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Sustainable and Cost-Efficient Cement Clinker Reduction: Retaining Strength and Lowering Carbon Emissions

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ABSTRACT

The Portland cement (PC) industry significantly contributes to global CO2 emissions, accounting for 5% to 8% of annual anthropogenic emissions. Supplementary cementitious materials (SCMs) offer sustainable alternatives that reduce the environmental impact of PC concrete. Still, the increased cost of conventional SCMs due to industry changes has created a need for more cost-effective solutions. Pulverised limestone (PL) has emerged as a promising alternative due to its abundance and cost-efficiency. While PL can reduce clinker by up to 15% in North America and 35% in Europe, it often leads to a significant decrease in compressive strength. Recent studies suggest that the ideal clinker replacement is around 12% by weight, beyond which strength is compromised. To produce eco-friendly concrete block, in this study, a new proprietary mineral admixture called Duraflex was assessed for its potential to maintain strength in cementitious pastes with higher PL content. Duraflex has previously shown promise in improving soil stabilization by enhancing strength and reducing porosity. The findings indicate that small amounts of Duraflex (2%) can effectively retain strength in cement mixes containing up to 30% PL, surpassing the performance of conventional SCMs. While PL replacement beyond 12-15% generally reduces strength, Duraflex mitigates this effect, allowing for greater clinker reduction without sacrificing performance. The study shows that using 30% PL with 2% Duraflex could reduce greenhouse gas (GHG) emissions by up to 20% compared to current Portland Limestone Cement formulations, which achieve a 10% GHG reduction with lower strength. This research highlights the potential of combining PL with Duraflex as a sustainable and cost-efficient approach to reducing clinker content while maintaining concrete strength and workability and lowering carbon emissions. In addition, an alternative approach to the problem is shown to be almost ready to be transferred to industry.

KEYWORDS

compressive strength, concrete masonry units, duraflex, granulated blast furnace slag (GBFS), portland cement, pulverised limestone

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Introduction

The Portland cement (PC) industry is one of the largest global emitters of carbon dioxide (CO2), accounting for 5% to 8% of the annual anthropogenic emissions worldwide [1, 2]. For every kilogram of Portland cement produced, an equivalent amount of CO_2 is released into the atmosphere, exacerbating environmental degradation and contributing significantly to global warming. PC is one of the main pillars of "concrete" and the ubiquitous materials produced from it, such as blocks, pavers, and roof tiles; for instance, 6% to 10% of the mass of a concrete block is made up of cement [3, 4]. While calcination produces most of the CO_2, the producing PC requires high amounts of energy because temperatures around 1450°C are required to convert the raw materials into aluminates of lime and silicate [3]. According to Cheng et al. [1], global demand for PC will grow by 12% to 23% by 2050, and its annual emissions will need to decrease by at least 16% to align with the Paris Agreement 2015 [5] by 2030. Reducing CO_2 emissions is complex because several factors need to be considered collectively - changes in the high greenhouse gas (GHG) emitting raw materials, decreasing the energy used in the extraction process, using cleaner energy in the production process, and reducing the amount of cement used in the construction process.

Supplementary cementitious materials (SCMs) offer sustainable alternatives that reduce the global warming potential of PC concrete without sacrificing its performance [6]. SCMs are divided into conventional, like fly ash (FA), ground granulated blast furnace slag, silica fume, and natural pozzolans, which are recognized in industry standards and known as by-products of established industries, and alternative SCMs such as calcinated clay, rice husk ash, sugarcane bagasse ash, and recycled glass powder that are recognized as substitutes for conventional SCMs and known as agricultural waste, recycled materials, or natural minerals. Pulverised limestone (PL) has been used as a filler. By substituting the PC clinker with SCMs or a filler, the pressure on the cement supply chain will be reduced, providing more alternatives, potentially reducing the cost, decreasing the GHG emissions, and achieving sustainable development goals. Ground granulated blast furnace slag, which acts as hydraulic material that hardens when in cement, allows for 35-50% clinker substitution, while FA, which acts as a pozzolanic material forming additional C-(A-)S-H (calcium (alumino)silicate hydrate), allows only 10-20% clinker substitution. Recently, using conventional SCMs has increased the cost of cement due to their scarcity because of changes in industry processes. For example, Fly Ash is a by-product of burning coal, but coal burning has been significantly reduced, yielding less FA and increasing its cost. Until recently, PL powder was used to reduce clinker by only 5%: however, cement factories are now permitted to reduce the clinker by up to 15% in North America and 35% in Europe [7]. Research studies have found that substituting 2% of the clinker with PL results in a Portland Limestone Cement (PLC) that can have 10% higher compressive strength than Ordinary Portland Cement. Substituting between 2%-13% of the clinker with PL provides PLC that performs slightly better but substituting more than 13% of the clinker with PL produces weaker PLC [8].

