Leaching Characteristics of Recycled Asphalt Shingles Mixtures

2013

Student Investigators: Jiannan Chen, Kyle J. Groves, Liangquan Wang
Advisors: Tuncer B. Edil, Ali Soleimanbeigi

University of Wisconsin-Madison
Leaching Characteristics of Recycled Asphalt Shingles Mixtures

Jiannan Chen
Graduate Research Assistant, Department of Geological Engineering, University of Wisconsin-Madison.
Email: jchen229@wisc.edu

Kyle J. Groves
Undergraduate Research Assistant, Department of Geological Engineering, University of Wisconsin-Madison.
Email: kgroves@wisc.edu

Liangquan Wang
Undergraduate Research Assistant, Department of Geological Engineering, University of Wisconsin-Madison.
Email: lwang263@wisc.edu

Tuncer B. Edil
Professor and Research Director, Recycled Materials Resource Center, Department of Geological Engineering, University of Wisconsin-Madison.
E-mail: tbedil@wisc.edu

Ali Soleimanbeigi
Research Associate, Department of Geological Engineering, Department of Geological Engineering, University of Wisconsin-Madison.
E-mail: soleimanbeig@wisc.edu

ABSTRACT:
Asphalt shingle waste has been identified by Environmental Protection Agency within top five priority material for reuse application. Use of RAS in hot mix asphalt is common but does not consume all available reclaimed asphalt shingles. Previous research results showed that recycled asphalt shingles (RAS) stabilized with fly ash (FA) or mixed with granular materials such as bottom ash (BA) have suitable geotechnical properties for use as structural fill in highway embankments opening new areas of application (Soleimanbeigi 2012). However, leaching of heavy metals from the coal ash mixed in the RAS could have the potential to contaminate the surrounding soil and groundwater. In this study, the leaching characteristics of heavy metals from different RAS/Coal Ash mixtures were evaluated. Long-term column leaching tests were conducted on the compacted RAS/BA (25/75 wt/wt), RAS:FA (80/20 and 90/10 wt/wt) mixtures, and the concentrations of heavy metals in leachate were compared to the USEPA drinking water standard and health advisory. From the results of column leaching tests, RAS/BA (25/75 wt/wt) has less environmental impact than the RAS/FA mixtures.

KEYWORDS: RAS, BA, FA, fly ash, leachate, temperature, season, long-term, column
INTRODUCTION
Recycling and reusing industrial wastes is an excellent way of saving energy, resources and reducing the greenhouse gas emission (Edil 2006, Lee et al. 2010). Nearly 80% of structures in the United States are covered by asphalt shingles and due to roof renovation, approximately 11 million Mg of asphalt roofing shingle waste is generated each year in the US. (Krivit, 2007). Re-roofing jobs account for 10 million Mg, with another 1 million Mg manufacturing scrap shingles. Applications including as a component of hot mix asphalt (HMA), cement kiln fuel, cold patch in paved roads and dust control in gravel roads account for reuse only between 10 to 20% of the total asphalt shingle waste and therefore the remaining large amount is disposed of in landfill (Townsend, 2007; Turley, 2011). Since highway construction usually consumes large volumes of material, the potential for using the asphalt shingle in other highway applications such as in unbound layers or fill is an appropriate option for reusing this material.

Typically, an asphalt shingle is made by impregnating a layer of organic or fiberglass material with liquid-blown asphalt. After being coated with proper thickness of asphalt, one side of the shingle is covered by granules in order to protect the shingle from being damaged physically and from the ultraviolet rays; the other side is covered by fine sand or fly ash to prevent the shingles from sticking with each other during packing and transport processes.

The asphalt shingle wastes are produced by either removing old asphalt shingles from the existing houses during house renovation process, in which they are called “tear-off shingle”, or rejecting asphalt shingle/shingle tabs that are discarded during the manufacturing process of new asphalt shingles, these shingle wastes are called “manufacturing shingle scrap”. Because RAS itself is too compressible to provide serviceable highway embankments, engineering properties of RAS mixed with granular byproducts, e.g., bottom ash (BA) and sand, and self-cementing fly ash (FA) are evaluated by Soleimanbeigi in 2012. It is suggested that geotechnical properties of compacted RAS/BA and RAS/FA mixtures are appropriate for structural fill application.

