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Martin O'Malley, *Governor*
Anthony G. Brown, *Lt. Governor*

State Highway Administration
Maryland Department of Transportation

James T. Smith, Jr., *Secretary*
Melinda B. Peters, *Administrator*

STATE HIGHWAY ADMINISTRATION

RESEARCH REPORT

ENVIRONMENTAL SUITABILITY OF RECYCLED CONCRETE AGGREGATE IN HIGHWAYS

Project Title: Development of Design Guidelines for Proper Selection of Graded Aggregate Base in Maryland State Highways
Task Order: SHA/UM/2-21

Ahmet H. Aydilek

UNIVERSITY OF MARYLAND, COLLEGE PARK

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16. Abstract

The use of recycled concrete aggregate materials in highway constructions as compared to the use of virgin materials reduces virgin natural resource demands on the environment. In order to evaluate their potential use of recycle materials in highway construction, two different recycled concrete aggregate (RCA) materials and two conventional graded aggregate base (GAB) materials and the mixtures of RCA-GAB materials were tested in the laboratory for their strength (California Bearing Ratio), resilient modulus, permanent deformation, and durability. In addition, laboratory water leach tests (WLTs), and pH-dependent leaching tests were conducted to determine the environmental suitability of RCA materials. The leaching behavior of Ca, Cr, Cu, Fe, and Zn from RCAs and the effects of pH, curing time, freeze/thaw cycles, liquid-to-solid ratio, and particle size on leaching of these metals were investigated. The summary resilient moduli (SM_R) of RCAs were 2.6 and 2 times higher than that of the natural GAB material and their stiffness increased when subjected to freezing and thawing cycles. Consistent trend could not be observed with CBR and SM_R values and percent RCA addition. Overall, GAB yielded lower permanent deformations compared to the RCA materials. In pH-dependent leaching tests, Ca showed cationic leaching patterns, while Cr, Cu, Zn, and Fe showed amphoteric leaching patterns. Leaching tests results indicated that leached Zn concentrations were below the detection limits at pH>5. Increasing curing time caused rehydration of cement particles and generally yielded a decrease in pH and leached concentrations of metals. Freezing and thawing of RCA also led to self-cementing and decreased pH, Ca concentrations while no consistent trends were found between the leaching of Cr, Cu, and Fe concentration and freeze-thaw cycles. Increasing the liquid-to-solid ratio decreased leaching of elements significantly due to the dilution of leached elements in the aqueous solutions.

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INTRODUCTION

Unbound aggregate materials are commonly used in the highway base/subbase construction to provide adequate mechanical support to the upper layer (asphalt layer) of pavement structure (Tutumluer and Pan 2008). A properly designed highway base layer would reduce the risk of the occurrence of asphalt fatigue cracking and rutting. Therefore, millions of tons of graded aggregate bases are used in the construction of highway base layers which cause an environmental problem and depletion of natural resources (Cetin et al. 2014).

Large amounts of recycled concrete aggregates (RCAs) are generated every year from the demolition of State highway concrete structures annually and part of these concrete wastes are being landfilled. In addition, disposed concrete waste contains high Ca content (cement) and it poses risk to clog the drainage system of the landfills due to potential occurrence of calcite precipitation (Mandal and Gupta, 2002). It is highly important to find a way to recycle these concrete waste materials. There is great potential for the use of recycled concrete wastes in highways. The reuse of RCA as compared to the use of virgin materials is economically feasible and reduces energy and need of virgin natural demands on the environment. However, State Departments of Transportation (DOTs) are still questioning the superiority of the mechanical performance of RCAs (Wen et al. 2010). Thus, there is a need to provide comparative information on the physical and mechanical behavior of conventional graded aggregate base materials (GABs) with respect to RCAs.

Furthermore environmental suitability of RCA materials used in such application has to be investigated even though it provides satisfactory mechanical performance. RCA may contain heavy metals due to additives such as fly ash, steel slag etc. used during the production of concrete and cement. The cement paste in the material is the primary source of the leached constituents (Engelsen et al. 2010), and certain cement additives like fly ash and steel slag may intensify this issue. Appreciable amounts of heavy metals from fly ash have been shown to leach (Cetin et al. 2012b, Cetin et al. 2013) and leaching of these constituents may have adverse effect to the environment (Cetin et al. 2014). Leaching of the metals from RCA is influenced by the chemical and physical properties of the material and environmental conditions. Several studies showed that RCA materials possess a great potential of leaching of certain metals, such as chromium (Cr), copper (Cu) and zinc (Zn) (Chen et al. 2013, Engelsen et al. 2009, and Engelsen et al. 2010) . Chen et al. (2013) also observed that leached Cr concentrations from the RCA materials used in their study also exceeded the Environmental Protection Agency Maximum Contaminant Level (EPA MCL), which is 0.1 mg/L. These studies show RCA leachate has critical concentrations of certain toxic metals, yet insufficient research regarding factors affecting leaching of RCA has been conducted. These factors include curing time, freeze-thaw, liquid-to-solid ratio, particle size and pH.

A battery of laboratory geotechnical tests was conducted on graded aggregate base (GAB) course material, recycled concrete aggregates (RCAs) as well as mixtures of GAB-RCA to determine the mechanical performance of these materials. California bearing ratio (CBR), resilient modulus, and permanent deformation tests were conducted to investigate the engineering properties of GAB, RCA and their mixtures, as well as the effect of curing time on RCA. The effects of winter conditions were also evaluated by performing resilient modulus tests on the RCA specimens after a series of freeze-thaw cycles. In addition, laboratory batch water leach tests (WLTs) were conducted to determine potential groundwater contamination due to leaching of heavy metals from RCA materials used in this study. Laboratory leaching tests investigated the effects of freeze/thaw cycles, curing time, liquid-to- solid ratio, pH, and particle sizes on the leaching behavior of metals leached from RCA materials.

MATERIALS

Two conventional graded aggregate base (GAB) materials and two recycled concrete aggregate materials were used in the current study. GAB materials contain coarse and fine aggregate particles as well as fines (clay and silt). Generally, the ratio between coarse and fine aggregate particles varies between 1:1 and 7:3. GAB and RCA materials were collected from different quarries in Maryland and tested in the laboratory. GABs and RCA materials used in the current study were named as: GAB1, GAB2, RCA-Plant A, and RCA-Plant B. All materials were classified as high quality base materials (A-1-a (0)) (AASHTO M-145) (Table 1). Figure 1 shows the grain size distributions of the materials. The fines contents of GAB1, GAB2 and RCA plant A and B materials were 7.6%, 10%, 6.4% and 4% by weight, respectively. Grain size distribution curves of GAB 2 and RCA Plant A were barely in the AASHTO M-147 limits (Figure 1). On the other hand, the gradation curve of the RCA Plant B material exceeded the AASHTO upper gradation curve limit indicating that it did not meet the AASHTO M-147 specifications (Figure 1)- . However, these specifications were designed for the use of regular GAB materials.

The specific gravity of GAB materials ranged from 2.55 to 2.83, while the range was 2.29 to 2.49 for the RCAs. The absorption values of both RCAs were significantly higher than the values for natural GAB materials (Table 1). The Los Angeles abrasion of the RCA Plant A sample exceeded the specification limit of 50%. Based on our laboratory testing, this number exceeds 50%. The percent losses based on sodium sulfate tests were 15.7 and 14% for RCA

Plant A and RCA Plant B, respectively, and exceeded the local department of transportation specification limit of 12% (MDSHA2013). The high percent losses could be due to a reaction of sodium sulfate with cement contents present in the material.

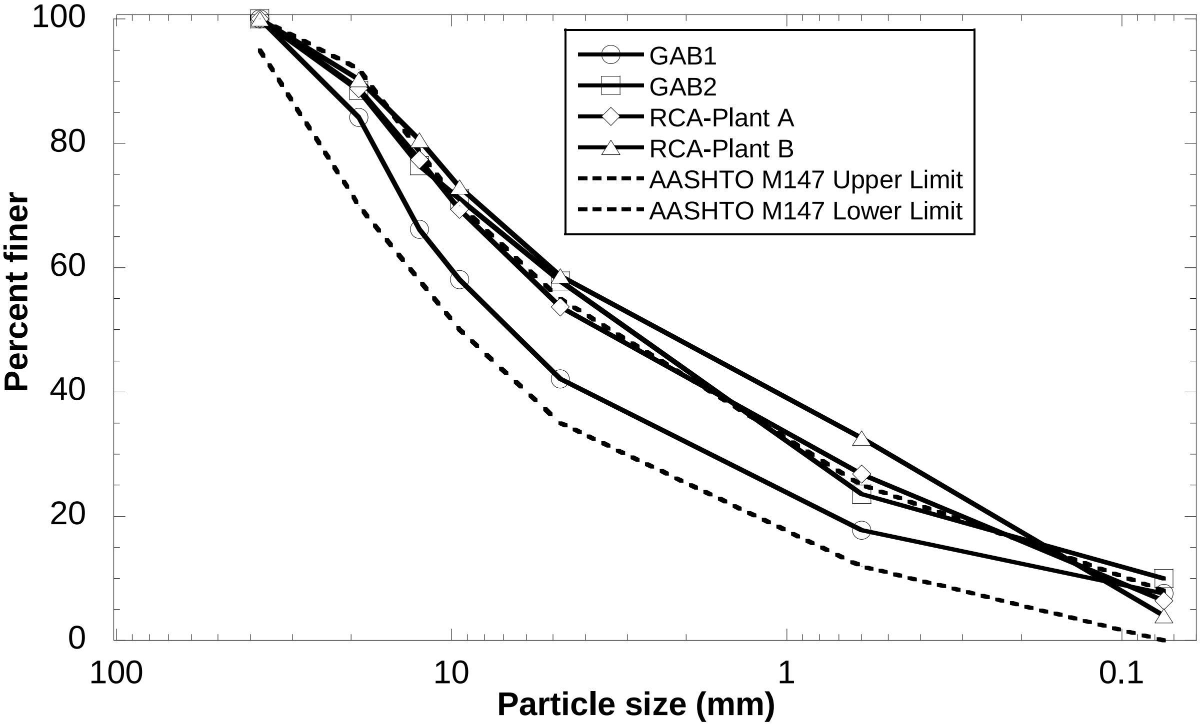

Figure 1. Particle size distributions of GAB and RCA materials

Table 1. Physical properties of the GAB and RCA materials

Material	Physical Properties									Soil Classification
	γ_{dmax} (pcf)	OMC (%)	G_s		Absorption		LA (%)	MD (%)	SS (%)	AASHTO
			F	C	F (%)	C (%)				
GAB1	158	4.7	2.55	2.77	5.33	0.78	16.4	22	1.6	A-1-a(0)
GAB2	158	4.5	2.72	2.83	3.09	0.55	23.6	7.6	1.1	A-1-a(0)
RCA-Plant A	128.5	9.5	2.29	2.49	9.23	4.20	55	17	15.7	A-1-a(0)
RCA-Plant B	127.8	9.5	2.29	2.53	9.05	4.19	47	18	14	A-1-a(0)

Notes: γ_d: maximum dry density, OMC: optimum moisture content, $G_{s:}$ specific gravity, F: fine aggregate, C: coarse aggregate, LA: Los Angeles abrasion, MD: Micro deval, SS: loss in sodium sulfate test.

