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Peer reviewed

1 **Utilization of Post-Consumer Carpet Calcium Carbonate (PC4)**
2 **from Carpet Recycling as a Mineral Resource in Concrete**

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7

8 ***Abstract:***

9 The short residence time of certain building materials, such as carpets, results in a notable waste
10 stream that has the potential to be a resource for other material systems. In this work, we explore
11 the utilization of post-consumer carpet calcium carbonate (PC4) consisting primarily of the
12 material from carpet backing, which has thus far found few markets, as a potential mineral
13 admixture in the production of concrete. The effects on mechanical and material properties are
14 examined for concrete mixtures with PC4 used as 5 and 15% mass replacement of Portland
15 cement and used as 5 and 15% mass replacement of fine aggregates. The effects of PC4 on the
16 slump, air content, unit weight, and setting times were measured. Additionally, compressive
17 strength, flexural strength, coefficient of thermal expansion, shrinkage, bulk density, void
18 volume, and absorption were measured. Due to the known presence of antimony in PC4, leachate
19 from cured specimens was examined. Additionally, greenhouse gas emissions for the mixtures
20 were quantified. The results indicate that PC4 can increase air content in concrete by 3.5-10%
21 and decrease the unit weight by up to 11% in fresh concrete. PC4 also led to a loss in
22 compressive strength by up to 60%. Thus, the findings indicate that it is possible to form a less
23 dense, more porous concrete by using PC4 as a mineral constituent in concrete, but with some
24 loss in performance. Through appropriate selection of applications or through supplementary
25 treatment of PC4, it may find a suitable market in the concrete industry.

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31 ***Keywords:***

32 Post-consumer carpet; Concrete; Mechanical properties; Greenhouse gas emissions; Mineral
33 admixtures; Filler
34

35 **1. Introduction**

36 Construction of building and infrastructure systems is the largest consumer of materials in
37 the United States (Horvath, 2004). There is continued expected growth in building demand
38 (Breunig et al., 2018; IEA, 2017) and with it, demand for infrastructure materials. There are
39 many materials that are used in construction practice, including metal alloys (AISI, 2014; Bray,
40 2016), plastics (PlasticsEurope, 2010), and cement-based materials (Kapur et al., 2008). Portland
41 cement is the primary binder in concrete, which is typically composed of a hydraulic Portland
42 cement binder, water, and aggregates. Of the common construction materials, concrete is the
43 most consumed by mass and has a relatively long in-use service period once constructed (Kapur
44 et al., 2008; Monteiro et al., 2017). These factors suggest that waste minerals, even if used in
45 small fractions, could be incorporated into concrete in large quantities and, in doing so, be kept
46 from entering waste streams for long periods of time.

47 The components in built systems have varying lifespans, which leads to material waste flows
48 occurring at different times. A prime example of this service-life effect is the difference between
49 the lifespan of carpet in a building and the concrete building components. A piece of carpet
50 could be in-use for less than a fifth of the in-use period of concrete (Scheuer et al., 2003). While
51 materials such as carpets may not comprise a large mass of buildings, their high component
52 embodied energy and the frequency of replacement could lead to their being a significant
53 contributor to the total embodied energy of a building over its life cycle (Scheuer et al., 2003).
54 Currently, the United States (US) produces nearly 90 Mt of cement annually (van Oss, 2019),
55 with the majority being used in concrete (Xi et al., 2016). From the national average cement
56 content per cubic meter of concrete, 270 kg/m³ (ERMCO, 2016), it can be estimated that
57 approximately 320 million m³ of concrete are produced every year. Approximately 821 million

58 square meters of carpet is used in the US annually, based on sales in 2017 (CARE, 2018). Each
59 year, total discards of carpet are estimated at 1.5 Mt of carpet (CARE, 2018), less than 2% the
60 mass of cement production. Due to this low relative mass fraction, there is a potential to use
61 waste streams from one building component, carpets, in another, concrete, to increase the
62 potential for construction materials to contribute to the circular economy, in which material loops
63 are closed to extract maximum utility.

64 For decades, the exploration of using waste streams in concrete to reduce landfilling
65 materials and, in certain cases, improve the properties of concrete has been an active area of
66 study. It is well known that several industrial by-products, such as coal fly ash and blast furnace
67 slag, have pozzolanic and/or cementitious properties that can benefit the performance of concrete
68 (Mehta and Monteiro, 2006). The incorporation of other waste streams into concrete mixtures
69 have been studied with various effects on material performance. For example, studies in
70 literature range from replacing aggregates with crushed glass bottles and used tires (Khaloo et
71 al., 2008; Limbachiya et al., 2012) to fillers produced from waste ceramic powder (Subaşı et al.,
72 2017; Torkittikul and Chaipanich, 2010) and marble dust (Topçu et al., 2009). Previous research
73 on the utilization of carpet waste in concrete has focused on the use of plastic fibers (Wang et al.,
74 2000). The focus of these studies has been using carpet fiber to bridge concrete crack and
75 improve factors such as concrete tensile performance and impact resistance (e.g.,
76 (Mohammadhosseini et al., 2017)) and toughness (Wang et al., 2000). Classically, carpet backing
77 has been considered to have no significant use and has been the major part of carpet waste being
78 sent to landfills (Wang et al., 2000). While initial investigation has been performed on the
79 properties of carpet backing that could contribute to the performance of concrete, these studies
80 have been limited (Schmidt and Cieślak, 2008). The rising demand for Portland-limestone

81 cements, which use 5 to 15% limestone blended with Portland cement (Tennis et al., 2011),
82 could lead to a notable market for the utilization of the calcium carbonate from post-consumer
83 carpet backing if it is found to contribute to adequate concrete performance. Further, the
84 increased use of blended cements, such as these, could act as a mechanism to mitigate
85 greenhouse gas (GHG) emissions by lowering the overall clinker content in a set volume of
86 concrete (John et al., 2018).

87 The objective of this work is to provide a foundation for understanding the functionality of
88 using recycled calcium carbonate (CaCO_3) from deconstructed carpet— Post-consumer Carpet
89 Calcium Carbonate (PC4)— as a constituent in concrete production. Limestone is used as a raw
90 material for the production of Portland cement; however, there are other common uses for
91 limestone in concrete, namely: (i) as a mineral admixture; and (ii) as aggregate. If PC4 can be
92 used as a mineral admixture that can partially reduce cement demand while providing the same
93 performance as Portland-limestone cement, there could be environmental benefits, most notably
94 a reduction in GHG emissions. If PC4 can be used as aggregate, benefits could be obtained if
95 there are improvements in concrete performance or if there is a reduction in impacts through a
96 reduction in landfilling of PC4 and the need to excavate and transport limestone rock. Thus, this
97 work examines the effects of using PC4 to offset use of Portland cement or fine aggregates.
98 Comparisons for the influence on material properties from using PC4 were drawn to
99 conventional limestone fines (LS) as a partial replacement and a concrete mixture containing
100 neither PC4 nor LS. Properties assessed in this work include the coefficient of thermal
101 expansion, drying shrinkage, workability, flexural strength, compressive strength, void volume,
102 and specific gravity. Additionally, changes in the associated GHG emissions of each mixture

103 were quantified by performing environmental impact assessments of the concrete mixtures
104 studied.