BACKGROUND
An important step to evaluate suitability of RAS as structural fill is to investigate its environmental impact. Use of coal ash raises environmental concerns related to heavy metal leaching (Li et al. 2006, Kossen et al. 1996, Kossen al. 2002, O’Donnell et al. 2010, Komonweeraket et al. 2011). Concentrations of leaching of Arsenic (As), Boron (B), Chromium (Cr) and Mercury (Hg) from fly ash and fly ash stabilized soils are observed in the laboratory.
batch tests to exceed the USEPA groundwater standard (Komonweeraket et al. 2011). To wisely and safely use RAS/additive mixtures as structural material, investigation of the leaching behavior is crucial.

In this study, RAS/Coal Ash mixtures are proposed to be used in construction of highway embankment. Physical properties of fly ash, and bottom ash including grain size distribution, specific gravity and maximum dry unit weight were evaluated. Previous studies on RAS showed limited information on the chemical properties and leaching of contaminants in unbound applications. Laboratory-scale column leaching tests on the RAS/Coal Ash mixture compacted to the expected field densities were performed to characterize the long-term leaching behavior of heavy metals.

TEST MATERIALS
The Resample used in this study was provided by Stratford Building Supply Company, Stratford, WI. The non-friable RAS samples were processed from tear-off roofing shingle waste to remove common impurities including nails, paper, plastic and wood chips. The percent impurities measured from the received RAS sample was less than 0.3% by weight. Fig. 1 (a) shows the typical shape of RAS particles, the sand cover and mineral coating on RAS particle surfaces. AS particles are plate-like, irregular in shape, highly angular and have rough surface texture. The angularity of RAS particles reduces to semi-round to round as the particle size decreases.

Self-cementing class C fly ash (FA) used for RAS stabilization was supplied by Columbia Power Plant, Portage, WI. Bottom ash (BA) was selected as a granular industrial byproduct additive to RAS. Fig. 1 (b) shows the microphotograph of the BA, internally porous, angular and rough in surface texture.

In this study, RAS was mixed with FA and BA respectively. RAS/FA ratio of 90/10 (wt/wt) and 80/20 (wt/wt) were selected for RAS stabilization, Andres/BA ratio of 25/75 (wt/wt) was selected for BA as granular mixture for RAS. The determination of RAS/Coal Ash ratio is based on both material strength and drainage considerations, the details are described in Soleimanbeigi (2012).

TEST METHODS
Physical Property Tests
The physical property tests, including grain size distribution, specific gravity, and compaction tests, were conducted on RAS, BA and FA samples.
Grain Size Distribution. The Grain size distributions of RAS, BA and FA samples were determined in accordance with ASTM D422. The specimens were first wet sieved through sieve No. 200 to separate coarse and fine particles. The coarse portions of BA and FA samples were oven dried (105°C) for 24 hours prior to mechanical sieving. The coarse portion of RAS sample was air dried to prevent binding of the particles at oven temperature.

Specific Gravity. The specific gravities of RAS, BA and FA were measured based on ASTM D854. A pycnometer is continuously agitated for an hour under a consistent vacuum to remove the entrapped air in the slurry and to prevent clogging by sample particles during the test. De-airing is accomplished by vacuuming distilled water.

Compaction. Standard Proctor compaction tests were conducted based on ASTM D698. RAS was mixed with FA at 80/20 and 90/10 wt/wt and with BA at 25/75 wt/wt.

Column Leaching Tests
RAS/FA (80/20 and 90/10) and RAS/BA (25/75) mixtures were used to conduct the long-term column leaching test (CLT). These specimens were compacted using the standard Proctor effort (ASTM D698) at 90% of the maximum dry unit weight (i.e., the field specification) and optimum water content into a PVC column, which has a diameter of 20 cm and a height of 10 cm. A schematic view of column setup is presented in Fig.2. All fittings in contact with specimens are non-metallic. Specimens with RAS/FA mixtures were cured 7 days right after compaction. Synthetic rainwater, as described in Scalia and Benson (2010) was used as inflow permeant. A continuous, upward flow was generated by a peristaltic pump at a Darcy flux of 1.6 cm/day (approximately 0.5 pore volumes of flow, PVF, per day), which was sufficient to avoid preferential flow paths, wet-dry cycles, and air bubbles in the system. Specimens were saturated 24 h before test initiation, and leachate samples were collected every day in the first 2 pore volumes of flow (PVFs), and twice per week afterward. The column tests were terminated at 20 PVFs, which is equivalent to approximately 4 to 5 years of permeation in the field (calculated from Chen et al. 2013).

