Executive Summary: Factors controlling the pH of recycled concrete aggregate (RCA)

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Introduction

The use of recycled concrete aggregate (RCA) as a substitute for natural, virgin aggregate in pavement construction applications is a wise choice of engineering material, as it is recognized as a readily available, mechanically sufficient, construction and demolition waste product with life-cycle economic and environmental benefits (Edil et al. 2012; "Recycling Concrete Pavements" 2009, Sanger et al. 2019). Environmentally safe and responsible applications of RCA must also consider the high alkalinity, high pH leachate as well as the heavy metal leaching risks (Abbaspour et al. 2016; Chen et al. 2013a; Engelsen et al. 2006, 2009, 2010, 2012, 2017; Gomes et al. 2016). RCA leachate generation from stockpiles and road base applications is unavoidable, and therefore it is of important to understand the fate and transport of the leachate, and whether pre-treatment, prescribed aging, or remediation is necessary to limit the environmental impact of RCA.

Generation of high pH leachate

High pH leachate is generated from the interaction of water with portlandite ($Ca(OH)_2$) and/or unreacted cement minerals that are exposed during the recycling process. In experimental systems with limited sample agitation, leachate pH increases from neutral to its maximum value in 6 hours, with the majority of that increase happening in the first minutes of reaction. Depending on how long it's been since the RCA has been crushed, the peak pH value is frequently between 10 and 12, with the maximum pH decreasing with increasing time since crush date. If water is left in contact with the RCA for 30 days, the leachate pH gradually decreases to pH 10 due to atmospheric CO_2 dissolution into the solution and reaction with the RCA mineral surface to generate $CaCO_3$.

Neutralization of high pH leachate

AASHTO drainage quality standards for roadbeds include excellent, (water is removed within 2 hours), good (1 day), fair (1 week), and poor (1 month). Upon the leachate's separation from the solid phase, high pH RCA leachate is rapidly neutralized by common, naturally occurring weak acids. Potential acids include atmospheric carbon dioxide and soil acidity, with soil acidity including acid bound to soil minerals, soil CO_2, and acid generated by the dissolution of common clay minerals. Independent of contact time, leachate in contact with atmospheric CO_2 is rapidly neutralized reaching a pH of 8 within 4-6 hours (Figure 1). Soils with high CECs (e.g., clay minerals) effectively neutralize high pH leachate by the dissolution of clay minerals, within a matter of minutes (Chen et al. 2020). Chen et al. (2020) demonstrates that common subgrade soils with moderate clay content will effectively neutralize high pH leachate initially produced by RCA for

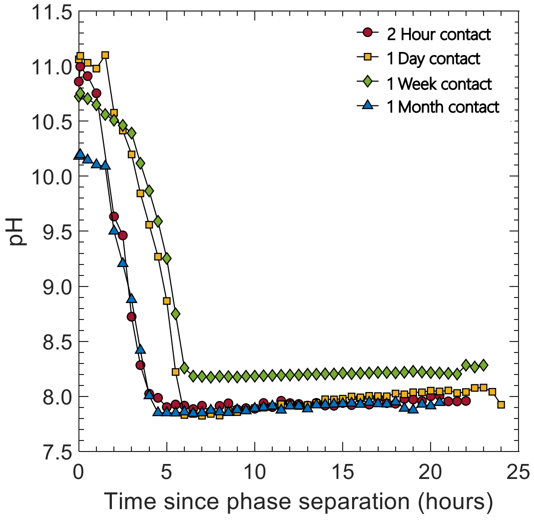

Figure 1. Change in pH of RCA leachate after separation from solid-phase RCA. Samples were exposed to RCA for 2 hours, 1 day, 1 week or month, and then stirred in contact with atmospheric CO_2.

decades and in many cases limits its migration to less than a meter in 50 years. Since, subbase layers under pavements are designed to drain water away from the road bed, leachate pH should be rapidly neutralized by common environmental acids.