105 **2. Materials and Methods**

106 **2.1. Materials and Mixtures**

107 For this work, PC4 from recycled nylon 66 carpet was obtained from Circular Polymers
108 located in Lincoln, California on March 14, 2019. From the recycling processes implemented at
109 the plant, the PC4 was obtained with 99.39% passing a No. 40 mesh (420 um) and 64.47%
110 passing a No. 100 mesh (150 um). To remove long remnant fibers, the PC4 was sieved through a
111 No. 40 sieve (420 um). Comparisons were made with mixtures using LS obtained from Blue
112 Mountain Minerals located in Columbia, California that had a fine gradation with 99.8% passing
113 a No. 120 (125 um) mesh and 95.8% passing a No. 200 (75 um) mesh. To assess the effects of
114 using PC4 relative to a typical concrete blend and relative to use of ground limestone, concrete
115 mixtures were batched. The Portland cement used was ASTM Type II/V, obtained from Lehigh
116 Southwest Cement Co in Stockton, CA. The fine aggregate was alluvial concrete sand sourced
117 from Esparto, California. Two sizes of coarse aggregates were used, an intermediate gravel
118 aggregate and a larger, crushed gravel aggregate, both obtained from Esparto, California
119 (aggregate, PC4, and LS gradations are given in Appendix A).

120 This work examined how PC4 can act as a partial replacement for Portland cement or fine
121 aggregate. For this assessment, two partial replacement levels were examined, namely 5% and
122 15% by mass. These replacement levels were selected because they are currently used in
123 Portland-limestone cements. The same replacement levels were considered for both the partial
124 replacement of Portland cement and the partial replacement of fine aggregates. Control group
125 blends were used for comparison, namely: 0% replacement, 5% replacement with LS, and 15%

126 replacement with LS. The mixture proportions and mixture nomenclature used are shown in
 127 Table 1.

128 **Table 1.** Mixture proportions for concrete and mortar specimens

Concrete Mixture Proportions (units in kg/m³)									
Mixture Name	Binder			Aggregates					
	Portland Cement	PC4	Limestone	5 mm	10 mm	25 mm	PC4	Limestone	Water
PC	411	0	0	516	382	763	0	0	193
C-PC4-5	390	21	0	505	382	763	0	0	193
C-PC4-15	349	62	0	482	382	763	0	0	193
C-L-5	390	0	21	514	382	763	0	0	193
C-L-15	349	0	62	510	382	763	0	0	193
FA-PC4-5	411	0	0	491	382	763	26	0	193
FA-PC4-15	411	0	0	439	382	763	77	0	193
FA-L-5	411	0	0	491	382	763	0	26	193
FA-L-15	411	0	0	439	382	763	0	77	193

Shrinkage and Setting Time Mortar Mixture Proportions (units in kg/m³)									
Mixture Name	Binder			Aggregates					
	Portland Cement	PC4	Limestone	5 mm	10 mm	25 mm	PC4	Limestone	Water
PC	411	0	0	516	0	0	0	0	193
C-PC4-5	390	21	0	505	0	0	0	0	193
C-PC4-15	349	62	0	482	0	0	0	0	193
C-L-5	390	0	21	514	0	0	0	0	193
C-L-15	349	0	62	510	0	0	0	0	193
FA-PC4-5	411	0	0	491	0	0	41	0	193
FA-PC4-15	411	0	0	439	0	0	123	0	193
FA-L-5	411	0	0	491	0	0	0	41	193
FA-L-15	411	0	0	439	0	0	0	123	193

129
 130 Concrete was batched using a Multiquip MC64SE concrete mixer. Aggregates were mixed
 131 for 2 minutes before adding Portland cement and PC4 or LS, where applicable. Mixing then
 132 commenced for an additional 2 minutes prior to the inclusion of water. Water was gradually
 133 added as constituents were mixed for 3 additional minutes. Blended mixtures were allowed to sit
 134 for 3 minutes followed by a final mixing of 2 minutes. Batches were then molded. After 1 day,
 135 specimens were demolded and placed in a conditioning chamber set at 25°C and a relative
 136 humidity of 95%. Compressive strength testing was performed on cylinders measuring 100 mm
 137 X 200 mm (4 X 8 inches); flexural strength tests were performed on prisms of 100 X 100 X 300
 138 mm (4 X 4 X 12 inches); coefficient of thermal expansion test specimens were cut from 100 mm

139 X 200 mm (4 X 8 inch) cylinders; density, permeable air voids, and porosity tests were
140 performed on 100 mm X 200 mm (4 X 8 inch) cylinder specimens.

141 For the setting time and shrinkage experiments, mortar was prepared separately from the
142 concrete mixtures, but the same replacement levels were considered. The mortar mixtures were
143 batched in Hobart A200 dough mixer. To make mortars, fine aggregates were dried out in an
144 oven at 120°C, then allowed to cool before mixing. Sand and powder were combined in the
145 mixing bowl and mixed for 1 minute. After 1 minute, water was gradually added, and the
146 mixture was mixed for 2 more minutes. The mixture was allowed to sit for 1 minute and finally
147 mixed for 2 minutes before being molded. Shrinkage prism specimens were cast with dimensions
148 of 25 mm X 25 mm X 285 mm (1 X 1 X 11.2 inches). Setting time mixtures were tested in
149 154 mm X 154 mm (6 X 6 inch) cylinders forms.

150

151 **2.2. Experimental Procedures to Test Fresh Concrete Properties**

152 ***2.2.1. Slump, Air Content, and Unit Weight***

153 The partial replacement of aggregates and cement with mineral admixtures can have a
154 significant influence on the fresh properties of concrete (Esping, 2008). These fresh properties
155 can affect the ability to easily place the concrete or inform the need to use certain chemical
156 admixtures. For this reason, several fresh-state properties of the concrete mixtures were assessed.
157 To determine the effects concrete workability, slump was measured following ASTM C143
158 (ASTM, 2015). To determine the effects of PC4 on air content and density, the air content in the
159 fresh concrete was assessed following ASTM C231 (ASTM, 2017d) and the unit weight of the
160 fresh concrete mixtures was assessed following ASTM C138 (ASTM, 2017b).

161 **2.2.2. Setting Times**

162 To evaluate the effects of PC4 on setting time for the mixtures, experimental investigation of
163 initial and final set times was performed following ASTM C403 (ASTM, 2016b). For each
164 mixture, three specimens were produced for testing. As the mortars set, the resistance to
165 penetration was measured using an Acme Penetrometer and the initial and final set times were
166 determined.