The dilution of the clinker by a filler that is relatively inert reduces the quantity of material that is available for reaction, which effectively increases the water-to-clinker ratio in the cement paste. This dilution, however, decreases the chemical shrinkage, raises the overall degree of hydration of the cement paste, and increases the flowability and slump [7]. Moreover, the increase in surface area attributed to fine filler particles can promote heterogeneous nucleation of the hydration products on the surfaces of the limestone particles, leading to more rapid hydration rates [7]. The nucleation of calcium silicate hydrate (C-S-H) on the surfaces of the limestone precipitates in the open capillary pores which facilitates the reduction of their volume and partially compensates for the dilution [9, 10]. In addition, calcium ions get released into the pore solution of the cement matrix due to the dissolution of the limestone, which increases the tendency to precipitate more C-S-H gel [9, 11].

Hence, adding limestone to cement may result in increasing the matrix density and consequently the compressive strength. However, the influence of limestone on the mechanical properties varies based on the amount of limestone used. This is because the change in matrix density and in turn, its strength, depends on the balance between the negative impact of dilution and the positive impact of improved hydration and C-S-H formation. This balance depends on the water/binder ratio and the percentage of limestone in the mix, which determines the overall effect of replacing the cement with limestone on the strength.

Some studies have been conducted on the use of PL and other conventional SCMs [8] because the addition of more than 15% PL affects the mechanical properties of the product negatively. Thus, while PL has been used to reduce the amount of clinker in cement, PL does not provide sufficient confidence for the cement industry to shift from conventional to alternative SCMs for major clinker reduction. Thus, the use of PL only decreases the GHG emissions of PC production by about 10% [9].

One of the important mechanical properties of cementitious products, including concrete and mortar, is compressive strength. Most studies examine 7d-fc (compressive strength at 7 days age) and 28d-fc (compressive strength at 28 days age) after casting. In North America, testing was conducted on concrete with 12% wt. of PL [8] with a 0.43 water/binder (w/b) ratio and by Thomas et al. [12] with different w/b ratios. It was found that the 7d-fc and 28d-fc increased by 6% and 15%, respectively, when compared to the reference mix with 0% wt. of PL. In contrast, when Chong et al. [13] cast mortar cubes with 15% and 35% wt. of PL and approximately 0.4 w/b, they found that the compressive strength decreased in all 4 cases when their strengths were compared to those of the control specimens: 15% wt. of PL decreased 7d-fc by 9% and 35% wt. of PL decreased 7d-fc by 27%, while 15% wt. of PL decreased 28d-fc by 6% and 35% wt. of PL decreased 28d-fc by 22%. In Europe, Alunno-Rossetti and Curcio [14] and Tsivilis et al. [15, 16] studied the effect of adding 20% wt. of PL to concrete and found that 7d-fc and 28d-fc were like the reference mixes' strengths with 10% deviation. Tsivilis et al. [15, 16] also found that adding between 20% to 35% wt. of PL to concrete reduced 7d-fc and 28d-fc by 20% to 40%. In other parts of the world, Muthu et al. [9] studied the addition of 35% wt. of PL to concrete and different w/b 0.35, 0.4, 0.45, and 0.5 and found that 28d-fc was reduced by 36% to 54%. Furthermore, Her et al. [17] reported that adding 10% to 30% wt. of PL to mortar samples reduced the 7d-fc and 28d-fc by 3% to 40% and 5% to 30%, respectively.