Table 2. Chemical compositions and total contents of elements of the GAB and RCA materials.

Materials	pH	LOI (%)	Percentage by Weight (%)				(mg/kg)				
			SiO_2	Al_2O_3	Fe_2O_3	CaO	Ca	Cu	Fe	Cr	Zn
GAB1	6.7	10	38	10	7	6	41600	64	47000	1170	36
GAB2	8.5	45	2.4	0.7	1.3	29	215000	12	2300	7	33
RCA-Plant A	11.8	17	52	4.6	26	17	133164	30	19656	39	48
RCA-Plant B	10.4	13.4	61	4	1.9	13	95500	13	14580	19	40

LOI: Loss on ignition, NA: not applicable.

Table 2 shows the physical and chemical properties of the materials. The pH of RCA-Plant A and Plant B materials were 11.8 and 10.4, respectively. It is well known that there is a relationship between the pH of the materials and the CaO contents since dissolution of CaO causes an increase in pH (Cetin et al. 2012a). Similar relationship was observed between the pH and CaO contents of the RCA materials used in the current study as the RCA Plant A had higher CaO content by weight (~17%) which yielded higher pH (11.8) than the RCA Plant B with CaO content at 13% resulting in pH of 10.4 (Table 2).

METHODS

Laboratory Geotechnical Engineering Tests

GABs were blended with two RCAs at 75:25, 50:50, and 25:75 ratios by weight. The specimens for vibratory compaction was prepared in three equal layers using a vibration frequency of 55 Hz for 60±5 seconds per layer in accordance with ASTM D7382. All specimens were compacted at their optimum moisture contents (OMC). Table 1 provides the optimum moisture contents (OMCs) and maximum dry unit weights (γ_{dm}) of the materials. AASHTO T-193 and ASTM D 1883 were followed to conduct CBR tests. The strain rate was 1.27 mm/min during shearing process.

The procedure outlined in AASHTO T 307-99, a protocol for testing of highway base and subbase materials, was followed for resilient modulus tests. All specimens were compacted by vibratory compactor in split mold of 152 mm in diameter and 305 mm in height. Resilient modulus tests were performed on GAB, RCA and mixtures of GAB and RCA prepared at the same ratios of those tested for CBR. Each sample was compacted in six layers at their optimum moisture contents (OMC) and maximum dry densities using a vibratory compactor (ASTM

D7382). RCA specimens were removed from the molds after compaction, sealed in plastic wrap, and cured at 100% relative humidity and controlled temperature ($21 \pm 2\ ^{O}C$) for 1, 7 and 28 days before testing.

To observe the effect of winter conditions on resilient moduli, some of the mixtures were tested after a series of freeze-thaw cycles after compacted at their optimum moisture contents. After 28 days of curing, the specimens were frozen in a temperature chamber at $-23 \pm 1\ ^{O}C$ for 24 hours and then thawed in a humidity chamber at 100% relative humidity and controlled temperature ($21 \pm 2\ ^{O}C$) for 23 hours. The specimens were subjected to 1, 4, 8, 16, and 20 freeze-thaw cycles.

A Geocomp LoadTrac-II loading frame and associated hydraulic power unit system was used to load the specimens. The specimens were subjected to conditioning before the actual test loading under the confining and axial stress of 103 kPa for 500 repetitions. Confining stress was kept between 20.7 and 138 kPa during loading stages, and the deviator stress was increased from 20.7 kPa to 276 kPa and applied 100 repetitions at each step.

Resilient moduli from the last five cycles of each test sequence were averaged to obtain resilient modulus for each load sequence. This nonlinear behavior of unbound granular material was defined in this study using the model developed by Witczak and Uzan (1988)

$$M_R = k_1 p_a \left(\frac{\sigma_3}{p_a} \right)^{k_2} \left(\frac{\sigma_d}{p_a} \right)^{k_3} \tag{1}$$

where M_R is resilient modulus, k_1, k_2, and k_3 are constants, σ_3 is isotropic confining pressure, and σ_d is the deviator stress, p_a is atmospheric pressure. A summary resilient modulus (SM_R) was computed at a bulk stress of 30 psi (~208 kPa), following the guidelines provided in NCHRP 1-

28A. With few exceptions, high R^2 values (R^2 >0.9) were obtained from regression analyses performed on the model.

AASHTO T-307 test guidelines were followed to run the permanent deformation tests. During the permanent deformation test, same preconditioning load sequence of resilient modulus tests was followed. After the preconditioning stage, the specimens were subjected to 10,000 load repetitions under 103.4 kPa confining pressure and 206.8 kPa deviator stresses. Permanent deformation tests were performed until either 10,000 load repetitions were completed or the permanent deformation of the tested specimen was exceeded the original length of the specimen by 5%.

Laboratory Leaching Tests

The leaching tests used in this study were water leach tests (WLT), and pH-dependent leaching tests (pHLT). Preparation of the RCA materials was consistent for both tests: RCA materials were sieved through a U.S. No. 10 sieve (2 mm) (unless otherwise noted) and mixed with deionized water at the optimum moisture content (w_{opt}) (9.5% for both materials) in a polyethylene zip-top bag or a ceramic bowl. The samples cured for 1, 7 and 28 days in a moisture-controlled humidity chamber (21 ± 2 ^{O}C and 98% relative humidity). The zip-top bags were left open in this chamber during curing, and the ceramic bowls were covered with Parafilm® with several holes poked through it. Samples were weighed to the nearest hundredth gram on weighing paper, and reaction vessels were acid-cleaned 50 mL polypropylene centrifuge tubes.

At the end of the curing period, the samples were prepared at a differentliquid-to-solid (L:S) ratio of 10. This ratio was used per suggestion of Kosson et al. (2002) who claimed that it is the typical L:S ratio observed in natural environments. The influent solutions were prepared with 0.02M NaCl solution to provide stable reaction conditions for the release of metals (Cetin et al. 2012b). The tubes were rotated at 28±2 rpm for 72hours to reach equilibrium conditions. The samples settled for several minutes, the pH was measured, then the samples were filtered through a 0.2 µm pore size membrane disk filter using a 25 mm Easy Pressure syringe filter holder and a 60 mL plastic syringe into an acid-cleaned 50 mL centrifuge tube. The samples were acidified with 10% trace metal grade nitric acid (HNO_3) to a pH<2 and stored in a refrigerator at a temperature between 0°C and 4°C.

L:S ratios of 5:1, 15:1, and 20:1 were also studied to determine the effects of saturation level on leaching of metals from RCA materials. Moreover, effects of particle sizes on the leaching of metals were investigated using nine particle size fractions (9.5-4.75 mm, 4.75- 2.36 mm, 2.36-1.18 mm, 1.18-0.6 mm, 0.6-0.3 mm, 0.3-0.15 mm, 0.15-0.105 mm, 0.105-0.075 mm, and <0.075 mm). In addition, freeze/thaw (F/T) cycles were completed to study climatic effects. One F/T cycle involved freezing the samples at less than -18°C for 24 hours then thawing in the humidity chamber ($21 \pm 2\ ^{O}C$ and 98% relative humidity) for 24 hours. RCA samples were subjected to 4, 8, and 12 F/T cycles.

A PerkinElmer AAnalyst 100 atomic absorption spectrometer (AA) was used to quantify the metal concentrations of Ca, Fe, Cr, Cu, and Zn in the leachate samples. Minimum detection limits (MDLs) for AA were determined for each element. The MDLs for Ca, Cr, Cu, Fe, and Zn were determined as 2.5 mg/L, 2.1 µg/L, 3 µg/L, 4 µg/L, and 10 µg/L, respectively. These metals were considered important components of concrete and have been shown to leach from RCA materials (Engelsen et al. 2009 and 2010). Ca is the major metal in concrete, and Cr and Cu, Fe, and Zn have been designated EPA MCLs and/or secondary MCLs (SMCL) due to health risks and the potential to contaminate the environment.

RESULTS

Geomechanical Tests

CBR Tests

Table 3 shows the CBR results for GAB and RCA materials. CBR values presented for RCA Plant A and RCA Plant B were the CBR values measured after 1 day of curing mention figure number to represent the test data. The CBR of GAB2 was the highest (218) while the GAB1 resulted in the lowest CBR (68). RCA Plant A and RCA Plant B materials provided CBR values 2.18 and 1.67 times higher than the GAB1, respectively. On the other hand, the CBR of GAB2 was 1.47 and 1.91 higher than the CBR of RCA-Plant A and RCA-Plant B, respectively. CaO is the main mineral that initiates the pozzolanic reactions to improve the mechanical properties of cementitious materials (Cetin et al. 2010) and Table 1 indicates that CaO contents of GAB2, RCA-Plant A and RCA-Plant B are 29%, 17% and 13% while it is 6% for GAB1 which probably the main reason for GAB2 and RCA materials to provide higher CBR values than the GAB1 material.

Table 2. Chemical compositions and total contents of elements of the GAB and RCA materials.

Materials	pH	LOI (%)	Percentage by Weight (%)				(mg/kg)				
			SiO_2	Al_2O_3	Fe_2O_3	CaO	Ca	Cu	Fe	Cr	Zn
GAB1	6.7	10	38	10	7	6	41600	64	47000	1170	36
GAB2	8.5	45	2.4	0.7	1.3	29	215000	12	2300	7	33
RCA-Plant A	11.8	17	52	4.6	26	17	133164	30	19656	39	48
RCA-Plant B	10.4	13.4	61	4	1.9	13	95500	13	14580	19	40

Table 3. CBR, SM_R, power fitting parameters and plastic strain of the GAB materials

Material	**CBR**		**SM_R (psi)**	**Power model fitting parameters**			**$\varepsilon_{plastic}$ (%)**
				k_1	**k_2**	**k_3**	
GAB1	68		30,457	1025	0.88	-0.22	0.03
GAB2	218		16,534	1121	0.91	-0.16	0.04
RCA-Plant A	148 (1 day cured)	167 (7days cured)	42,786	355.8	1.40	-0.18	0.2
RCA-Plant B	114 (1 day cured)	131 (7days cured)	31,908	493.3	1.18	-0.13	0.1

Table 4. CBR, SM_R and power model fitting parameters of RCA/GAB mixtures.