167 **2.2.3. Isothermal Calorimetry**

168 The heat flow from hydration of the binder mixtures was evaluated to examine the effect of
169 PC4 on the initial hydration. As with the concrete and mortar tests, comparisons were drawn to a
170 control binder with no mineral admixture and to mixtures with LS. For the binder containing
171 PC4 and for the binder containing LS, the powder components were made with 5% and 15%
172 mass replacement of Portland cement with each mineral admixture, respectively. Binders were
173 proportioned such that the total mass of the paste mixture was 5g, with a 0.47 water-to-binder
174 ratio. Reference samples were made to match the heat capacity of the paste mixtures using inert
175 silica sand. Samples were tested in a TAM Air Isothermal Microcalorimeter (TA instruments) at
176 30°C using glass admixture ampoules and a syringe-and-paddle mixing apparatus that allows for
177 water addition as well as paste mixing within the calorimeter. Powder mixtures were weighed
178 into the ampoules and distilled water was loaded into a syringe, the syringe tips were dried, and
179 then dipped into silicon grease to prevent water from prematurely interacting with the powder.
180 The ampoules were loaded into the calorimeter sample chamber with a corresponding reference
181 sample added to the reference chamber. After the samples equilibrated to the chamber
182 temperature, the baseline was measured, and the signal was corrected automatically by the
183 accompanying software package. Water was then injected into the first sample, the time of

184 injection was marked, and the paste was mixed for approximately 90 seconds. This process was
185 then repeated for each sample.

186 **2.3. Experimental Procedures to Test Hardened Concrete Properties**

187 ***2.3.1. Compressive Strength***

188 It is known that mineral additions like limestone filler can induce early hydration in concrete
189 mixtures and can lead to a dilution effect which lowers compressive strength at later ages relative
190 to a Portland cement mixture without limestone (Ghrici et al., 2007). For this reason,
191 compressive strength was determined for each of the concrete mixtures at several curing ages,
192 namely, after 7, 14, 28, and 56 days of curing. Experiments were performed on a SoilTest CT-
193 950 load frame following ASTM C39 testing procedures (ASTM, 2017a). Cylinder specimens
194 were capped on either end with neoprene-padded aluminum cap and specimens were loaded
195 under force control. Five specimens were tested for each mixture at each of the four ages.
196 Ultimate strength of the concrete mixtures was determined based on the average maximum load
197 reached before softening or failure occurred.

198 ***2.3.2. Flexural Strength***

199 Flexural strength was determined by performing three-point bend tests after 28 days of
200 curing using a MTS Testline Component load frame managed by an MTS TestStarII controller
201 following ASTM C293 testing procedures (ASTM, 2016a). Three specimens were tested from
202 each mixture and average ultimate flexural strength was determined.

203 ***2.3.3. Coefficient of Thermal Expansion***

204 Coefficient of thermal expansion is known to have significant effects on the feasibility of
205 utilizing concrete mixtures in systems that undergo changes in temperature, such as roadways
206 (Uygunoğlu and Topçu, 2009). To assess the effects of PC4 and limestone on Portland cement-

207 based mixtures, this work followed AASHTO T336 testing procedure (AASHTO, 2015). Using
208 100 mm X 200 mm (4 X 8 inch) cylinders, specimens for testing were cut to have a length of
209 177.9 ± 2.54 mm (7 ± 0.1 inches). One concrete specimen was tested for each of the mixtures
210 after curing for 42 days. Experiments were performed at the University of California Pavement
211 Research Center.

212 **2.3.4. Shrinkage**

213 To determine how replacement of Portland cement or fine aggregates with PC4 affects
214 drying shrinkage, mortar specimens were cast to have the same binder content and replacement
215 ratios as the concrete mixtures tested. Following ASTM C157 testing procedures (ASTM,
216 2017c), specimens were removed from molds after 1 day of curing and placed in a conditioning
217 chamber set at 25°C and 50% relative humidity. Three mortar prisms were cast for each mixture.
218 The length change of each mortar specimen was measured at 0, 14, 21, and 28 days and the
219 average length changes were reported.

220 **2.3.5. Bulk Density, Void Volume, and Absorption**

221 Density, absorption, and voids in each of the concrete mixtures were assessed following
222 ASTM C642 testing procedures (ASTM, 2013). One 101.6 mm X 203.2 mm (4 X 8 inch)
223 cylinder specimen from each concrete mixture was tested after curing for 28 days. To determine
224 oven-dry weight, specimens were dried at 100-110°C and weighed every 24 hours until less than
225 0.5% weight fluctuation between two successive measurements were achieved. Specimens were
226 then submerged in water and weight measurements of the specimens after surface drying were
227 taken every 24 hours until less than 0.5% weight fluctuation between two successive
228 measurements occurred. Specimens were then placed in water and boiled for 5 hours. The boiled
229 specimens were allowed to cool for 14 hours before being submerged in water using a specific

230 gravity bench to determine the apparent weight. Finally, the surface was dried and the specimens
231 were weighed once more. Water absorption, void volume, apparent density, and bulk density
232 were determined using the formulae stipulated in ASTM C642 (ASTM, 2013).

233 **2.4. Experimental Procedure to Test Leaching from Concrete**

234 Additionally, due to potential remnant material in the PC4 obtained from their first life cycle,
235 leaching tests were performed on the concrete mixtures to assess whether the cement had
236 adequately stabilized major and minor metals, trace metals and semimetals. Leaching tests were
237 performed on concrete specimens after 105 days of curing. Tests were conducted on one 100 mm
238 X 200 mm (4 X 8 inch) cylinder specimen for each of the concrete mixtures. Leachate containing
239 soluble minerals was tested after 3 days of immersion and after 90 days of immersion.

240 **2.5. Estimating Greenhouse Gas Emissions from Producing Concrete Mixtures**

241 Noting that mineral admixtures can facilitate reductions of cement content in concrete, which
242 can in turn lower GHG emissions, a preliminary assessment of changes to GHG emissions from
243 using ground limestone or PC4 in concrete was examined in this work. For each of the mixtures
244 (proportions in Table 1), GHG emissions were modeled using the GreenConcrete LCA Webtool
245 developed at the University of California Berkeley (Gursel and Horvath, 2012). Within this tool,
246 100-year global warming potentials were used to draw comparisons. Production was modeled as
247 occurring in California, including all constituent production and mixture batching. For each
248 constituent requiring electricity during processing and for concrete batching, the average
249 California grid mix for 2018 was used (California Energy Commission, 2019). For modeling the
250 production of cement clinker, the California average kiln fuel mix for 2017 was used (California
251 Air Resources Board, 2019). Cement production was modeled in the GreenConcrete Tool
252 following the most common production modes in the US (Marceau et al., 2006). Specifically, the

253 raw material processing was modeled as dry ball milling and preheater/precalciner processing.
254 The clinker was modeled as being cooled through a reciprocating grate cooler and finish milled
255 in a ball mill. For each of the concrete constituents, transportation by truck was selected since it
256 is the predominant mode throughout the cement and concrete production process in the US
257 (Marceau et al., 2006, 2007). Transportation distances are the average for each constituent
258 provided in reports published by the Portland Cement Association (Marceau et al., 2006, 2007).
259 Namely, these distances were modeled as 146 km for cement, 61 km for fine aggregate, 43 km
260 for coarse aggregate, and 43 km for LS. Water was considered to be available on site with
261 negligible energy resources necessary for its acquisition. Because PC4 is a residual waste
262 product of the carpet recycling process, the impacts from PC4 removal from carpet were
263 allocated to the recycled plastic product. As such, no GHG emissions were modeled from the
264 production of PC4. However, transportation of PC4 to the concrete batching site was considered.
265 This PC4 transportation was modeled as 197 km by truck, based on the US average distance for
266 post-industrial product transport for use in concrete (Marceau et al., 2007).