Clearly, using PL in cementitious products, including mortar and concrete, promotes the reduction of GHG emissions in terms of the amount of PC clinker in the cement. Some studies show that the ideal replacement appears to be approximately 12% wt. of PL, after which the strength decreases. Tennis et al. [8] mentioned that the cement industries are producing Portland cement in North America including, but not limited to, Type IL in the USA as per ASTM C595 and AASHTO M 240 and GUL, MSL, HEL, and HSL in Canada as per CSA A3001, with up to 15% wt. of PL and 10% reduction in the compressive strength as outlined in the literature. On the other hand, European cement industries are producing Portland cement including, but not limited to, CEM II/A-L and CEM II/B-L as per EN 197-1, with approximately 6 to 20% and 21% to 35% wt. of PL respectively and 40-50% reduction in the compressive strength.

Based on the above, recent work in Calgary has concentrated on reducing the GHGs associated with the production of concrete blocks, specifically by reducing the amount of PC clinker in the concrete mix. Two approaches have been explored. The first is described in detail in a companion paper in these proceedings by Ghasemalizadeh and Khoshnazar [18]. That approach is well advanced and involves the use of ground granulated blast furnace slag. More recently, a second approach has been investigated using a new proprietary mineral admixture called Duraflex. This admixture was used to enhance soil stabilization by mixing the soil with PC and Duraflex. The mixes including Duraflex at 2% and 5% by weight of cement were found promising from previous unpublished research studies as they increased strength and durability by reducing porosity and increasing the density when compared to the control specimens without Duraflex.

The idea of using the Duraflex admixture to reduce the amount of PC clinker substantially while potentially retaining compressive strength was therefore investigated.

METHODOLOGY

The current goal of the second approach is to determine an appropriate cementitious paste mix with different PL, Duraflex wt., and w/b ratios that will provide a similar compressive strength to the traditional cementitious paste mix. In addition, the effect of Duraflex on flowability as per ASTM C230 is being examined to maintain workability.

Materials

Quikrete GU (General Use) Portland cement was used in all the tests. A mix of 95% clinker and 5% PL by wt. were interground as per manufacturer specifications and CSA A3001. The PL was acquired from Graymont with a particle size of 150 microns, while Duraflex (a powder) was obtained from Duraflex Solutions Global. In the mixes with Duraflex, the Duraflex, and Graymont PL are not interground with the Portland cement but rather added separately to the mix in different wt. as shown in Table 1.

Mix Design and Testing

The mixes implemented in this study are based on a trial-and-error methodology. Two different w/b ratios were used, 0.3 and 0.42. These were selected as one being close to stoichiometric and the other more typical of use in PC concretes. The 0.42 w/b was selected as an average value following the studies by Ghasemalizadeh and Khoshnazar [19], Chong et al. [13], and Tennis et al. [8], which have w/b ranging between 0.4-0.43. The 0.42 was used in 10 mixes and 1 control mix resulting in high flowability and low compressive strength results. Since block manufacturing requires a mix with no slump, the w/b was reduced to 0.3 w/b to produce less flowable mixes and to examine the effect of changing the w/b ratio on the compressive strength. The nomenclature is defined by the percentages wt. of clinker, PL, Duraflex and the w/b ratio; for example, C83%-PL15%-D2%-0.3 means 83% wt. of clinker, 15% wt. of PL, 2% wt. of Duraflex, and a 0.3 w/b ratio, while the ordinary Portland cement with a 0.42 w/b ratio will be C95%-PL5%-D0%-0.42. The mixtures are summarized in Table 1.