RCA/GAB mixtures	**CBR**	**SM_R (psi)**	**Power model fitting parameters**		
			k_1	**k_2**	**k_3**
25A75GAB1	209	23,206	1430	0.82	-0.20
50A50GAB1	131	18,855	356.5	1.54	-0.17
75A25GAB1	154	40,611	478.1	1.45	-0.34
25A75GAB2	282	20,306	1495	0.80	-0.04
50A50GAB2	319	21,756	543.5	1.30	-0.10
75A25GAB2	301	37,710	492.3	1.29	-0.11
25B75GAB1	141	10,153	510.61	1.29	-0.18
50B50GAB1	194	17,405	450.63	1.27	-0.11
75B25GAB1	189	21,756	356.25	1.39	-0.21
25B75GAB2	NA	49,314	452.3	1.36	-0.05
50B50GAB2	NA	40,611	1689	0.68	-0.08
75B25GAB2	NA	17,405	2313	0.53	-0.18

Table 3 shows that the RCA specimens cured for 1 day resulted in lower CBR than those subjected to 7 day-curing. Poon et al. (2006) stated that unhydrated cement content retained within the adhered mortar was the cause of self-cementing in RCA used as unbound base. Table 4 presents the results of CBR tests performed on mixtures of RCA and GABs. GAB2-based mixtures resulted in higher CBRs but a consistent trend cannot be observed with CBR value and percent RCA addition.

Resilient Modulus

Average SM_R of two RAC and GAB materials are shown in Table 3. The SM_R values of RCA materials were higher than those of GAB materials. It was noted that the differences between the SM_R values of RCA materials and GAB1 were not as significant as they were with CBR test. However, the GAB2 materials had SM_R 1.85-2.6 times lower than the SM_R of other materials. These results indicated that the strength and stiffness of materials were not directly related parameters. Figure 2 shows that SM_R of all materials increases considerably with an increase in bulk stress, consistent with the findings of previous studies (Sweere 1990 and Cetin et al. 2010).

Resilient modulus tests were also performed on RCAs and mixtures prepared at varying RCA-to-GAB ratios. It can be seen from Figure 3 that 100% RCA and 100% GAB provide relatively higher M_R values as compared to their mixtures, with few exceptions. Similar observations were made by Kazmee et al. (2012) who attributed this behavior to poor packing of particles and change in gradation parameters. Figure 4 indicates that the SM_R of RCAs tend to increase with an increase in freezing and thawing cycles. Similar trends are reported by Bozyurt et.al (2011). The stiffness increase in the current study is attributed to the continuation of hydration (cementation) reactions in RCA during the freeze-thaw cycles.

Permanent Deformation

Granular materials exhibit permanent deformation if they are subjected to repetitive loading for extended periods of time. The permanent deformation values are dependent on rigidity, shear stress and load capacity of the granular materials. Use of the resilient modulus by itself is not sufficient to fully characterize the mechanical behavior of a pavement structure and should be coupled with permanent deformation tests (Khogali and Mohammad, 2004).

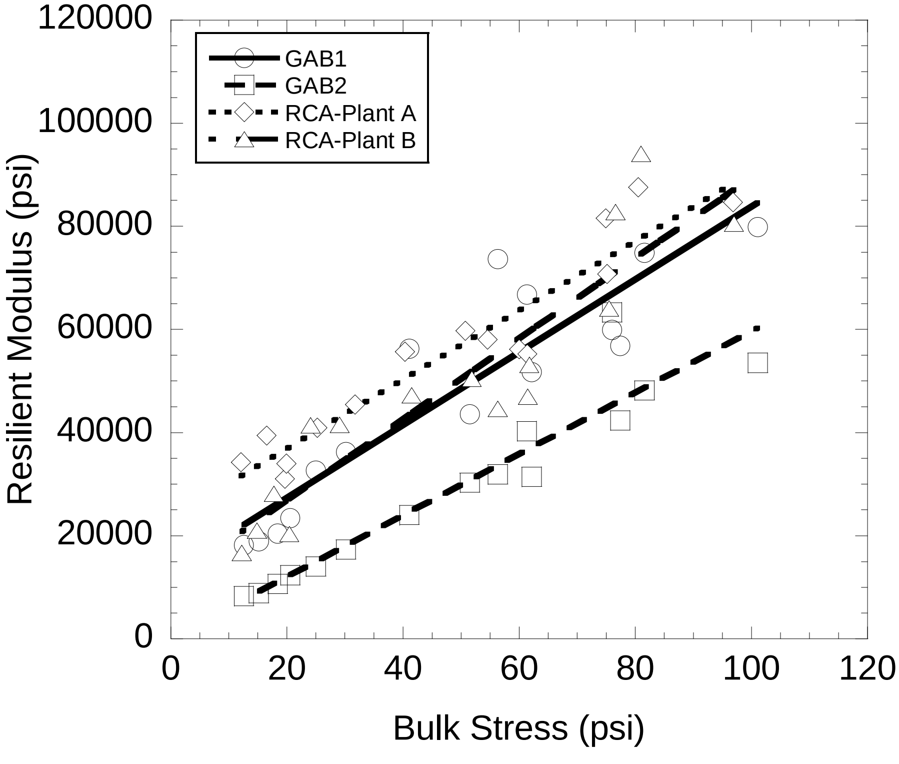

Figure 2. GAB and RCA resilient moduli at different loading sequences

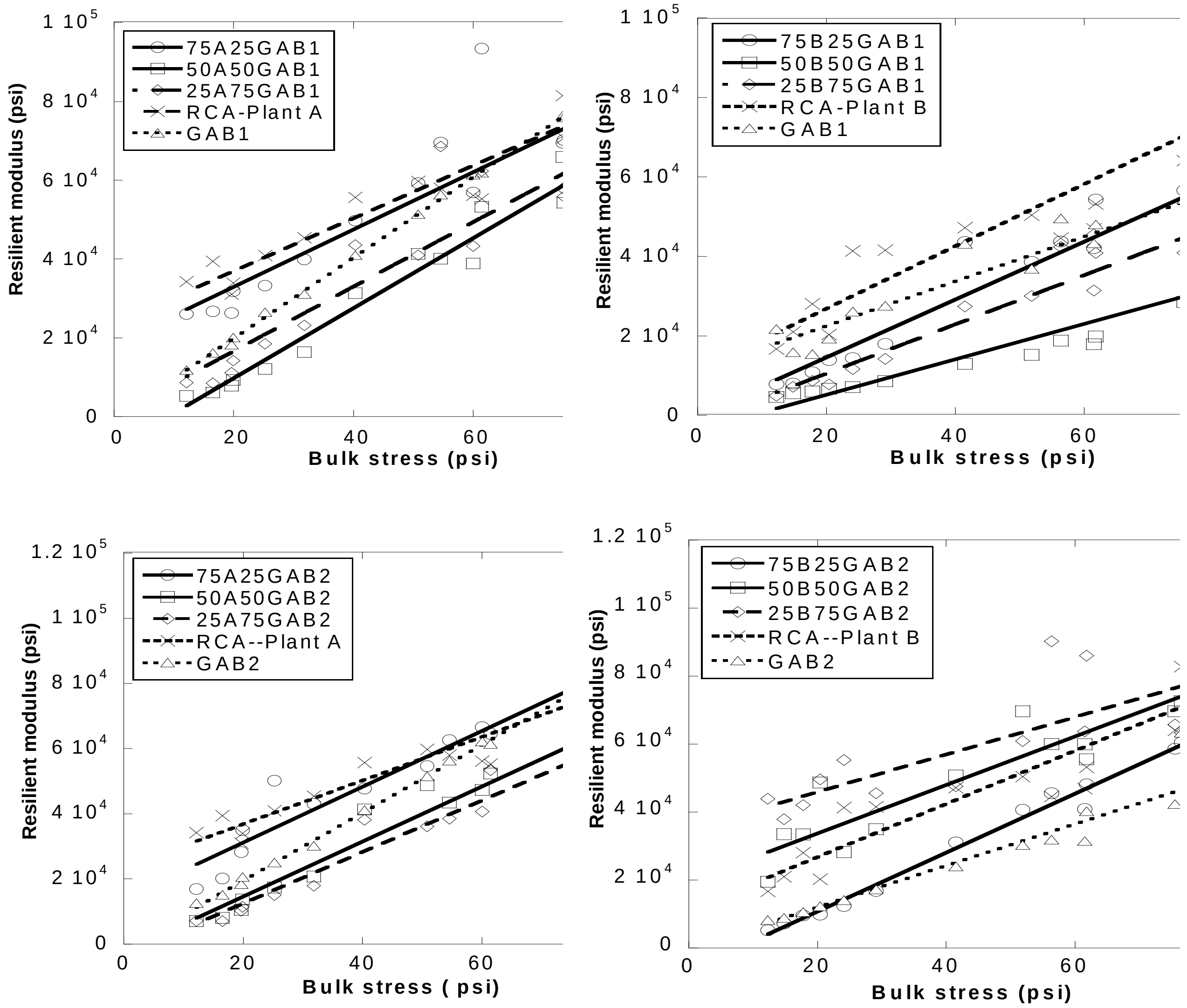

Figure 3. Resilient moduli of recycled concrete aggregate A and B, and RCA- GAB mixtures.