Chemical Analysis
pH, Electrical Conductivity (EC) and Redox Potential (Eh) measurements of the leachate were recorded immediately after the sampling. 0.45μm filter paper was used for leachate sample filtering process. Samples were then preserved with trade-grade nitric acid (HNO₃) and stored at 4 °C. Metals that are listed in USEPA drinking water standard and health advisories,
i.e., Arsenic (As), Barium (Ba), Beryllium (Be), Boron (B), Cadmium (Cd), Chromium (Cr), Copper (Cu), Lead (Pb), Antimony (Sb), Selenium (Se), Thallium (Tl) and Zinc (Zn), were analyzed in this study. The maximum contamination level (MCLs) and method detection limits (MDLs) of the elements are listed in Table 1.

**TEST RESULTS**

*Physical Properties*

**Grain Size Distribution and Specific Gravity.** The grain size distributions of RAS, BA and FA particles are shown in Fig.3. More than 80% of the particles of RAS and BA are sand size with the fine contents less than 5%. RAS and BA have very similar grain size distributions. According to the Unified Soil Classification System (USCS), RAS is classified as well graded sand and BA is classified as poorly graded sand. RAS has a relatively low specific gravity (1.74), this low specific gravity is attributed to organic cellulose felt and asphalt cement contents which together constitute about 50% by mass of RAS. The specific gravity of BA is 2.67. The FA sample has 95.3% fines, and is classified as class C fly ash because of its self-cementing properties. The compositional properties of the fly ash are 6.0% loss on ignition (LOI), minimum 50% of SiO2, Al2O3, and Fe2O3, and minimum 75% of strength activity at 7 days. The specific gravity of this class C fly ash is 2.70 (Edil et al. 2006). The physical properties of RAS, BA and FA are summarized in Table 2.

**Compaction Test.** Fig.4 shows the compaction curves of RAS/FA (80: 20), RAS/FA (90/10) and RAS/BA (25/75). RAS/FA mixtures have well-defined compaction curves with γdmax varying from 12.5kN/m³ to 13.8kN/m³, and increasing FA content results in higher γdmax. The γdmax of RAS/FA (80: 20) mixtures is identical to γdmax of RAS/BA (25/75). The low γdmax of RAS mixtures makes the BA favorable alternative to compacted sandy soils for construction of highway embankments over weak subgrade.

**Column Leaching Tests**

**pH, Electrical Conductivity (EC) and Oxidation-Reduction Potential (ORP).** pH, EC and ORP of leachates as a function of pore volumes of flow (PVFs) are shown in Fig.5. All three specimens presented a trend of increasing pH and decreasing EC and ORP over the first 5 PVFs, and the values leveled off afterwards up to 20 PVFs. The pH and ORP of first flush (<2 PVFs) for all three specimens are similar and within a range of pH = 7.7 to 8.2 and ORP = 235.5 to 253.3 mV. The EC value of first flush from RAS/FA (80/20) (EC = 6570 us/cm) is much higher
than those of RAS/BA (25/75) and RAS/FA(90/10), while the EC values of RAS/BA(25/75) and RAS/FA(90/10) are very similar (2260 and 2380 us/cm).

After 5 PVFs, both RAS/FA mixtures have higher leachate pH and EC than RAS/BA mixture, with pH_{RAS/FA} = 8.7 to 11.4, EC_{RAS/FA} = 243 to 1003 us/cm compared with pH_{RAS/BA} = 7.4 to 8.2, EC_{RAS/BA} = 290 to 521 us/cm. Since FA is a cementitious material and also fine-grained material, it is more active than bottom ash when it comes to contact with water. The hydration process of FA generates alkaline substances, such as portlandite and brucite, which help enhance the leachate pH. Simultaneously, leaching of hydration products increases the total dissolved solids (TDS) in leachates and contributes to the higher leachate EC. Additionally, when compared both specimens containing FA, the specimen with higher FA content (i.e., 20/80) has higher leachate pH and EC, i.e., pH_{20\%FA} = 11.1 to 11.4, EC_{20\%FA} = 610 to 1003 us/cm compared to the specimen with lower FA content, i.e., pH_{10\%FA} = 8.7 to 10.8, EC_{20\%FA} = 243 to 456 us/cm. The leachate ORP values from all three specimens are within the range of -100 to 250 mV, with RAS/FA (80/20) has lower ORP (-100 to 29.5 mV) than the other two specimens.