Table 1: Specimens and Flowability as per ASTM C230

Mixes	Clinker (%)	PL (%)	Duraflex (%)	Flowability (%)
C95%-PL5%-D0%-0.42	95.00	5.00	0.00	165
C95%-PL5%-D0%-0.3	95.00	5.00	0.00	80
C90%-PL5%-D5%-0.42	90.25	4.75	5.00	200
C93%-PL5%-D2%-0.42	93.10	4.90	2.00	110
C90%-PL5%-D5%-0.3	90.25	4.75	5.00	68
C93%-PL5%-D2%-0.3	93.10	4.90	2.00	66
C86%-PL15%-D0%-0.42	85.50	14.50	0.00	225
C76%-PL24%-D0%-0.42	76.00	24.00	0.00	230
C67%-PL34%-D0%-0.42	66.50	33.50	0.00	230
C57%-PL43%-D0%-0.42	57.00	43.00	0.00	230
C57%-PL43%-D0%-0.3	57.00	43.00	0.00	100
C81%-PL14%-D5%-0.42	80.75	14.25	5.00	218
C71%-PL24%-D5%-0.42	71.25	23.75	5.00	228
C61%-PL33%-D5%-0.42	61.75	33.25	5.00	230

C52%-PL43%-D5%-0.42	52.25	42.75	5.00	230
C57%-PL41%-D2%-0.3	57.00	41.00	2.00	90
C69%-PL29%-D2%-0.3	68.89	29.11	2.00	80
C70%-PL28%-D1%-0.3	70.30	28.30	1.40	85

The specimens were prepared to substitute between 15%-35% wt. of clinker as defined in the literature, and specimens with 43% wt. were tested to examine the possibility of exceeding the wt. of clinker replacement stated in the literature without sacrificing the mechanical properties. The specimens were cast in 50 mm cubes as per ASTM C109 as shown in Figure 1 (a). The mixture which included Duraflex, PL, or both was initially dry mixed mechanically for 1 min by a KitchenAid stand mixer at slow speed 1-2. Then, the dry mix was added to water gradually and was mixed at a slow speed for 3 mins, average speed 4-6 for 1 min, and high speed 8-10 for 1 min. The mixing was done at room temperature (20°C) by the same person to guarantee consistency, following ASTM C305. The flowability test was applied by calculating the average diameter of 4 different measurements with a caliper in 4 different directions and then applying Eq. (1), as per ASTM C230, to calculate the flowability percentage.

$$(1)\ Flowability\,\% = \frac{Avg.Dia\,(mm) - 100}{100}\ x\,100\%$$

After casting, the specimens were covered with plastic wrap for 24 hrs, then demolded and placed in a closed fog room with controlled temperature and humidity (20°C, 100% RH) for the desired curing periods. Compressive strength was obtained by testing sets of three specimens at the ages of 7 and 28 days using the Riehle testing machine shown in Figure 1 (b).

(a) **(b)**

Figure 1: (a) Specimens in 50 mm cube molds, and (b) the Riehle testing machine

RESULTS AND DISCUSSION

As shown in Table 1, the flowability results show that in lower w/b ratios the addition of PL or 5% wt. of Duraflex increases the flowability, while the addition of 2% wt. of Duraflex decreases the flowability. Adding Duraflex and PL to the mix provides greater flowability than those of the reference mixes C95%-PL5%-D0%-0.3 or C95%-PL5%-D0%-0.42. Adding PL to the mix with 0.42 w/b ratio creates a highly flowable mix with about 200% to 250% flowability. For lower w/b ratios, it might be better to use a superplasticizer to increase the flowability.

Regarding 7d-fc and 28d-fc, two control samples were tested, with 0.3 w/c and 0.42 w/c ratios, to provide the reference strengths. Decreasing the w/b ratio increased the strength of the control sample by approximately 10%. All compressive strength results, including standard deviation (S.D.) and coefficient of variation (C.O.V.), are summarized in Table 2. As shown in Figure 2, four different mixes with w/b ratios of 0.3 and 0.42, and wt. of Duraflex, 2% and 5%, were tested: the resulting strengths and standard deviations are plotted on the bar chart. It was found that using the lower w/b ratio and only 2% Duraflex caused the greatest increase in strength. Adding more Duraflex did not yield more strength compared to the reference values or those with only 2% Duraflex.