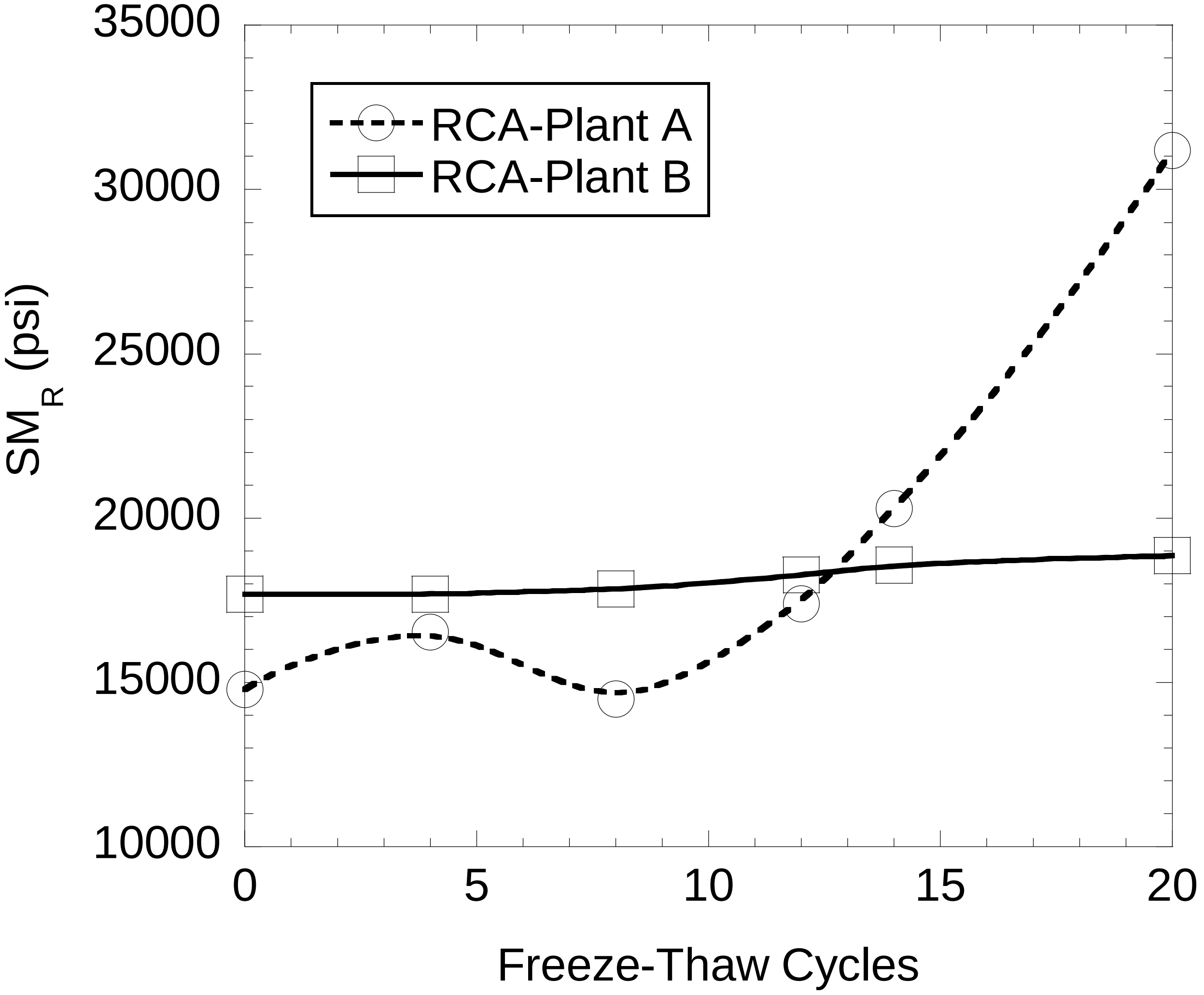

Figure 4. Effect of freeze-thaw cycles on SM_R of the two RCAs.

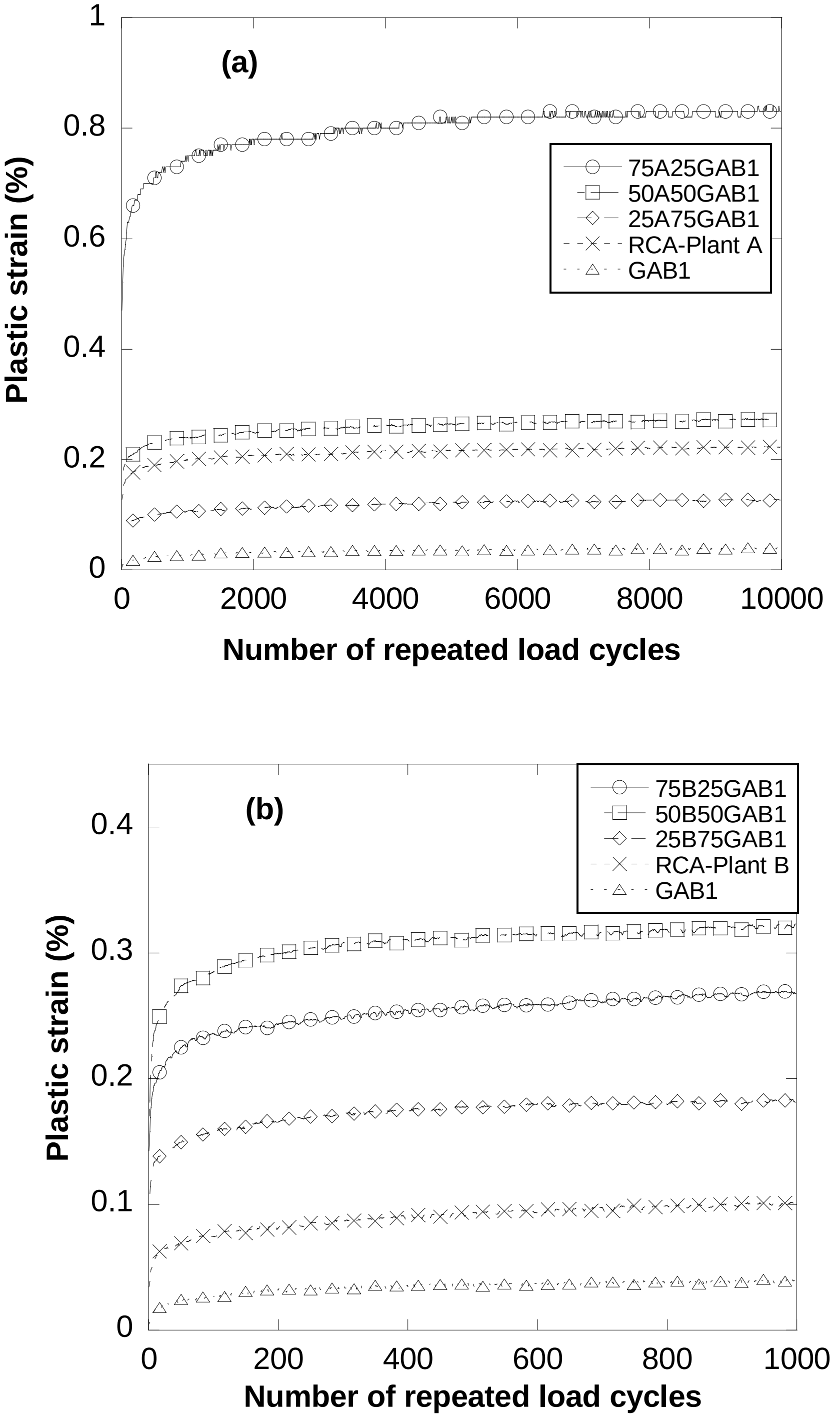

Figure 5. Plastic strain of (a) RCA-Plant A, and (b) RCA-Plant B , and their mixtures with GAB1.

Figure 5 shows the variation of cumulative permanent axial strain (plastic strain) with applied number of load repetitions. To model the relationship between the applied number of load repetitions and plastic strain, a power model was used:

$$\varepsilon_P = aN^b \tag{2}$$

where a and b are fitting parameters, ε_P is the cumulative permanent axial strain and N is the number load repetitions. The permanent deformation (i.e., plastic strain) depends on the packing arrangement of particles, grain size distribution, and particle contact area. Table 3 shows the plastic strain of all materials used in the current study. RCA Plant A had the maximum plastic strain (0.2%) while GAB1 had the minimum plastic strain (0.03%) after 10,000 repeated cycles of loading.

As shown in Figure 5, the permanent deformation of GAB increases upon mixing with RCA, suggesting higher likelihood of rutting of a pavement system built with GAB/RCA blends. Similar observations were made by Kazmee et al (2012). It was also noted that the plastic strain in individual GAB and RCA materials was less than that of their mixtures with an exception of 25A75GAB1, which may be attributed to poor packing arrangement of particles when these two materials were mixed.

Effect of Resilient Modulus on Highway Base Design

Resilient modulus test results were used to estimate the thickness of the base layer in a pavement by following the procedures defined in the AASHTO Guide (1993). The 50 million ESALwas assumed for this analysis. The detailed information about the analysis and the assumptions that were made to back-calculate the structural numbers (*SN*) can be found in Cetin et al. (2010). The laboratory-based SM_R values of GAB and RCA materials, summarized in Table 3, vary between 114 MPa and 295 MPa (30500 psi), which correspond to a base layer coefficient (a_2) of 0.06-0.18 according to AASHTO pavement design guidelines (1993). a_2 values of GAB1, GAB2, RCA-Plant A and RCA Plant B were 0.14, 0.08, 0.16 and 0.15, respectively. Finally, the base thicknesses were calculated using the following formula:

$$D_2 = \frac{SN - a_1 D_1 - a_3 D_3 m_3}{a_2 m_2} \tag{5}$$

where m_2 and m_3 are drainage modification factors for base and subbase layer, respectively, and were chosen as 1.2, 1.0, 0.8, and 0.6 for excellent, good, fair, and poor drainage conditions, respectively, within the pavement system (Huang 1993). D_1, D_2, and D_3 are the layer thicknesses of asphalt, base, and subbase layers, respectively. It can be concluded from Figure 6 that an increase in the base layer coefficient (a_2) yields a decrease in required base layer thickness while all other factors are kept constant. Furthermore, RCA materials require smaller base layer thickness than the GAB materials indicating that use of RCA could be more economical. On the other hand, the decrease in drainage modification factor increases the required thickness of the base course.

Leaching Tests

Batch Water Leach Tests-Effluent pH

Triplicate water leach tests were conducted and the averages of the triplicate test results were reported in this paper. Leaching tests were only conducted on RCA materials since they may pose potential environmental threat due to addition of supplemental materials such as fly ash.