267

268 **3. Results**

269 **3.1. Fresh Concrete Properties**

270 ***3.1.1. Slump, Air Content, and Unit Weight***

271 The incorporation of PC4 influenced slump, air content, and unit weight of concrete (see
272 Table 2). Partial replacement of Portland cement with PC4 and 5% replacement of fine aggregate
273 with PC4 increased concrete slump by 5 to 27%. However, a 15% replacement of fine aggregate
274 resulted in a 64% decrease in slump. The partial replacement of cement with PC4 had similar
275 effects on slump as partial replacement with LS. This similarity suggests that, like the LS, a filler

276 effect or a net increase in water to Portland cement ratio could have contributed to an increase in
 277 workability. The same reduction in workability was noted when 15% of fine aggregate was
 278 replaced with PC4 and LS relative to the Portland cement mixture (without any PC4 or LS).
 279 However, noting the slump increased at 5% fine aggregate replacement, there is an apparent
 280 varying effect of the PC4 based on the concentration. The carpet backing from which PC4 is
 281 produced from is known to contain polymer fibers and latex adhesives (Brentin, 1982; Realff,
 282 2007; Realff et al., 1999). It is possible that remnant compounds from the backing, such as latex
 283 and microfibers, are affecting surface interactions and, possibly, contribute to the noted
 284 differences.

285 **Table 2.** Slump, air content, and unit weight of fresh concrete mixtures and the average
 286 compressive strength, flexural strength, and coefficient of thermal expansion of hardened
 287 concrete specimens.

Mixture Name	Fresh Properties Measured			Hardened Properties		
	Slump, mm	Air Content, %	Unit Weight, kg/m ³	f _c ', MPa (range)	f _r ', MPa (range)	CTE, microstrain / °F (range)
PC	140	6	2431	43.8 (4.1)	6.89 (0.12)	7.08 (0.03)
C-PC4-5	165	12.5	2161	26.8 (5.5)	4.29 (0.52)	7.15 (0.01)
C-PC4-15	146	16	2087	14.4 (1.4)	3.17 * (0.10)	7.16 (0.03)
C-L-5	152	1.8	2835	46.5 (4.1)	3.18 (0.19)	7.28 (0.02)
C-L-15	165	1.2	2739	44.8 (2.7)	3.84 (2.12)	7.05 (0.07)
FA-PC4-5	178	11	2609	24.9 (4.1)	4.46 (0.47)	7.25 (0.05)
FA-PC4-15	51	9.5	2649	22.6 (1.6)	4.06 (0.51)	7.11 (0.03)
FA-L-5	114	1.5	2846	44.2 (5.2)	2.93 (0.32)	6.93 (0.05)
FA-L-15	51	1.4	2860	43.6 (3.3)	3.25 (0.41)	7.15 (0.07)

288 '* * ' indicates only two specimens tested; f_c' = 28-day compressive strength; f_r' = 28-day flexural strength; CTE = 42-
 289 day coefficient of thermal expansion. Values in '() ' indicate the range between values used to calculate the reported
 290 average

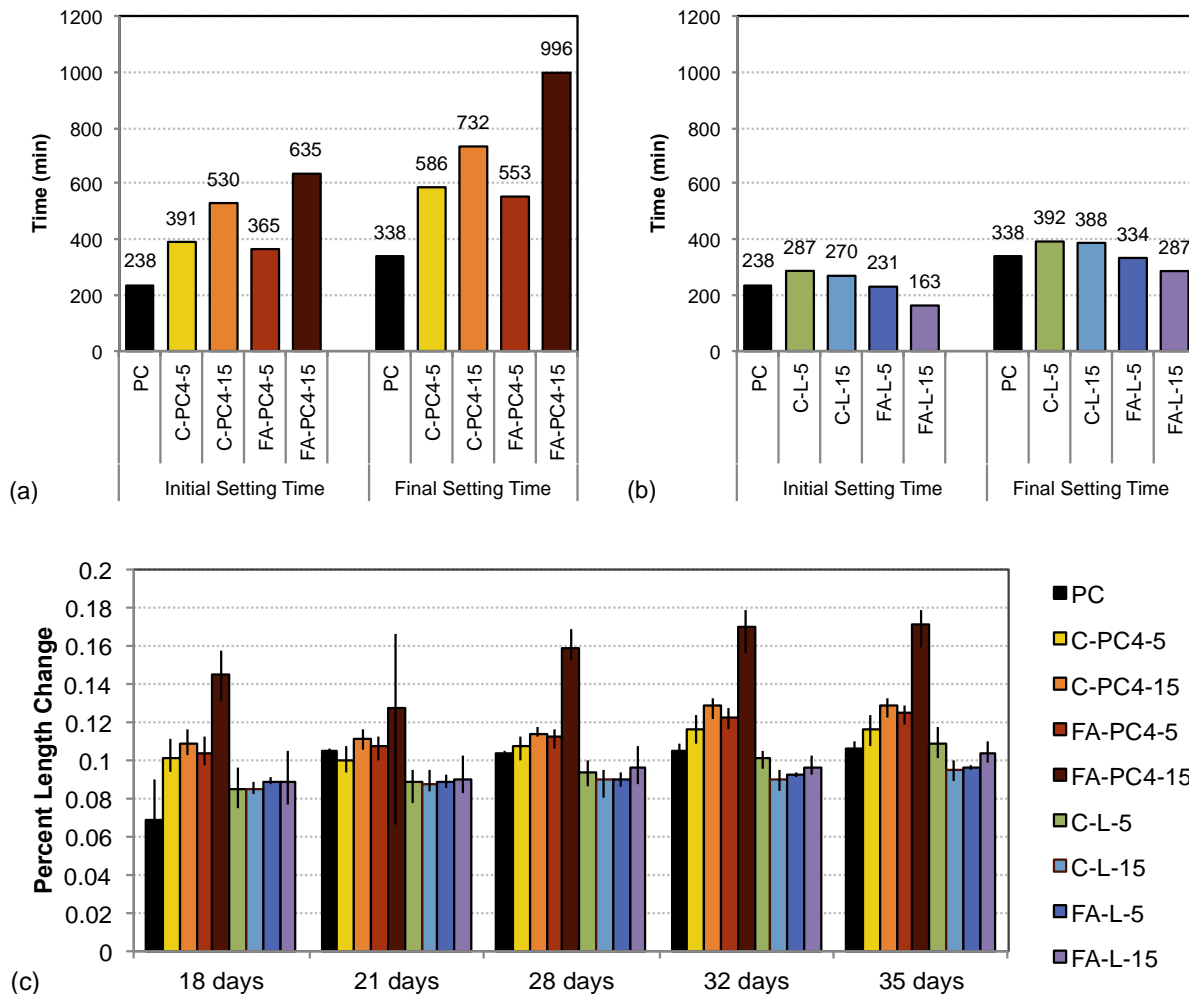
291 The use of PC4 did not consistently affect the air content or unit weight of fresh concrete in
292 the same manner as the LS. The PC4 consistently increased air content relative to the control
293 mixtures. This change could be a function of the size and air content of the PC4 particles.
294 Additionally, any remnant latex could contribute to increased air entrainment during mixing. The
295 use of LS had the opposite effect, leading to a consistent reduction in air content. The LS also
296 increased the concrete unit weight relative to the Portland cement mixture. Both parameters
297 indicated that the finer LS particles likely increased particle packing. While the PC4 increased
298 air content, the replacement of Portland cement with PC4 led to a reduction of unit weight and
299 the replacement of fine aggregate with PC4 led to an increase in unit weight. These results
300 suggest that the PC4 may improve gradation when used as a fine aggregate replacement, but not
301 when used as a partial cement replacement. These differing effects based on constituent replaced
302 could be due to the gradation of PC4, which is coarser than the LS.

