi}}{C} \left\{ \frac{1}{[H^+] + K_{Hi}} - \frac{1}{[H^+]_0 + K_{Hi}} \right\} \quad (1)$$

where ΔVss is the net volume of the acid/base consumed by surface sites for protonation or deprotonation reactions (mL). It is the difference between the volume of acid/base added during the titration and that used for changing the water pH under the same ionic strength and pH. The volume of acid/base that is used to change water pH can be determined by titrating a water sample that has the same volume and same ionic strength as that used for particle titration. V_0 is the total volume of tire particle suspension used for titration (mL). S_{Ti} is the total surface site concentration of acid site *i* (M), which is the product of particle concentration (g/L) and surface site density (mol/g). K_{Hi} is the acidity constant of acid site *i* (M). C is the concentration of acid/base stock solution (M). $[H^+]$ is the hydrogen ion concentration in bulk solution (M). $[H^+]_0$ is the initial hydrogen ion concentration before any acid/base addition (M).

2.2. Chemical reactions

In the liquid suspension that contains tire particles and water, the following reactions could occur:

(a) Tire surface site deprotonation for site i:

$$\underline{S_i}OH = \underline{S_i}O^- + H^+, K_{Hi} \quad (2)$$

where $\underline{S_i}OH$ and $\underline{S_i}O^-$ are neutral and deprotonated surface site i, respectively; K_{Hi} is the corresponding acidity constant for $\underline{S_i}OH$ (M). Only the deprotonated surface site, $\underline{S_i}O^-$, is responsible for the adsorption of cationic or neutral species.

(b) Zn-hydroxide formation:

$$Zn^{2+} + OH^- = Zn(OH)^+; \beta_{(OH)_1} \quad (3)$$

$$Zn^{2+} + 2OH^- = Zn(OH)_2; \beta_{(OH)_2} \quad (4)$$

$$Zn^{2+} + 3OH^- = Zn(OH)_3^-; \beta_{(OH)_3} \quad (5)$$

$$Zn^{2+} + 4OH^- = Zn(OH)_4^{2-}; \beta_{(OH)_4} \quad (6)$$

where $\beta_{(OH)_1}$, $\beta_{(OH)_2}$, $\beta_{(OH)_3}$, and $\beta_{(OH)_4}$ are overall formation constants of $Zn(OH)^+$, $Zn(OH)_2$, $Zn(OH)_3^-$, and $Zn(OH)_4^{2-}$, respectively.

(c) Zn adsorption:

Using the formation constants of Zn-hydroxide species, we can calculate the speciation of soluble Zn under different pH conditions. Based on the calculation, the major soluble Zn species under an experimental pH range of less than 10 are Zn^{2+}, $Zn(OH)^+$, and $Zn(OH)_2$. The adsorption of these species by the deprotonated free surface site i, $\underline{S_i}O^-$, can be expressed as:

$$\underline{S_i}O^- + Zn^{2+} = \underline{S_i}O\text{-}Zn^+; K_{iS(OH)0} \quad (7)$$

$$\underline{S_i}O^- + Zn(OH)^+ = \underline{S_i}O\text{-}Zn(OH); K_{iS(OH)1} \quad (8)$$

$$\underline{S_i}O^- + Zn(OH)_2 = \underline{S_i}O\text{-}Zn(OH)_2^-; K_{iS(OH)2} \quad (9)$$

where $\underline{S_i}O\text{-}Zn^+$, $\underline{S_i}O\text{-}Zn(OH)$, and $\underline{S_i}O\text{-}Zn(OH)_2^-$ are surface complexes of free Zn ion, $Zn(OH)^+$, and $Zn(OH)_2$ with site i; $K_{iS(OH)0}$, $K_{iS(OH)1}$, and $K_{iS(OH)2}$ are the corresponding adsorption constants of different soluble Zn species (M^{-1}).

2.3. Metal adsorption modeling

If a metal concentration is relatively low compared with the total surface site concentration, the adsorption of free metal ion or metal hydroxide species is in the linear range of the Langmuir adsorption isotherm (Wang et al., 2004).

The concentrations of the two sorbable Zn-hydroxide species can be expressed as:

$$[Zn(OH)^+] = \beta_{(OH)_1}[OH^-][Zn^{2+}] \quad (10)$$

$$[Zn(OH)_2] = \beta_{(OH)_2}[OH^-]^2[Zn^{2+}] \quad (11)$$

The concentration of the deprotonated free surface site i, $\underline{S_i}O^-$, can be expressed as:

$$\{S_iO^-\} = \frac{K_{Hi}S_{Ti}}{[H^+] + K_{Hi}} = \alpha_{Hi}S_{Ti} \quad (12)$$

where $\{S_iO^-\}$ is the free surface site concentration of site i (M); α_{Hi} is the ratio of free surface site concentration to the total surface site concentration, $\alpha_{Hi} = K_{Hi}/\{[H^+] + K_{Hi}\}$; S_{Ti} is the total surface site concentration of site i (M).

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