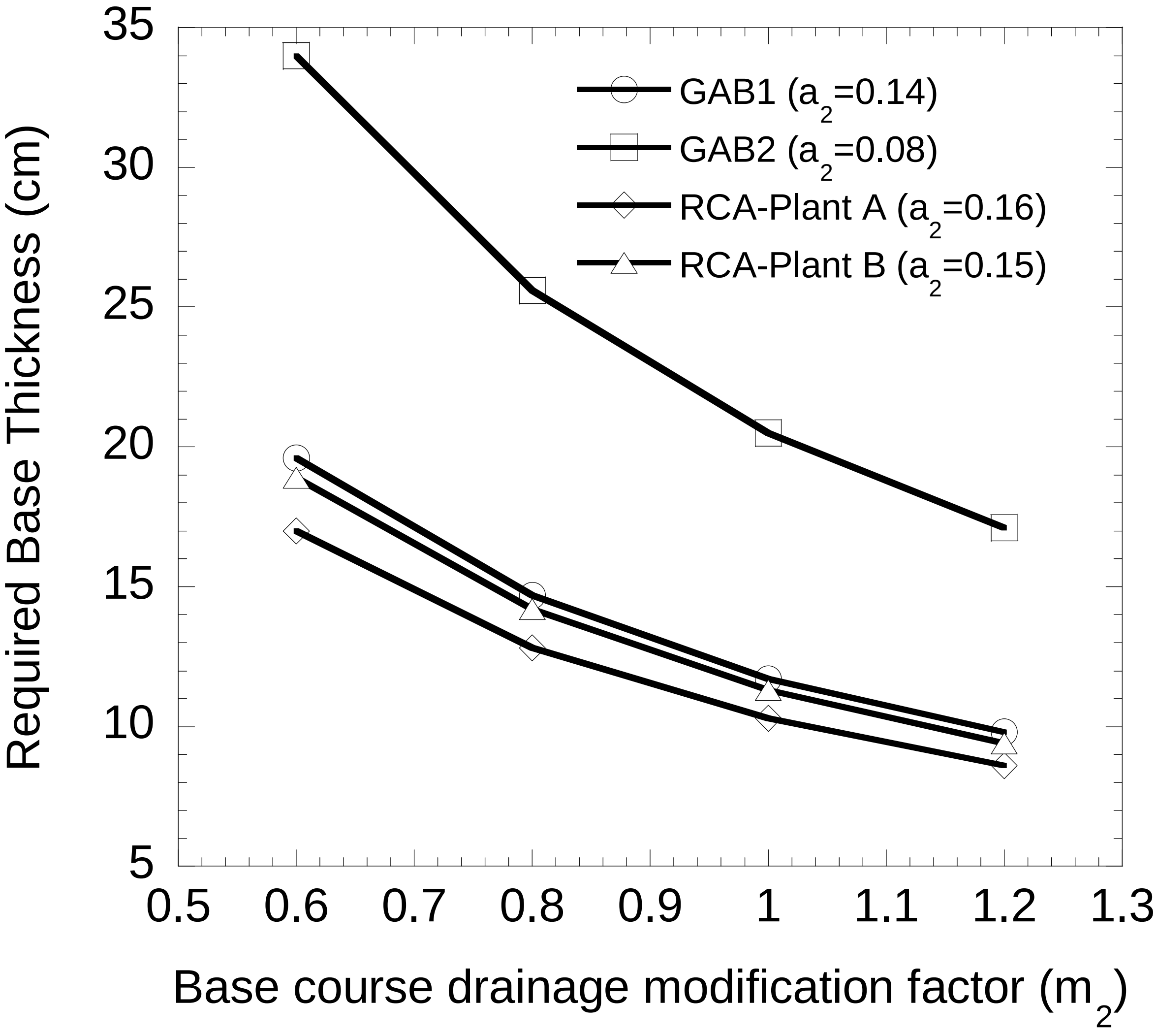

Figure 6. Effect of (a) base course drainage modification factor on required thickness,

steel slag etc. during the production of cement and concrete (Lewis et al. 2014). The core focus of this research included the effects of the following parameters on leaching of RCA: curing time (CT), freeze/thaw (F/T) cycles, liquid-to-solid (L:S) ratio, and particle size. Zn concentrations were below the detection limit at all WLTs. Therefore, no discussion regarding leaching of Zn is included in this section. Figure 7 summarizes the effects of CT, F/T cycles, L:S ratio and particle sizes on the effluent pH of RCA materials. The pH values indicated the RCA materials were highly alkaline (pH>10), which was consistent with the findings from previous studies (Engelsen et al. 2010, Chen et al. 2012). Overall, the effluent pH of RCA-Plant A material is higher than the effluent pH of RCA-Plant B material. This is consistent with the data provided in Table 2. CaO and total Ca amounts are the most important contents that contribute to the alkalinity of the aqueoussolutions (Cetin et al. 2014). Relatively higher CaO content (17%) and total Ca concentration (133164 mg/kg) of RCA-Plant A than the CaO content (13.1%) and total Ca concentration (95500 mg/kg) of RCA-Plant B probably yielded higher effluent pHs.

All RCAs were subjected to 1 day, 7 days, and 28 days curing times. Effluent pHs of RCA-Plant A and RCA-Plant B after 1 day and 28 days were reduced by 2% and 10%, respectively. This decrease in the effluent pH is most probably due to rehydration of Ca minerals in RCA aggregate matrix which reduced the leached Ca concentrations and ultimately the pH. Similar trends in the effluent pHs were observed when RCA materials were subjected to 4, 8 and 12 freeze/thaw cycles. The pH of RCA materials decreased with freeze/thaw cycles. The pH of the leachates decreased by an average of 3%. It is speculated that curing of the RCA materials continued during freeze/thaw process. This is consistent with the results of geomechanical test section as mentioned before. Stiffness of RCA

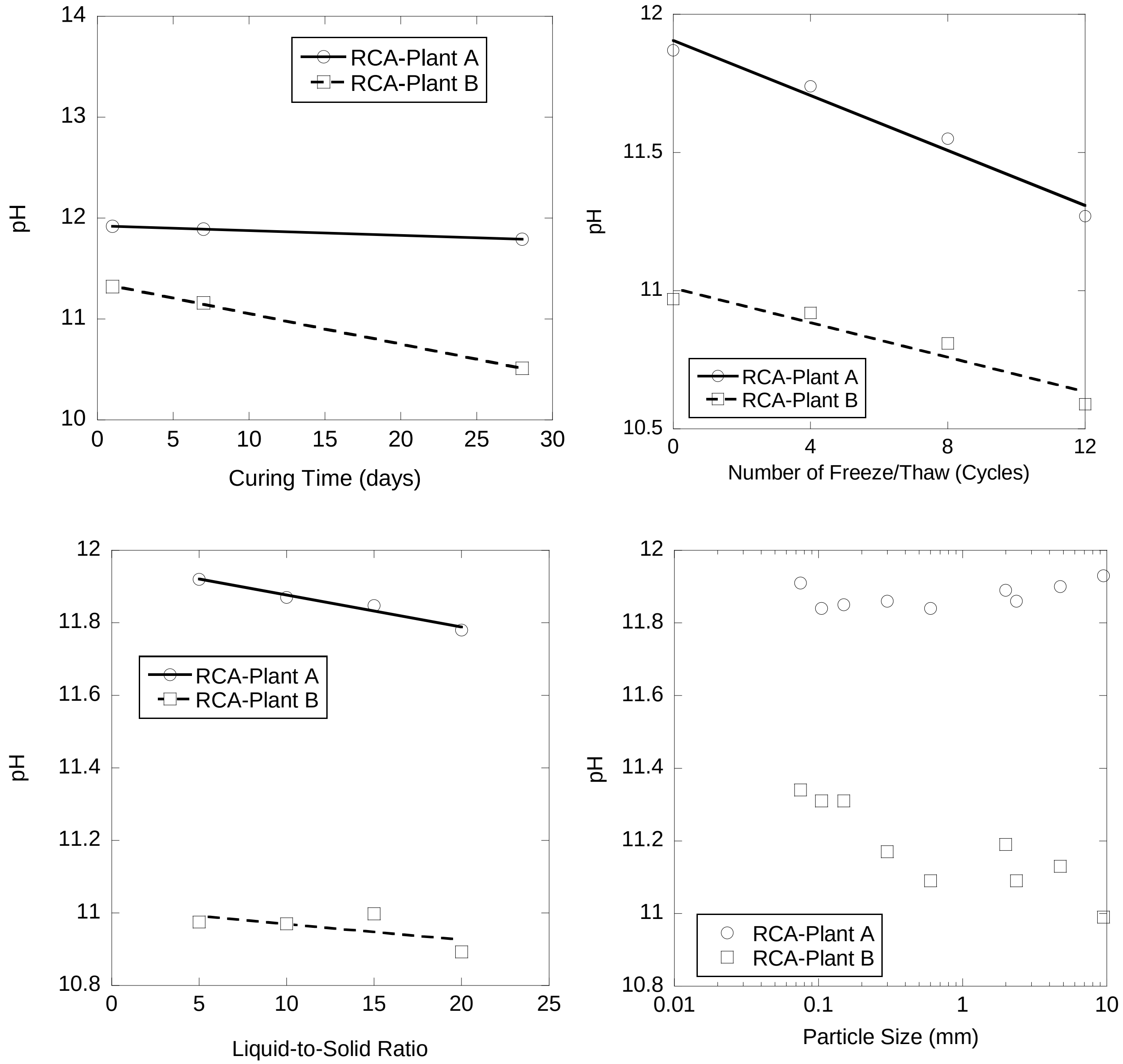

Figure 7. Effects of curing time, freeze/thaw cycles, liquid-to-solid ratio, and particle size on effluent pHs.

materials kept increasing which is an indicator of ongoing rehydration process. Sanchez et al. (2009) also found similar findings and claimed that precipitation of Ca as $CaCO_3$ could be a reason for the observed decrease in effluent pHs after materials were subjected to freeze/thaw cycles. Figure 7 also shows that the effluent pH of RCA materials decrease with an increase in L:S ratio. This was an expected behavior due to dilution of Ca minerals when L:S ratio was increased from 5 to 20. Furthermore, effects of particles size on the leachate pH of RCA materials were also investigated. As shown in Figure 7, the pH of RCA-Plant A materials was relatively stable for each particle size fraction indicating the leachate pH of these materials is more dependent on the chemical compositions of RCA-Plant A material itself rather than particle size. On the other hand, there is a slight decreasing trend in the effluent pH of RCA-Plant B material with an increase in particle size. This result showed that finer RCA-Plant B particles may possess higher CaO and total Ca content, which cause an increase in pH when the particles are finer.

Leaching of Ca, Cr, Fe, and Cu

Curing Time

Curing time is a very important parameter that has to be considered for RCA materials when used as base/sub-base materials in highway construction as increased curing time allows RCAs to reach designed geomechanical strength. Figure 8 illustrates the effect of curing time on leaching of the RCA materials. Results showed that Ca, Cr and Cu concentrations decreased with an increase in curing time while Fe showed initial increases followed by slight decreases. Allowing the RCA samples to cure for a longer period of time may increase the rehydration rate of cement particles in RCA which may eventually yield encapsulation of particles. Thus, this may also contribute on immobilization of metals that are attached to surface of RCA particles. The self-cementing process may not have fully completed after 1 day of curing which could be the main reason to observe initial increase in the leached Fe concentrations.