303

304 ***3.1.2. Setting time***

305 The inclusion of PC4 resulted in an increase to both the initial and final setting times relative
306 to the mixtures containing no PC4 (see Figure 1). Compared to the Portland cement mixture, the
307 replacement of cement with PC4 led to a 64-123% increase in initial set time and a 74-117%
308 increase in final set time. While 5% fine aggregate replacement did not lead to as large a change
309 to setting times as the 5% Portland cement replacement, 15% fine aggregate replacement with
310 PC4 resulted in 2.5- and 3-times greater initial and final setting times than the Portland cement
311 mixture, respectively. As a result, all of the PC4 mixtures studied here exceeded the 375 min final
312 setting time stipulated by code (Mehta and Monteiro, 2006). Under certain scenarios, it is
313 possible that the increase in initial setting time offered by the PC4 could improve the window
314 during which the concrete can be adequately mixed and placed. However, the delay in when

315 mechanical strength begins to develop could affect the use of PC4 in certain applications. For
 316 example, the delayed setting time would hamper the use of PC4 in applications when a load
 317 needs to be applied to the concrete quickly or it could increase the time between which concrete
 318 is poured and framework can be removed.



319 **Figure 1.** Initial and final setting times for (a) mixtures containing PC4 and (b) mixtures
 320 containing limestone relative to a mixture with neither and (c) shrinkage of specimens over a
 321 two-week period as shown by percent change in length
 322
 323

324 The use of LS in the concrete mixtures resulted in smaller changes in setting times than the
 325 use of PC4. Cement replacement with LS increased initial and final setting times relative to the
 326 Portland cement mixture, but by less than the use of PC4, namely by 13-21% and 15-16%,
 327 respectively. This shift in final setting times also resulted in the mixtures containing LS as a

328 partial cement replacement exceeding final set time limits. It is possible that these shifts were
329 caused by the reduction in Portland cement, which the LS partially compensates for by improved
330 cement dispersion and hydration. The larger particle size of PC4, however, may not be as
331 effective at cement particle dispersion. The use of LS as a partial replacement for fine aggregates
332 reduced both initial and final setting times by up to 31% and 15%, respectively. This change
333 could result in these LS mixtures being more difficult to handle, but it would lead to the earlier
334 onset of strength development.

335 ***3.1.3. Isothermal Calorimetry***

336 The partial replacement of cement with either PC4 or LS showed little effect on the early age
337 heat flow of the binders (Appendix B). The PC, C-L-5, and C-L-15 binders all reached maximum
338 heat flow approximately 8 hours after the introduction of water into the powder. This was
339 followed by C-PC4-5 with a delay of approximately 30 minutes, a 6% increase in time. The C-
340 PC4-15 binder was offset from the first three mixtures by approximately 2 hours, a 25% increase
341 in time; however, the peak heat flow of C-PC4-15 was approximately the same as the binders
342 with 5% replacement and the binder with neither PC4 nor LS. The C-L-15 paste had a small,
343 ~3%, decrease in heat flow at the peak compared to the other mixtures. This decrease for the C-
344 L-15 is consistent with mixtures reported in literature with similar levels of Portland cement
345 replacement with LS and it is also similar to the results for cement replacement with fly ash (De
346 Weerd et al., 2011; Thongsanitgarn et al., 2014). The 2-hour shift for C-PC4-15 is also similar
347 to the delays shown with fly ash replacement (De Weerd et al., 2011; Thongsanitgarn et al.,
348 2014).

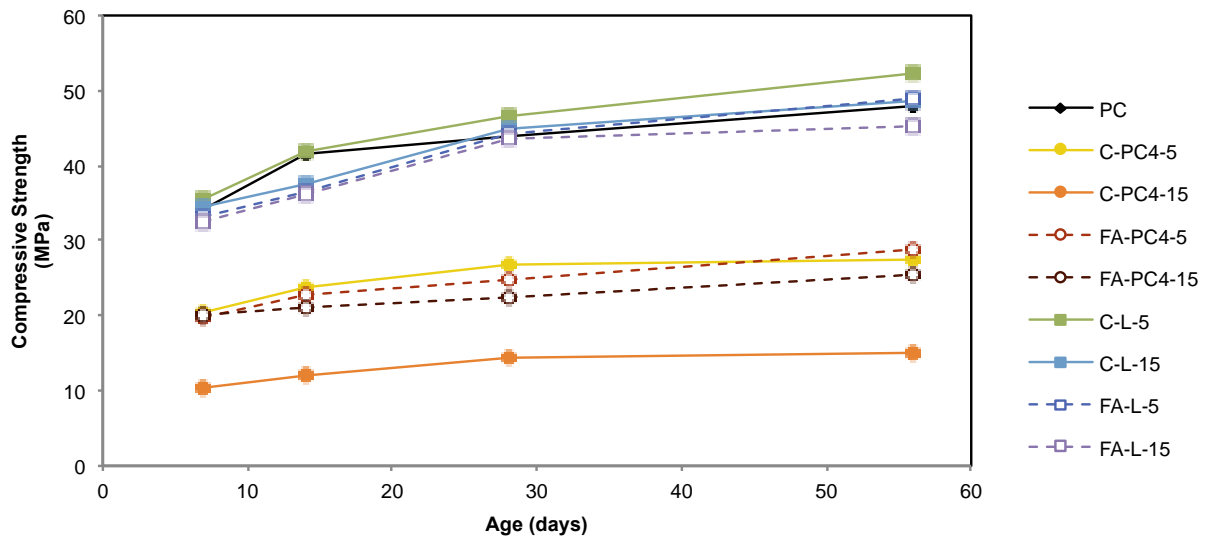
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351 **3.2. Hardened Concrete Properties**

352 **3.2.1. Compressive Strength**

353 Each of the concrete mixtures studied showed an increase in compressive strength over time,
354 ranging from a 27% to 48% increase between measurements taken at 7 days and 56 days (Figure
355 2). The largest increase in strength was exhibited by FA-L-5 and the smallest was exhibited by
356 the FA-PC4-15. The mixtures containing PC4 had significantly lower compressive strength than
357 the control mixtures (based on a T-test analysis, $p < 0.001$ for all ages). The PC4 mixtures varied
358 from 10.4 MPa at 7 days for C-PC4-15 to 28.7 MPa at 56 day for FA-PC4-15. At 28-day and 56-
359 day ages, all LS mixtures had higher compressive strengths than the Portland cement mixture.
360 However, while the compressive strength for mixture C-L-5 differed significantly from the
361 Portland cement mixture at 28- and 56-day ages ($p = 0.036$ and 0.015 , respectively), the other
362 mixtures containing LS did not ($p = 0.285$ and 0.655 , $p = 0.667$ and 0.448 , and $p = 0.867$ and $p =$
363 0.056 for mixtures C-L-15, FA-L-5, and FA-L-15 at each age, respectively).