**Heavy Metals Leaching.** The concentrations of metals in the leachate are shown in Fig.6, 7, 8 and 9 as a function of PVF. Exceedance of maximum contaminant level (MCL) for drinking water by As and Se in the leachates was observed during the first 10 PVFs for all three specimens. The peak concentration of As and Se happened during the first 5 PVFs with RAS/FA (80/20) having higher peak values than the other specimens. The peak value of As in leachate from RAS/FA (80/20) is 102.8 ppm, while RAS/FA (90/20) is 69.5 ppm and RAS/BA is 51.1 ppm. In the first 10 PVFs, the RAS/FA mixtures present a slightly higher As concentration in leachate than RAS/BA mixture, with AS_{RAS/FA} = 29.4 to 102.8 ppm, and AS_{RAS/BA} = 29.4 to 51.1 ppm. Moreover, higher FA content induced greater As leaching with a range of 38.1 to 102.8 ppm for RAS/FA (80/20) compared with 29.4 to 69.4 ppm for RAS/FA (90/10). After 10 PVFs, the As exceedance of MCL is not clear due to the limitation of analysis method. Since ICP-OES method is not quite sensitive to As, Sb and Tl in low level range (< 50 ppb), the MDLs of these three metals are all above their MCLs, which make the analysis doubtful. Although, the Sb and Tl concentration in the leachates are all under their corresponding MDL, exceedance is still possible. RAS/FA (80/20) leached much higher concentration of Se (312.2 to 941.3 ppm) during the first 2 PVFs than the other two specimens (68.6 to 90.5 ppm), which indicates the FA content also has great impact on the Se leaching. The Se concentration in leachate decreased as continuous flow through the specimens, and no exceedance was observed after 10 PVF.
No exceedance of Ag, Ba, Be, B, Cd, Cr, Cu, Pb and Zn was observed during the testing period (Fig.6, 7, 8 and 9). Concentration of Ag, Be, Cd, Cr and Pb in the leachate from the three specimens are all below or close to the MDL. The concentration of Ba and Cu in leachate presented a dependency on the fly ash content, i.e., RAS/FA (80/20) has higher Ba and Cu leaching than RAS/FA (90/10).

DISCUSSIONS
Exceedance of MCL by As and Se in first flush were observed from all three tested specimens. In the field, the heavy metal in the leachate could be naturally reduced by bioprocess, soil adsorption and dilution of rainwater or groundwater, which, to certain degree, can help relieve the environmental impact. However, to better evaluate the leaching behavior and contaminant transport, water sampling is highly recommended near the highway embankment. The leachate from RAS/FA (80/20) presented higher pH, electrical conductivity values and concentrations of As and Se than that from RAS/FA (80/20) and RAS/BA (25/75). The leaching of heavy metals from RAS/FA (80/20) and RAS/BA (25/75) are quite identical, however, the leachate pH values of both RAS/FA mixture are higher than the recommended value in the Secondary Drinking Water Regulations of USEPA Drinking Water Standards and Health Advisories which is 6.5 to 8.5 whereas RAS/BA does not produce alkaline substance to cause pH rise. Additionally, since (a) metal leaching from RAS/FA mixtures is FA content dependent, and (b) RAS content in RAS/BA (25/75) mixture is minority, RAS proved not to be a contributor to leachate quality, and the type and amount of coal ash admixture control the leachate quality. In conclusion, from an environmental perspective, RAS/BA (25/75) is a more benign mixture for highway construction.

CONCLUSIONS
Mixtures of recycled asphalt shingle (RAS) with bottom ash (BA) and fly ash (FA) were respectively tested in the laboratory. Three mixtures with different RAS/Coal Ash ratios and combinations were investigated: RAS/FA (90/10), RAS/FA (80/20) and RAS/FA (25/75). Laboratory column leaching tests were conducted on compacted specimens simulating field conditions. Leachate samples were collected periodically to track the effluent pH and released metals up to 20 pore volumes of flow. Based on the collected data, the following conclusions are drawn:
1. Exceedance of maximum contaminant limits (MCL) for drinking water by As and Se were observed from all three testing specimens during the first flush, while other elements listed in USEPA drinking water standard were either below the corresponding maximum
contaminated level or the method detection limit. Higher fly ash content results in enhanced heavy metal leaching, which may have environmental impacts.

2. RAS/FA mixtures have higher leachate pH than RAS/BA mixture, and the pH level is above the recommended range in Secondary Drinking Water Regulations of USEPA Drinking Water Standards and Health Advisories.

3. RAS indirectly proved not to be a contributor to leachate quality, since the type and amount of coal ash admixture is observed strongly affect the leachate quality.

4. RAS/BA (25/75) is a more environmental benign mixture out of the three tested mixtures, however, field water samples are still required to monitoring the water chemistry around the highway embankment.