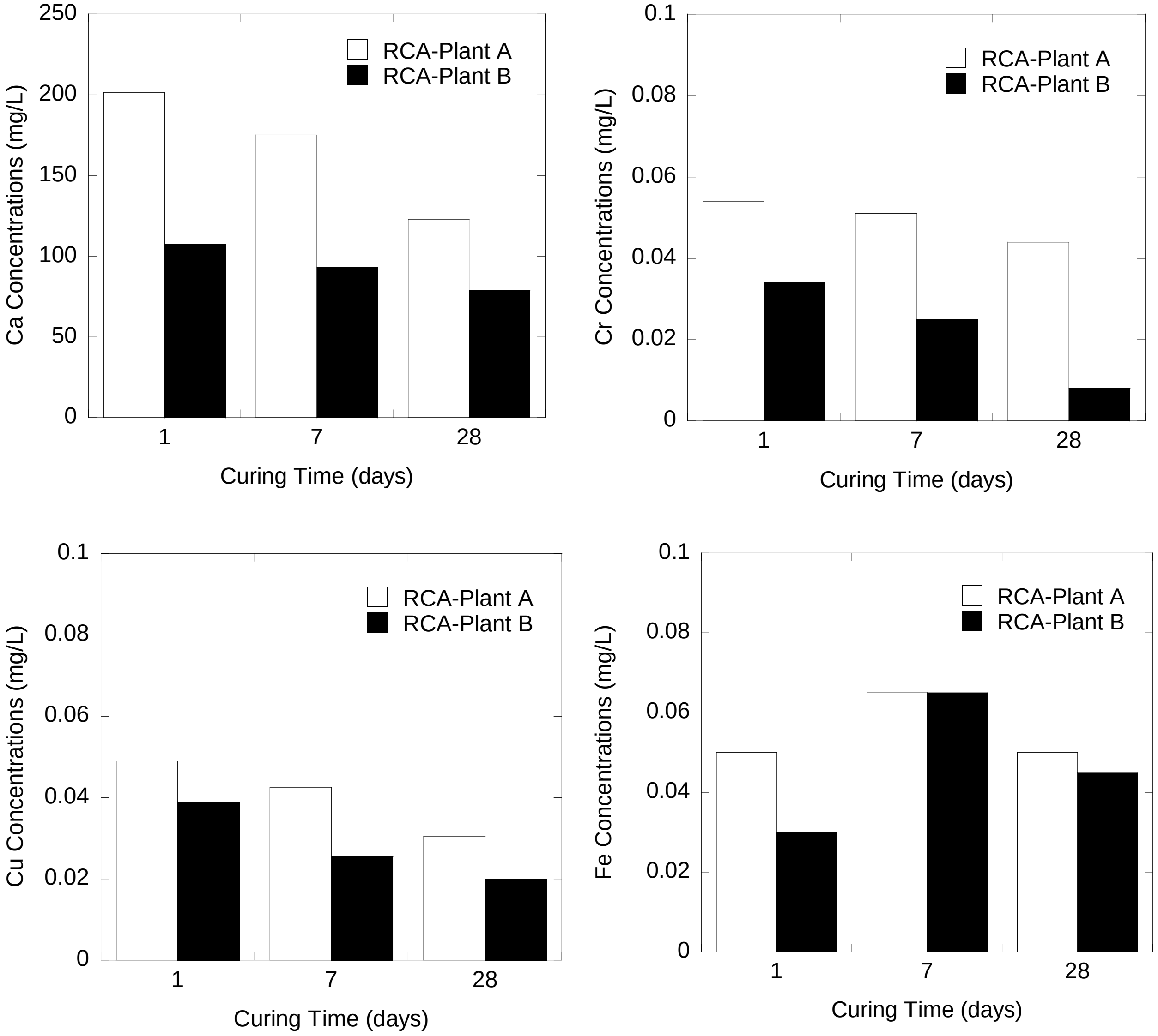

Figure 8. Effects of curing time on leaching of Ca, Cr, Cu, and Fe from RCA.

Leached Ca concentrations varied from 201 mg/L to 122 mg/L and 107 mg/L to 70 mg/L for RCA-Plant A and RCA-Plant B. The percent reduction (14%) in the leached Ca concentrations from RCA-Plant B was consistent across all curing times. However, the percent decrease in Ca concentration leached from RCA-Plant A was higher when samples were cured for 1 day and 7 days (12.5%) than the other ones cured for 28 days (30%) (Figure 8). Leached Cr and Cu concentrations ranged from 0.054 mg/L (RCA-Plant A) to 0.008 mg/L (RCA-Plant B) and 0.049 mg/L (RCA Plant A) to 0.02 mg/L (RCA-Plant B). Furthermore, leached Fe concentrations ranged from 0.065 mg/L (both RCAs) to 0.03 mg/L (RCA-Plant B). Cr and Cu concentrations did not decrease at a consistent rate pattern between curing times, which is partially due to differences in metal content based on total elemental analysis (Table 2). Fe concentrations increased until RCA materials were cured for 7 days then started decreasing again. Overall, metal concentrations leached from RCA-Plant A were consistently higher than the ones leached from RCA-Plant B. These results show that there was a relationship between the total metal content of RCA materials and leaching potential of these metals. As shown in Table 2, RCA-Plant A material contains higher metal content (Ca: 133,164 mg/kg, Cu: 30 mg/kg, Fe: 19,656 mg/kg, Cr: 39 mg/kg) compared to the metal content of RCA-Plant B (Ca: 95,500 mg/kg, Cu: 13.1 mg/kg, Fe: 14,580 mg/kg, Cr: 19 mg/kg).

Freeze/Thaw

Highways are continually exposed to climatic conditions during their service life, and these conditions can alter the physical and chemical properties of pavement systems significantly. Therefore, this study investigated the effect of climatic conditions on the leaching of metals from RCA materials. All specimens were subjected to 4, 8, and 12 Freeze/Thaw (F/T)

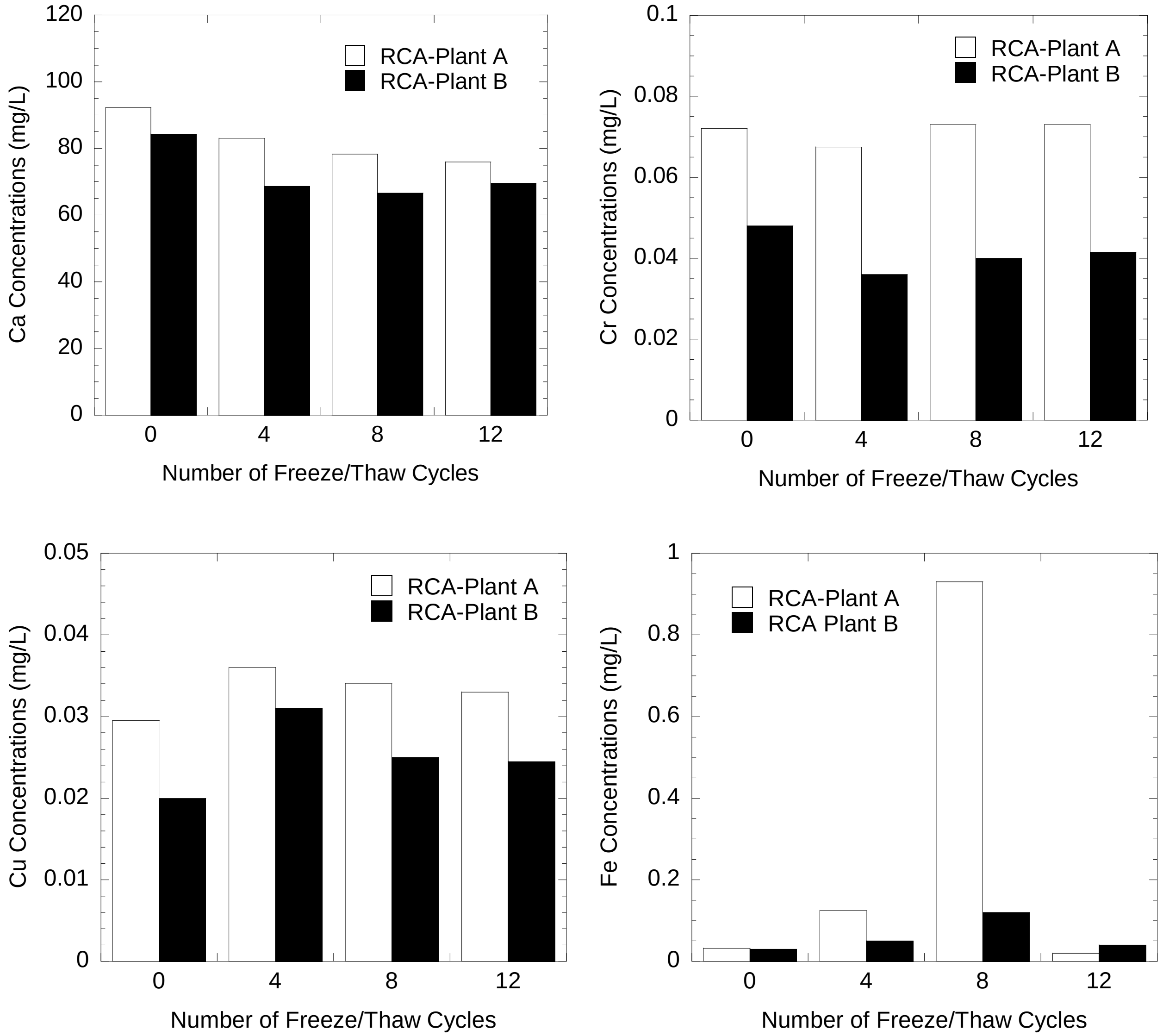

Figure 9. Effects of freeze/thaw cycles on leaching of Ca, Fe, Cr and Cu from RCA.

cycles. Figure 9 shows that Ca concentrations in the effluent solutions of RCA materials decreased with an increase in the number of F/T cycles while no consistent trend was observed for leached Cr, Cu, and Fe concentrations with F/T cycles.

Figures 7 and 9 show that variation of both leachate pH and leached Ca concentrations with F/T cycles illustrate similar leaching pattern indicating that effluent pH was directly controlled by the leached Ca concentrations. Similar findings were also observed on the previous studies (Engelsen et al. 2009, and Sanchez et al. 2009). It is concluded that precipitation of leached Ca concentrations as $CaCO_3$ due to carbonation may have contributed to the observed consistent decrease in Ca concentrations with an increase in F/T cycles.

Leached Cu concentrations increased for the RCAs subjected to 4 F/T cyclesbut decreased at F/T cycles greater than 4. A similar trend was also observed for leached Fe concentrations but these concentrations decreased when the materials were subjected to 12 F/T cycles. Rehydration of cement particles is a long process and takes times to be fully hydrated. Rehydration of RCA materials used in this study continued during F/T cycles and yielded an initial increase in the leaching of Cr and Fe. After rehydration process (8 F/T cycles) was almost complete, leaching of these two metals started decreasing.

Leaching concentrations of Cr decreased slightly when it was exposed to 4 F/T cycles and they were stabilized after being subjected to 8 number F/T cycles. Self-cementing processes were not as significant in leaching behavior of Cr in both RCA materials compared to the other metals fitting in this trend. These results showed that leaching of Cr from these two RCA materials were not affected significantly by F/T cycles.

Liquid-to- Solid Ratio (L:S)

The liquid-to-solid (L:S) ratio was studied to better understand the influence of saturation level on the leaching potential of metals. Four different L:S ratios were studied including 5:1, 10:1, 15:1, and 20:1. Figure 10 shows the leached metal concentrations decreased when L:S ratio of the influent solution was increased.

Increasing the liquid content of the leach test diluted the leachate as illustrated by the decreasing dissolved metal concentrations. Thus, dissolution of the samples did not increase indicating the RCA materials to be very insoluble at their original pH. These results are consistent with finding of previous studies. Garrabrants et al. (2004) claimed that leached Ca concentrations decreased with an increase in L:S ratio due to lower solubility of $CaCO_3$ mineral compared to portlandite and CaO. Gervais et al. (2004) also claimed that carbonation may have caused the precipitation of Cu when L:S ratio was increased (Garrabrants et al. 2004).