364 **Figure 2.** Compressive strength development of concrete mixtures containing PC4 or limestone
365 relative to a mixture with neither
366
367

368 In general, LS replacement appeared to either improve or have negligible effects on
369 compressive strength, while PC4 replacement led to notable decreases in compressive strength at
370 all ages and replacement levels tested, a 30 to 60% reduction in strength relative to the Portland
371 cement mixture. For the LS mixtures, most of the strength development occurred at early ages
372 (i.e., below 28 days). This trend was true for the PC4 mixtures with the partial replacement of
373 Portland cement. However, similar strength development was noted between 7 to 28 days and
374 between 28 to 56 days for the PC4 mixtures with fine aggregate replacement. The inclusion of
375 LS has been shown to contribute to early age strength development and a dilution effect that
376 influences later age strength development (Ghrici et al., 2007); however, it has also been shown
377 that the effects of such mineral additions can have varying influence on mechanical properties
378 depending on a variety of other factors (Wang et al., 2018). In addition to these contributing
379 factors, the PC4 could have a different internal microstructure than LS or remnant coating, such
380 as latex, both potential artifacts of the carpet manufacturing process (Brentin, 1982; Realff, 2007;
381 Realff et al., 1999). Styrene-acrylic ester copolymer latex, even in low concentrations, has been
382 shown to significantly reduce the compressive strength of cement mortars (Wang and Wang,
383 2010). It is possible remnant latex polymers in PC4 behaved similarly in the mixtures tested.
384 Such artifacts could either lead to flaws at the interface between matrix and inclusion or flaws
385 within the inclusions, which may affect strength. Further study, regarding the chemical and
386 microstructural properties of PC4 should be conducted to assess such parameters.

387 ***3.2.2. Flexural Strength***

388 Both PC4 and LS decreased flexural strength at 28 days (see Table 2) relative to the Portland
389 cement mixture. The partial replacement of fine aggregate with PC4 had the least effect on
390 flexural strength, resulting in a 35% reduction. The greatest reduction in flexural strength was

391 noted for the 5% replacement of fine aggregates by LS, but when considering specimen
392 variability, the reduction in flexural strength noted for this mixture was similar to that for several
393 others. The use of LS has been shown in past work to lead to varying effects on different
394 mechanical properties, dependent on factors such as gradation, composition, and water-to-binder
395 ratio (Ghrici et al., 2007; Kenai et al., 2004; Wang et al., 2018). However, the consistent
396 reduction in mechanical properties from PC4 could be a function of a dilution effect or inherent
397 flaws within the particles themselves, acting as either crack initiation or weak points that
398 facilitate propagation of failure through the concrete specimens.

399 ***3.2.3. Coefficient of Thermal Expansion***

400 The partial replacement of cement or fine aggregates with either PC4 or LS had limited effect
401 on the coefficient of thermal expansion (see Table 2). The Portland cement mixture, with neither
402 PC4 nor LS, had a coefficient of thermal expansion of approximately 7 microstrain / °F. The use
403 of PC4 in the concrete mixtures had a 0 to 3% increase in the coefficient of thermal expansion;
404 the use of LS had between a 2% reduction and a 3% increase in the coefficient of thermal
405 expansion. These minute variations resulted in all mixtures having coefficients of thermal
406 expansion within a normal range for conventional concretes (Uygunoğlu and Topçu, 2009) and
407 indicates these mixtures would behave similarly under typical temperature conditions.

408 ***3.2.4. Shrinkage***

409 In most cases, the inclusion of PC4 exhibited a larger impact on drying shrinkage than the
410 inclusion of LS (Figure 1). Mixtures with higher weight fractions of PC4 generally exhibited 3 to
411 110% greater arithmetic mean shrinkage than PC, depending on age and mixture. The exception
412 to this trend was the C-PC4-5 at 21 days, where, considering the variability present, the change
413 in length appears comparable to the Portland cement mixture. The inclusion of LS filler

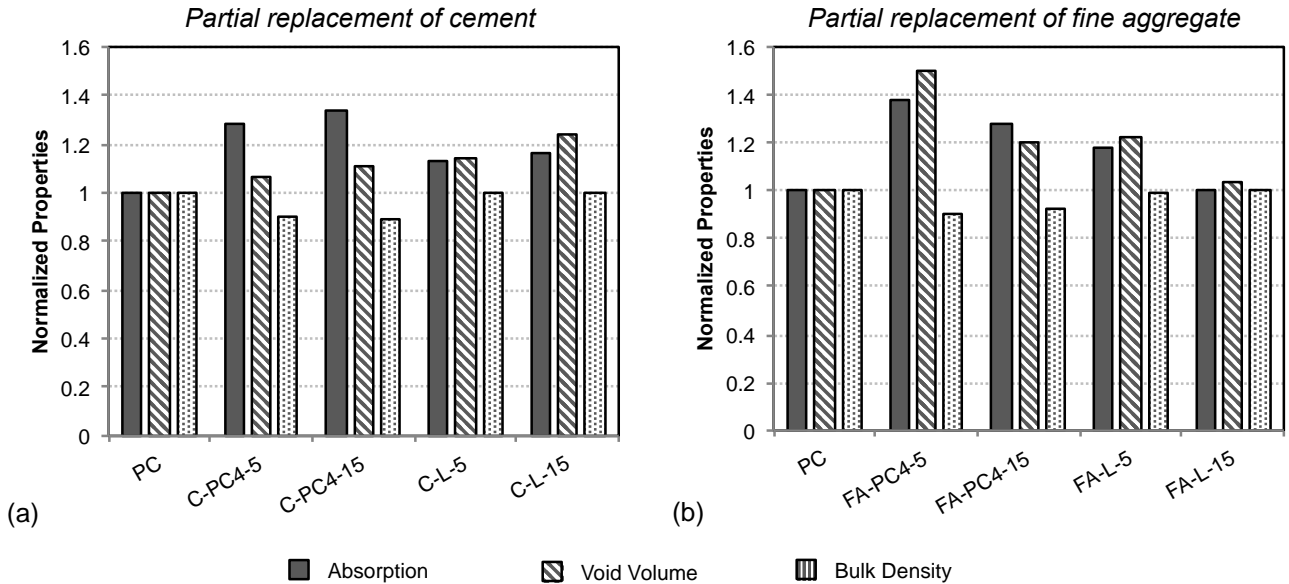
414 predominantly exhibited the opposite effect on drying shrinkage. With the exception of early age
415 measurements, the mixtures with LS filler generally exhibited smaller length changes than the
416 Portland cement mixture (3 to 15% lower change in length). The exception to this trend was the
417 C-L-5 mixture at 35 days, which had comparable shrinkage as the Portland cement mixture. It is
418 known that geometry, moisture, and material properties all affect the drying shrinkage of
419 cement-based materials. Given that the moisture and geometry are controlled, the changes
420 observed are potentially caused by the material properties of the LS and PC4 used. The LS, with
421 a higher modulus of elasticity, can reduce drying shrinkage (Mehta and Monteiro, 2006).
422 However, the opposite is observed for PC4 inclusion, suggesting the less-dense PC4 could lead
423 to a lower modulus elasticity and, thus, lead to the increased drying shrinkage observed.