Determining pH of RCA leachate

Commonly accepted experimental methods to determine the pH of leachate resulting from water contacting RCA are performed using a 10:1 water to solid ratio aggressively mixed in an end over end tumbler for 24 hours. Utilization of an end-over-end tumbler to determine material pH causes particle abrasion and removal of the protective calcium carbonate layer, again exposing uncarbonated matrix with reactive portlandite and cement phases. When assessing of leachate pH of stockpiled, aged, or otherwise carbonated RCA, particle abrasion and material handling during the measurement should be limited, additionally, the sample should be allowed to interact with atmospheric carbon dioxide. This will preserve the protective carbonate layers on the surface of RCA as a result of intermittent wetting and drying, which leads to a lower leachate pH (Sanger et al 2020), resulting in a more realistic leachate pH value.

Stockpiling Effects on Leachate pH

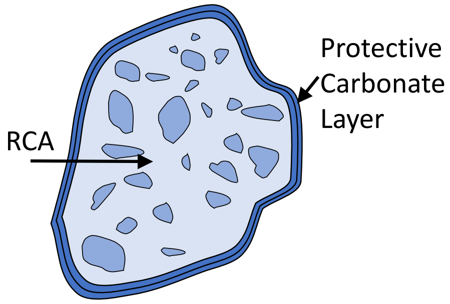

Figure 2. Schematic of protective calcium carbonate layer developed on RCA

Our comprehensive literature survey (Sanger et al. 2020a) of RCA leachate chemistry and recent experimental results (Ritchey et al. 2020, Sanger et al 2020b) support preliminary conclusions about the effect of stockpiling on leachate pH can be drawn. Crushing and material disrupt the protective carbonate later (Figure 2) that limits the direct contact of water with unreacted portlandite and unreacted cement phases. Immediately following crushing and material handling, water that comes into contact with the RCA will have an initial pH value greater than 11. As the stockpile is left undisturbed the protective carbonate layer will redevelop and leachate generated from this material will likely have a pH of ~10, although this process will likely take six months or longer. As long as water remains in contact with the RCA its pH will remain elevated (pH >9); however, once it is no longer in contact with the RCA the pH decreases to 7-8 due to neutralization by atmospheric CO_2. Contact with soils will likely result in more rapid neutralization. It is possible that addition of clay to the RCA stockpile could result in a lower leachate pH; however, this remains an area for further exploration. In order to minimize the environmental threat posed by high pH leachate RCA should be handled as little as possible. In recycling yards RCA leachate should be collected and treated to reduce the pH before discharge. At active construction sites care should be taken to limit the direct discharge of leachate to water bodies and high CEC clay barriers should be considered to retain the pH for 4-6 hours to allow neutralization by atmospheric CO_2 and/or dissolution of clay minerals.

Conclusions:

1. As long as water is contact with the RCA, it will have a pH value greater than 9. This cannot be avoided, water in contact with portlandite will rapidly increase to pH ~12, water in contact with $CaCO_3$ will have a pH value of ~9.5. This means that water will always start out at least at this level.
2. It may be possible to mix an "acid" in with the RCA to decrease this pH. Potential sources of acid could include clay minerals, aluminum sulfate or other additives; however, there are no published studies of these materials.
3. Once the leachate leaves the RCA, it is rapidly neutralized by common environmental acids. Road subbases are designed to transport water away from the road. CO_2 and clay minerals are potential neutralizing agents and are both are capable of rapidly (in a couple of hours) reducing the pH to ~7.
4. Several aspects of our research can directly inform RCA stockpile Management
    a. pH of water in contact with freshly crushed RCA will be higher than the pH of water in contact with RCA that has been exposed to atmospheric conditions (water and CO2)
    b. Every time the material is handled the protective carbonate layer that limits the pH of leachate to ~9.5 is disturbed. If this happens the leachate pH could be as high as 12.
    c. Leachate leaving stockpiles will be above pH 9.5, unless other acid sources are added
    d. Best practices for stockpiles would be to minimize materials handling and collect and control pH of leachate leaving the site. With a ~6 hour hold up, leachate will generally be at pH ~7.