Both the solubility of ettringite and CaCrO4(s), and the sorption to ferrihydrite (Engelsen et al. 2010) control the release of Cr. The concentration of aqueous Cr is decreased by the formation of inner-sphere complexes of oxyanions (e.g. CrO_4^-) and iron oxide (Cornelis et al. 2008). The concentrations of leached Cr decreased with the L:S ratio. This is likely due to insoluble ettringite and/or hydrous ferric oxide (HFO). The presence of insoluble Fe precipitates in the RCA materials most likely caused the slight decrease in Fe concentrations with increasing L:S ratio. The RCA also released aqueous Fe^{3+}, likely forming $Fe(OH)_3$ and/or $FeCO_3$ precipitates. These precipitates are insoluble at high pH levels which could also contribute to decrease in leached Fe concentrations with higher L:S ratios (Gitari et al. 2008; Goswami and Mahanta 2007).

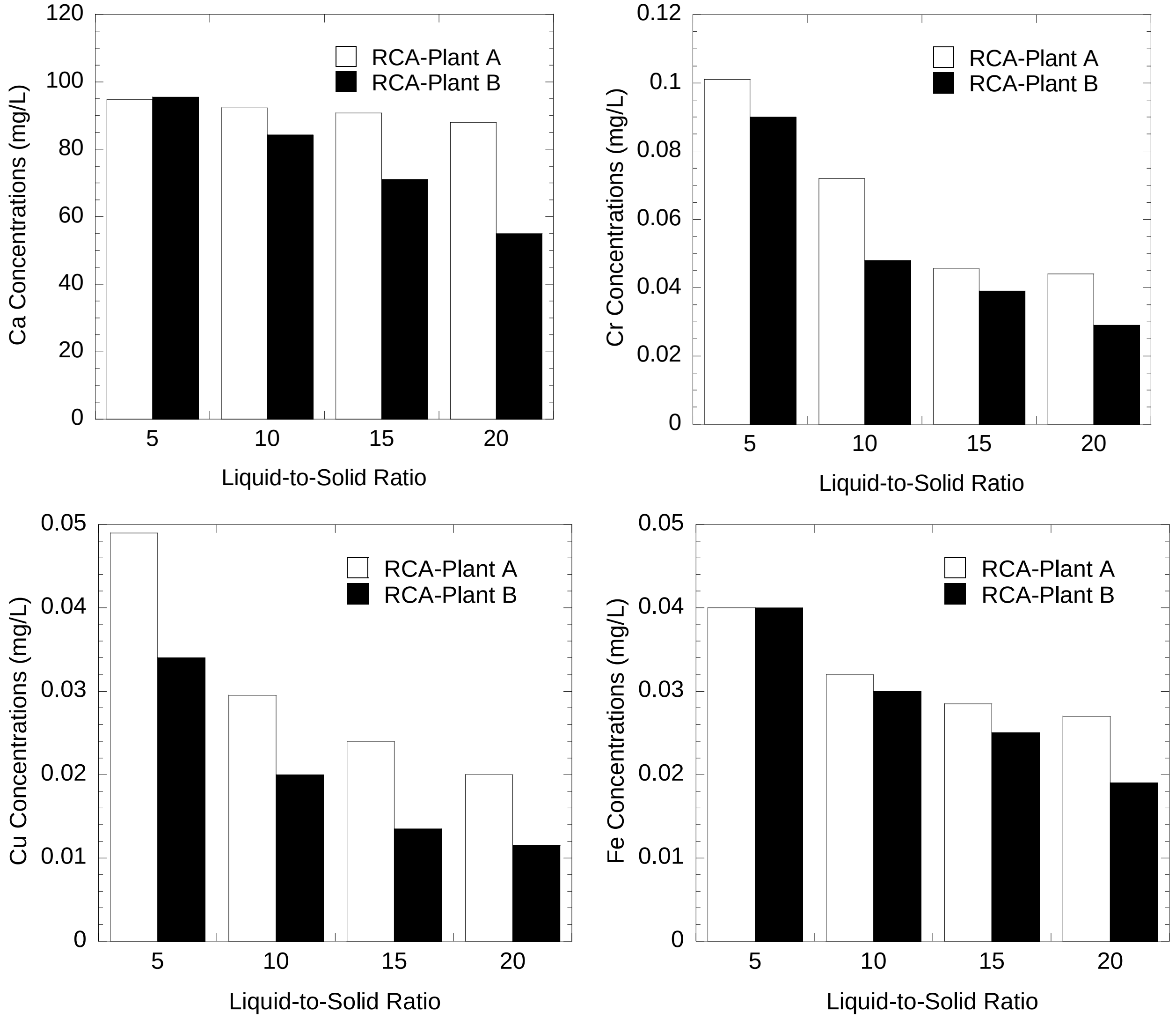

Figure 10. Effects of liquid-to-solid ration on leaching of Ca, Cr, Cu, and Fe from RCA.

Effect of Particle Size

Figure 11 shows that the leached Ca was relatively stable in each particle size fractions. The leaching patterns of Ca from RCA materials are similar to those trends observed for the effluent pH values at the corresponding particle sizes (Figure 7). This indicates that Ca contents in each sieve fractions possess great influence and directly impact the leachate pH. Figure 11 indicates that the concentrations of the leached Fe and Cu generally decreased with an increase in particle sizes while the leached Cr concentrations did not show any consistent trend.

The leaching behavior observed is due to differences in surface area in each particle size fraction. It is common knowledge that fine materials having a greater surface area than coarse materials. The larger surface area in the small particle sizes allowed for more interaction between the aqueous solution and the RCA to occur and thus enhanced leaching. Leaching occurs mainly by diffusion (Van Gervan et al. 2007), and a higher surface area would allow for more diffusion of elements into the aqueous phase. Furthermore, higher surface area of the smaller particles may yield more carbonation processes to occur on the particle surface. (Chen et al. 2013).

The degree of carbonation is dependent on the surface area of material (Houst and Witman 2002). Fine particles have a greater surface area than coarse particles yielding more carbonation to occur resulting in a lower pH which may eventually lead leaching of metals at higher concentrations (Chen et al. 2012). The pH of RCA-Plant B was slightly lower in the fines particles in this study as well (Figure 7). However, the results of this study showed that most of the RCA materials had leached lower Ca, Fe and Cu in the larger particles, which showed that surface area was the dominant factor that controlled the leaching behavior of elements more than carbonation. No consistent trend was observed between leaching of Cr and particle sizes. Cr is a

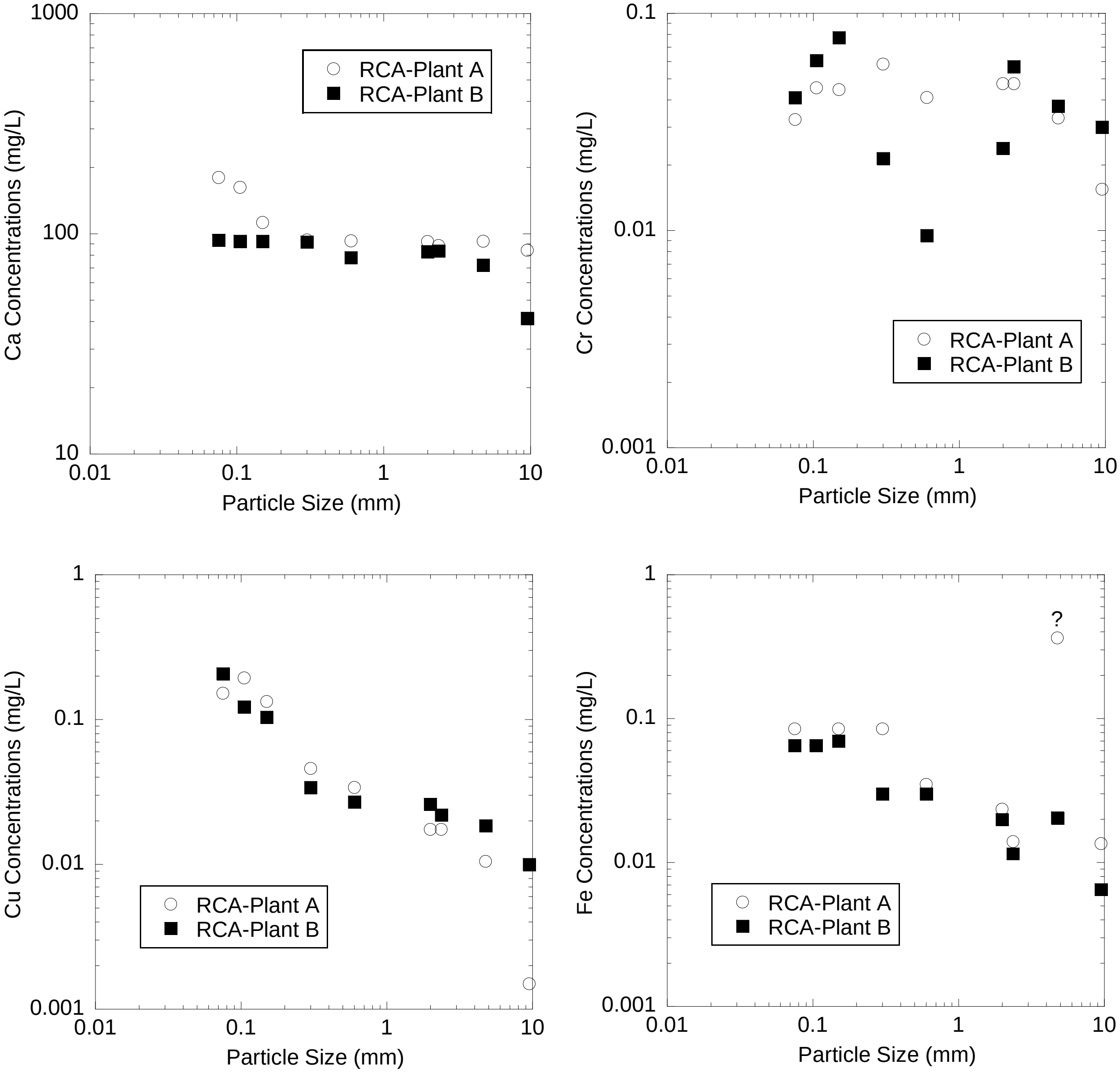

Figure 11. Effects of particle size on leaching of Ca, Cr, Cu and Fe from RCA.

redox and pH sensitive metal and its leaching is greatly affected by these parameters. This could be the reason that may impact the leaching of Cr from different size particles.

Another mechanism affecting leaching characteristics of elements by particle size is the cement composition within each fraction. Self-cementing in RCA is more likely to occur in the <0.15 and 0.3-0.6 mm size fractions due to increased C_2S concentrations within these fractions (Poon et al. 2006). Also, leaching of trace metals is mostly due to the cement paste content (Engelsen et al. 2010), and there may be more cement paste in certain fractions than others. It is expected that greater cement paste would occur in finer fractions.

pH - Dependent Leaching Tests

The pH of the leaching environment can greatly influence leaching behavior of metals. The leachate pH of RCA materials were adjusted from 2 to 13 and results are illustrated in Figure 12. The acid neutralizing capacity (ANC) and pH-dependent leaching of Ca, Cr, Cu, Fe, and Zn are shown in Figure 12. The leaching of Ca showed a cationic leaching pattern whereas the other metals followed an amphoteric leaching pattern. These patterns are consistent with past studies of RCA and Portland cement as presented by Chen et al. (2012), Engelsen et al. (2010, 2009), Garrabrants et al. (2004), and Sanchez et al. (2009).