424 ***3.2.5. Bulk Density, Void Volume, and Absorption***

425 Inclusion of PC4 consistently increased concrete absorption and void volume, while reducing
426 bulk density (see Figure 3). As a partial Portland cement replacement, PC4 increased absorption
427 by 28-34% and void volume by 7-11%, while reducing bulk density by ~10%. As a replacement
428 of fine aggregate, PC4 increased absorption by 28-38% and void volume by 20-50%, while
429 reducing bulk density by ~10%. While this notable increase in voids and absorption could lead to
430 less desirable attributes in a reinforced concrete system by facilitating additional ingress of
431 moisture, the ability to form a less dense, more permeable concrete could be desirable in other
432 applications requiring lighter or permeable concretes.

433 The partial replacement of Portland cement and fine aggregate with LS did not have as large
434 of an effect as the PC4, but trends of increased absorption and void volume were still noted.
435 Despite these changes, bulk density remained relatively consistent to the Portland cement
436 mixture. The partial replacement of Portland cement with LS led to a 13-16% increase in

437 absorption and a 15-24% increase in void volume, whereas the partial replacement of fine
 438 aggregates resulted in a 1-19% increase in absorption and a 4-23% increase in void volume
 439 relative to the Portland cement mixture. It is possible the shifts noted from the inclusion of LS
 440 are associated with both the density of the LS and its effect on particle packing; however, the
 441 trends noted from the inclusion of PC4 suggest that it is not consistently contributing to the same
 442 effects. As discussed above, the variability from the PC4 replacement could be attributed to the
 443 gradation of PC4 modifying the overall gradation of the mixture, dependent upon which
 444 constituent the PC4 replaces.



445
 446 **Figure 3.** Absorption, void volume, and bulk density for concrete mixtures normalized to the
 447 Portland cement mixture without either PC4 or quarried limestone for (a) partial cement
 448 replacement and (b) partial fine aggregate replacement
 449

450 3.3. Leaching

451 Three-day leachate (with stirring) at 105 days cured, show detectable antimony (Sb), a
 452 known PC4 impurity due to its use as a catalyst in production of PET (polyethylene
 453 terephthalate) plastic, a common carpet fiber. The Sb concentration was greater when the
 454 replacement was for Portland cement (vs. control replacement with LS) than for replacement for

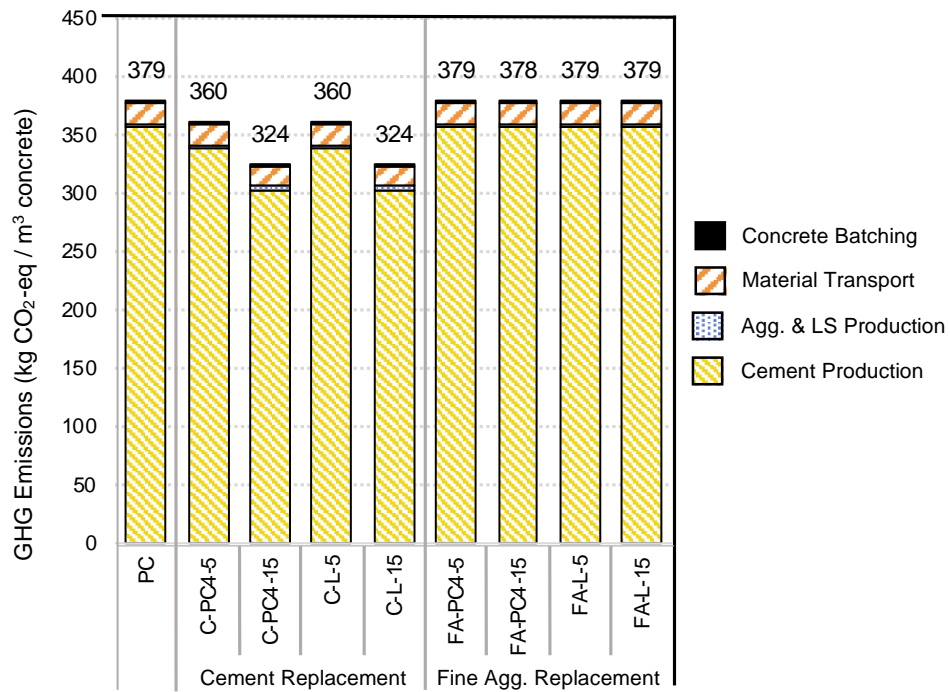
455 fine aggregate (again vs. control replacement): 118+-19 ng/L compared to 26+-18 ng/L (where
456 ng/L is nanograms of Sb per liter of leaching water). These concentrations are far below water
457 quality levels of concern; however, the Sb content of other PC4s have been studied and found to
458 be as high as 80-fold larger than the PC4 used in this study. Such a multiplier would move the
459 concentrations (for cement replacement only) to be above safe limits established by the EU (5
460 ug/L) and the US EPA (6 ug/L). After an additional 90 days leaching, there was no difference
461 between leachate from the PC4-substitution and the LS-substitution concrete mixtures. Another
462 known PC4 impurity, PBDEs (polybrominated diphenyl ethers) were tested in the leachate but
463 found to be undetectable.

464 **3.4. Greenhouse Gas Emissions**

465 The GHG emissions were examined for each mixture to understand possible reductions in
466 emissions that could be achieved through utilizing this building material waste stream. When
467 PC4 and LS were used as a partial replacement for Portland cement, notable reductions in GHG
468 emissions were found (Figure 4). These findings were expected as the cement production is
469 responsible for the majority of the GHG emissions attributed to concrete production. For
470 mixtures where cement is replaced with 5 or 15% PC4 or LS by mass, the GHG emissions were
471 approximately 5 and 15% lower, respectively, reflecting the direct mass replacement of the high
472 emitting cement. Mixtures with PC4 used for cement replacement provided slightly greater
473 reductions than the LS replacement. However, the differences between the use of PC4 and the
474 use of LS were less than 0.1% and would be considered negligible within the confines of this
475 type of environmental impact assessment.

476 For the mixtures in which PC4 and LS were used as partial fine aggregate replacement,
477 marginal differences in GHG emissions were noted compared to the PS mixture. The

478 replacement of 5 and 15% of fine aggregate with LS led to negligible increases in GHG
 479 emissions compared to PC4 replacement. This minute increase was a function of the additional
 480 emissions from limestone excavation and processing. Though, since the cement content—the
 481 primary contributor to concrete GHG emissions—is the same as that of the control for all
 482 mixtures with fine aggregate replacement, changes to GHG emissions from the use of either LS
 483 or PC4 were small.



484 **Figure 4.** Estimated GHG emissions (kg CO₂-eq/m³ of concrete) for mixtures produced
 485 evaluated by major process for production
 486
 487

488 4. Conclusions

489 In this work, we address how a waste mineral stream from the building industry can be
 490 utilized in the production of cement-based materials. Specifically, this work examines how the
 491 mineral components in carpet could be used to reduce demand for virgin materials in cement-
 492 based materials. The development of new, environmentally sustainable materials and practices is

493 of great societal necessity. The utilization of minerals that are readily available and recycled
494 from the same industry could be a significant mechanism in improving environmental
495 sustainability. Currently, however, the understanding of how the calcium carbonate used in
496 carpets can influence the material attributes of concrete is extremely limited. A foundation of
497 how these recycled mineral waste streams influence cement-based material performance is
498 critical in addressing waste-minimization strategies and routes to reduce dependence on virgin-
499 mineral resources.