ACKNOWLEDGEMENT
Support for this study was provided the University of Wisconsin System Solid Waste Research Program, TPF-5 (129) Recycled Unbound Materials Pool Fund and Recycled Materials Resource Center. Any opinions, findings, or conclusions expressed in this paper do not necessarily reflect the views of the sponsors. Special thanks to Professor Tuncer B Edil, James M. Tinjum, and Sabrina L. Bradshaw, lab managers Jackie B. Cooper and Xiaodong Wang, Undergraduate researcher Brigitte L. Brown, Geo-friends, and University of Wisconsin Madison.
REFERENCES


Table 1. Elements considered in column leaching tests

<table>
<thead>
<tr>
<th>Element</th>
<th>Instrument</th>
<th>MCL* (ug/L)</th>
<th>MDL (ug/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Antimony</td>
<td>ICP-OES</td>
<td>6</td>
<td>28.0</td>
</tr>
<tr>
<td>Arsenic</td>
<td>ICP-OES</td>
<td>10</td>
<td>28.6</td>
</tr>
<tr>
<td>Barium</td>
<td>ICP-OES</td>
<td>2</td>
<td>0.08</td>
</tr>
<tr>
<td>Beryllium</td>
<td>ICP-OES</td>
<td>4</td>
<td>0.11</td>
</tr>
<tr>
<td>Boron</td>
<td>ICP-OES</td>
<td>6000***</td>
<td>3.8</td>
</tr>
<tr>
<td>Cadmium</td>
<td>ICP-OES</td>
<td>5</td>
<td>0.53</td>
</tr>
<tr>
<td>Chromium</td>
<td>ICP-OES</td>
<td>100</td>
<td>0.3</td>
</tr>
<tr>
<td>Copper</td>
<td>ICP-OES</td>
<td>1000**</td>
<td>2.7</td>
</tr>
<tr>
<td>Lead</td>
<td>ICP-OES</td>
<td>15</td>
<td>3.8</td>
</tr>
<tr>
<td>Selenium</td>
<td>ICP-OES</td>
<td>50</td>
<td>44.2</td>
</tr>
<tr>
<td>Thallium</td>
<td>ICP-OES</td>
<td>2</td>
<td>14.2</td>
</tr>
<tr>
<td>Zinc</td>
<td>ICP-OES</td>
<td>5000</td>
<td>0.5</td>
</tr>
</tbody>
</table>

*USEPA drinking water standard and Health Advisories

**Secondary drinking water regulation

***Health Advisories - Life time
<table>
<thead>
<tr>
<th>Material</th>
<th>$d_{10}$ (mm)</th>
<th>$d_{50}$ (mm)</th>
<th>$C_u$</th>
<th>$C_c$</th>
<th>% Fines</th>
<th>$G_s$</th>
<th>USCS Classification</th>
</tr>
</thead>
<tbody>
<tr>
<td>RAS</td>
<td>0.17</td>
<td>1.1</td>
<td>7.6</td>
<td>1.6</td>
<td>3.8</td>
<td>1.74</td>
<td>SW</td>
</tr>
<tr>
<td>BA</td>
<td>0.19</td>
<td>0.9</td>
<td>6.3</td>
<td>0.8</td>
<td>1.9</td>
<td>2.67</td>
<td>SP</td>
</tr>
<tr>
<td>FA*</td>
<td>0.001</td>
<td>0.006</td>
<td>8.2</td>
<td>0.8</td>
<td>95.3</td>
<td>2.70</td>
<td>-</td>
</tr>
</tbody>
</table>

*From Edil et al. (2006)*
Figure 1. Photos of (a) RAS particles, the sand cover and mineral coating on RAS particle surfaces, (b) bottom ash.
Figure 2. Schematic view of column test apparatus
Figure 3. Grain size distributions of RAS, BA and FA samples.
Figure 4. Compaction curves of RAS/FA (80: 20), RAS/FA (90/10) and RAS/BA (25/75)
Figure 5. Leachate chemistry (a) pH, (b) EC and (c) ORP
Figure 6. Heavy metal leaching as the function of PVFs (a) Arsenic, (b) Barium, (c) Beryllium, (d) Cadmium
Figure 7. Heavy metal leaching as the function of PVFs (a) Chromium, (b) Copper, (c) Lead, (d) Antimony
Figure 8. Heavy metal leaching as the function of PVFs (a) Selenium, (b) Thallium, (c) Silver, (d) Zinc
Figure 9. Boron leaching as the function of PVFs