The titration curves of the RCA materials show steep pH changes. Both materials had similar buffering capacity at the extreme basic range, but RCA-Plant A had superior buffering capacity in the acidic range. Buffering in the range is due to the dissolution of calcite ($CaCO_3$) in the materials which forms as the hydrated cement products, such as portlandite ($CaOH_2$), react with CO_2 (Chen et al. 2012 and Garrabrants et al. 2004). Thus, RCA-Plant A likely has more calcite than RCA-Plant B. This is supported by a greater percent CaO in RCA-Plant A (24%)

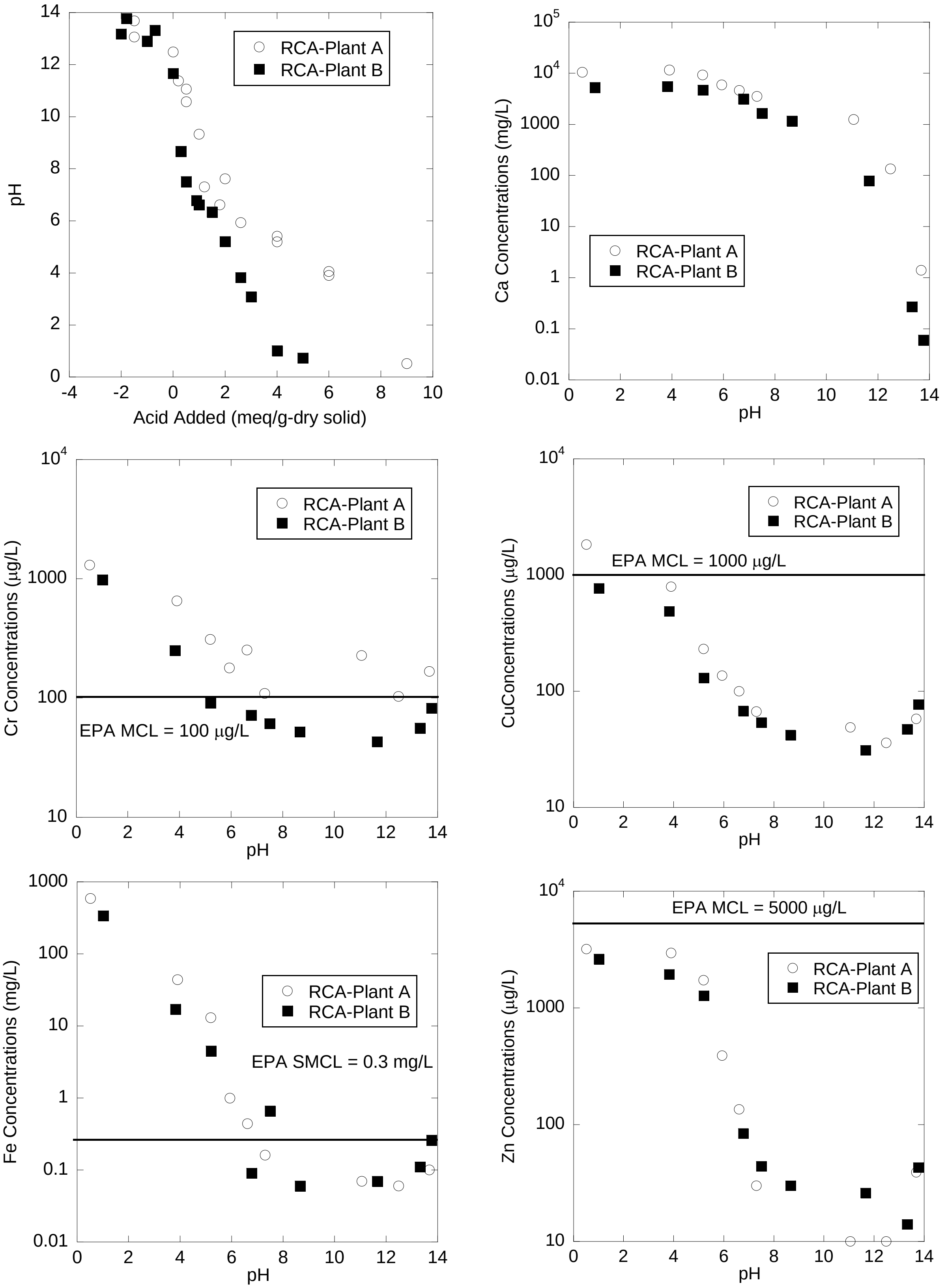

Figure 12. Results of pH-dependent leaching tests.

than RCA-Plant B (19%) through XRF analysis and higher concentrations of Ca leached from RCA-Plant A.

Leaching of major elements from wastes, under equilibrium conditions, is dominated by solubility processes (Kosson et al. 2002). Thus, Ca and Fe were likely solubility-controlled. For Ca, increasing the solution pH likely caused Ca^{2+} to precipitate as a carbonate, and decreasing the pH caused dissolution of this solid (Garrabrants et al. 2004 and Komonweeraket 2010). Ca showed a cationic leaching pattern which is characteristic of a monotonic decrease in leached concentrations as pH increases (Komonweeraket et al. 2011). Ca concentrations soared with increasing acidity due to dissolution of calcite and cement hydrates. More carbonated samples again would have less Ca released because $Ca(OH)_2$ is more soluble than $CaCO_3$ (Engelsen et al. 2009; Garrabrants et al. 2004).

Cr, Cu, Fe, and Zn had amphoteric leaching forming a U-shaped curve with increased concentrations at extreme acidic and alkaline pH values and minimum concentrations between these limits. At a pH near 10.5, the leached concentrations of these metals were the least. Acidic conditions caused the greatest release of Cr, Cu, Fe, and Zn as shown in Figure 12 due to greater dissolution of metal-bearing minerals. The pH-dependent release of Cr, Cu, Fe, and Zn is solubility-controlled and likely follows the solubility of the oxide and hydroxide species for each element (Engelsen et al. 2010; Garrabrants et al. 2004; Komonweeraket 2010), such as $Cr_2O_3/Cr(OH)_3$, $CuO/Cu(OH)_2/$ $Fe_2O_3/Fe(OH)_3$, $ZnO/Zn(OH)_2$. Cu may also be controlled by the dissolution/ precipitation of carbonate species (Komonweeraket 2010). The solubility of the cement hydrates in RCA is also an important factor in the leaching of Fe under different pH conditons (Engelsen et al. 2009). Lastly, trace metals may also be sorption-controlled under equilibrium conditions (Kosson et al. 2002). For example, geochemical modeling by Engelsen et al. (2010) showed the importance of adsorption of Cu^{2+} and CrO_4^{2-} to HFO at a pH of 5 to 10. Geochemical modeling of the RCA materials used in the current study would provide additional knowledge into the processes controlling leaching.

EPA drinking water standards were exceeded in the acidic leaching range for Cr, Cu, and Fe. Cr concentrations exceeded the MCL (100 μg/L) for both RCA materials after a pH of 5. Cu leached from RCA-Plant A exceeded the MCL (1 μg/L) at pH<2 and Fe leached from both RCA materials exceeded the SMCL (300 μg/L) at pH<7. A normal field environment would have neutral pH conditions, so it unlikely the leaching of Cr and Cu would cause health issues. Fe concentrations may exceed the SMCL in the environment, but this optional federal standard is in place for improved taste in drinking water (EPA 2009), and Fe leaching from RCA would likely not be detrimental to the environment.

CONCLUSIONS

A study was conducted to evaluate the mechanical properties of two different types of materials used in highway base layer constructions and the environmental suitability of recycled concrete aggregates (RCAs). Two recycled concrete materials, two natural virgin graded aggregate base (GAB) materials, and their selected mixtures were included in the testing program. The observations are summarized as follows:

1) RCA materials had higher CBR and SM_R values than the regular GAB materials, most likely due to the presence of higher CaO content which initiates the cementitious reaction in the soil matrix and yields an improvement in the mechanical properties of granular materials. The only exception was that CBR of GAB2 material was the highest among all of the specimens tested. On the other hand, GAB materials showed lower permanent

deformation (plastic strain) than both RCA materials indicating that regular GAB tended to provide better resistance under constant loads.

2) The RCA materials experienced a 0.98-2.1 times increase in SM_R with increasing freeze-thaw cycles due to the ongoing hydration process during freezing and thawing. The SM_R of RCA-GAB mixtures were lower than the ones for 100%RCA and 100%GAB materials. Similarly, permanent deformations of the mixtures were generally higher than those obtained for pure GAB or RCA.
3) Curing of RCA materials decreased the effluent pH and leached metal concentrations, except for Fe, mainly due to immobilization of metals caused by rehydration of cement particles in RCA aggregateLeached Fe concentrations increased when RCA materials were cured for 7 days and then decreased after 28 days of curing. These results indicated that cementation of RCA materials started encapsulating the leaching potential of Fe after 7days of curing.
4) All specimens were subjected to 4, 8, and 12 Freeze/Thaw (F/T) cycles. Effluent pH and leaching of Ca decreased with an increase in F/T cycles possibly due to the continued rehydration process. On the other hand no consistent trends were observed between leached Cr, Cu, and Fe concentrations and F/T cycles.
5) Several liquid-to-solid (L:S) ratio in the leaching tests were evaluated to better understand the influence of saturation level on the leaching potential of metals. Results indicated that increasing the liquid-to-solid ratio (L:S) (5,10,15, and 20) diluted the leached concentrations of metals from the RCA materials which yielded a significant decrease in leached metal concentrations.

6) The leached concentrations generally increased with decreasing particle size. These results suggested that the larger surface area in the small particle sizes allowed for more interaction between the aqueous solution and the RCA aggregate matrix to occur and thus enhanced leaching of metals.
7) pH-dependent leaching tests showed a cationic leaching pattern for Ca with concentrations increasing with decreasing pH. Amphoteric leaching patterns were observed in Cr, Cu, Fe, and Zn such that the concentrations were at minimum level at neutral pH and increased significantly at acidic and basic conditions.

Recycled concrete aggregate (RCA) is an especially valuable alternative material for use as highway base/sub-base as it has been shown to possess satisfactory mechanical properties. It is beneficial to recycle concrete for use as aggregate instead of landfilling of waste concrete. However, more laboratory and field tests and studies involving a variety of GAB and RCA materials are necessary to optimize the use of RCA in highway applications while safeguarding the environment.

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