500 This research serves as a contribution toward understanding the viability of Post-Consumer
501 Carpet Calcium Carbonate (PC4) for use in concrete. In this work, 5 and 15% mass replacements
502 of Portland cement as well as 5 and 15% replacements of fine aggregate with PC4 were
503 examined. To draw comparisons with commonly used materials, the effects of using these
504 mixtures were assessed relative to control mixtures with Portland cement as the only powder in
505 the binder as well as the same mass replacements with limestone. Fresh and hardened concrete
506 properties were assessed, as was the potential for leaching from hardened concrete. There were
507 several key findings from this work:

- 508 • The use of PC4 as an inclusion in concrete, either as a partial cement replacement or
509 as a partial fine aggregate replacement, increases initial and final set times by up to
510 167% and 195%, respectively;
- 511 • The use of PC4 can lead to a lower density concrete (~10% decrease) that is more
512 porous (11% increase in air voids) than a mixture with only Portland cement as the
513 binder or only conventional aggregates;
- 514 • PC4 lowers the strength of concrete mixtures by up to 60%; however, this may not be
515 of concern for low strength applications;

- 516 • Concentrations of antimony (Sb) leached from concretes containing PC4 were below
517 maximum safety levels; however, PC4 with higher Sb levels may surpass these limits;
- 518 • PC4, when used to replace cement, can lead to reduction up to 15% in GHG
519 emissions. However, such benefits should be weighed in the context of any shifts in
520 necessary performance.

521 In future studies, the effects of any variability between PC4 batches, such as those that might
522 occur as a function of the carpet fiber type, age of carpet, or recycling process, should be
523 examined. Additionally, the effects of PC4 on other durability properties of concrete should be
524 assessed. Future work should address possible beneficial traits that might occur from the low-
525 density PC4 mixtures, such as changes in thermal conductivity. Despite these areas for further
526 investigation, this work provides insight into the feasibility of utilizing PC4 as a constituent in
527 concrete. Noting the importance of decreasing the impact of concretes, the need to minimize
528 waste streams, and the value of immobilizing wastes in a long-service life material, such as
529 concrete, these findings suggest there is a potential market for PC4 in cement-based materials.
530 Due to alterations in material properties noted from this work, it is possible that PC4 would be
531 best utilized in low-strength applications, such as pavers. However, with further study and
532 refinement of processing methods, it is possible that additional uses for PC4 in cement-based
533 materials can be elucidated.

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537 quarried limestone tested, respectively. This work was supported by the Carpet America
538 Recovery Effort. Views expressed in this paper are those of the authors.

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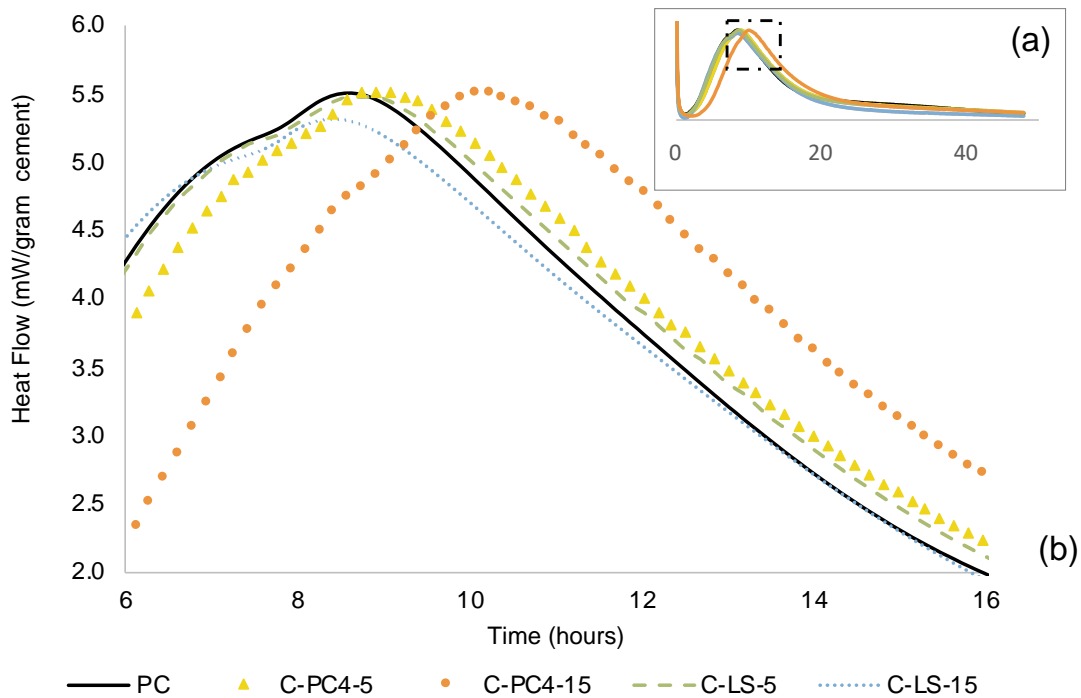
542 **Appendix A**

543 **Table A1.** Gradations for PC4, quarried ground limestone, fine aggregate, and coarse aggregate

Sieve Specification	PC4, % Passing	Ground Limestone, % Passing	5 mm Aggregate, % Passing	10 mm Aggregate, % Passing	25 mm Aggregate, % Passing
37.5 (1-1/2 in)	-	-	-	100.00%	100.00%
25 (1 in)	-	-	-	100.00%	100.00%
19 mm (3/4 in)	-	-	-	100.00%	94.1%
12.5 mm (1/2 in)	-	-	-	100.00%	47.8%
9.5 mm (3/8 in)	-	-	-	99.2%	15.7%
4.75 mm (No. 4)	100%	-	99.95%	17.7%	0.6%
2.36 mm (No. 8)	100%	-	89.1%	2.1%	0.2%
2.00 mm (No. 10)	100%	-	80.5%	-	-
1.18 mm (No. 16)	99.7%	-	56.3%	-	-
600 μm (No. 30)	99.9%	-	29.9%	-	-
420 μm (No. 40)	99.39%	100%	18.9%	-	-
300 μm (No. 50)	98.88%	100%	10.4%	-	-
150 μm (No. 100)	64.47%	100%	2.1%	-	-
125 μm (No. 120)	-	99.8%	-	-	-
75 μm (No. 200)	23.65%	95.8%	0.2%	-	-
45 μm (No. 325)	-	82.0%	-	-	-

544 “-” indicates sieve sizes not tested

545 **Appendix B**



546 **Figure B1.** Heat flow normalized to mass of cement of different binder mixtures with 5% or
 547 15% replacement of cement with either PC4 or Limestone showing (a) Heat flow over the 48-
 548 hour testing period and (b) heat flow for the region indicated by the rectangle in (a).
 549
 550

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