Table 2: Compressive strength testing results, standard deviations, and covariances

Mixes	7d-fc (MPa)	S.D. (MPa)	C.O.V. (%)	28d-fc (MPa)	S.D. (MPa)	C.O.V. (%)
C95%-PL5%-D0%-0.42	51.36	8.59	17	66.19	6.85	10
C95%-PL5%-D0%-0.3	57.24	12.63	22	67.03	4.58	7
C90%-PL5%-D5%-0.42	52.85	6.12	12	57.28	5.88	10
C93%-PL5%-D2%-0.42	44.84	9.17	20	43.94	3.03	7
C90%-PL5%-D5%-0.3	55.69	13.58	24	66.77	13.9	21
C93%-PL5%-D2%-0.3	70.69	10.03	14	71.47	14.78	21
C86%-PL15%-D0%-0.42	43.89	3.37	8	55.11	2.82	5
C76%-PL24%-D0%-0.42	40.81	1.79	4	43.29	4.75	11
C67%-PL34%-D0%-0.42	31.08	1.1	4	36.77	2.25	6
C57%-PL43%-D0%-0.42	24.44	1.65	7	27.81	4.38	16
C57%-PL43%-D0%-0.3	42.65	5.37	13	50.12	9.28	19
C81%-PL14%-D5%-0.42	44.53	3.35	8	56.76	5.28	9
C71%-PL24%-D5%-0.42	40.39	1.89	5	46.03	4.04	9
C61%-PL33%-D5%-0.42	28.35	3.03	11	37.01	5.91	16
C52%-PL43%-D5%-0.42	23.07	0.28	1	27.69	3.96	14
C57%-PL41%-D2%-0.3	46.97	5.6	12	52.61	4.73	9
C69%-PL29%-D2%-0.3	57.35	0.84	1	63.87	12.54	20
C70%-PL28%-D1%-0.3	59.25	2.99	5	59.96	9.69	16

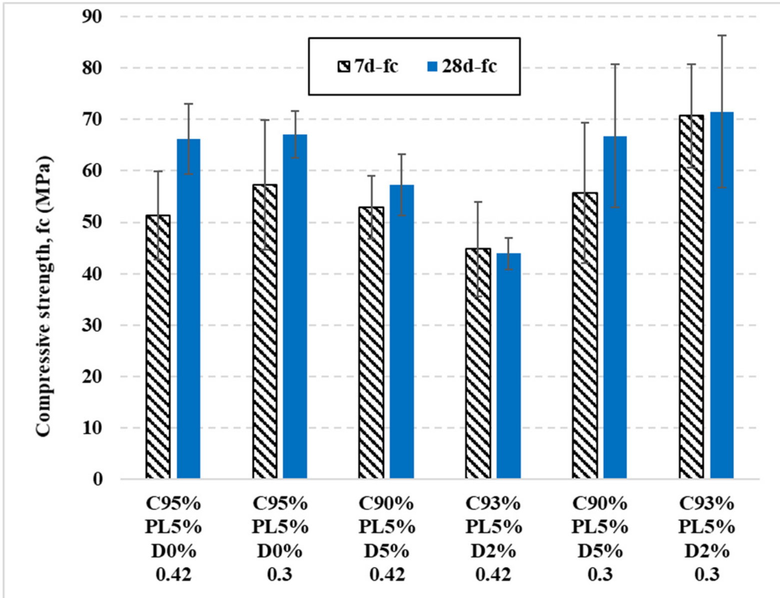


Figure 2: 7d-fc and 28d-fc for control specimens and specimens with Duraflex

Five different mixes with w/b ratios of 0.3 and 0.42, and wt. of PL, 15%, 24%, 34%, and 43% were tested: the compressive strength results are plotted in Figure 3, as well as their standard deviations. As with results in the literature, the 7d-fc and 28d-fc strengths were lower than the reference strengths by 15%-60%, when adding more PL because it dilutes the mix. However, decreasing the w/b ratio of the 43% PL mix increases the strength by 75%. 7d-fc and 28d-fc were decreased by approximately 25% for specimens with a w/b 0.3 and 43% wt. of PL and decreased by 55% for specimens with a w/b 0.42 and 43% wt. of PL. Moreover, the 7d-fc and 28d-fc for specimens with a w/b 0.42 and 34% wt. of PL were decreased by approximately 42%.

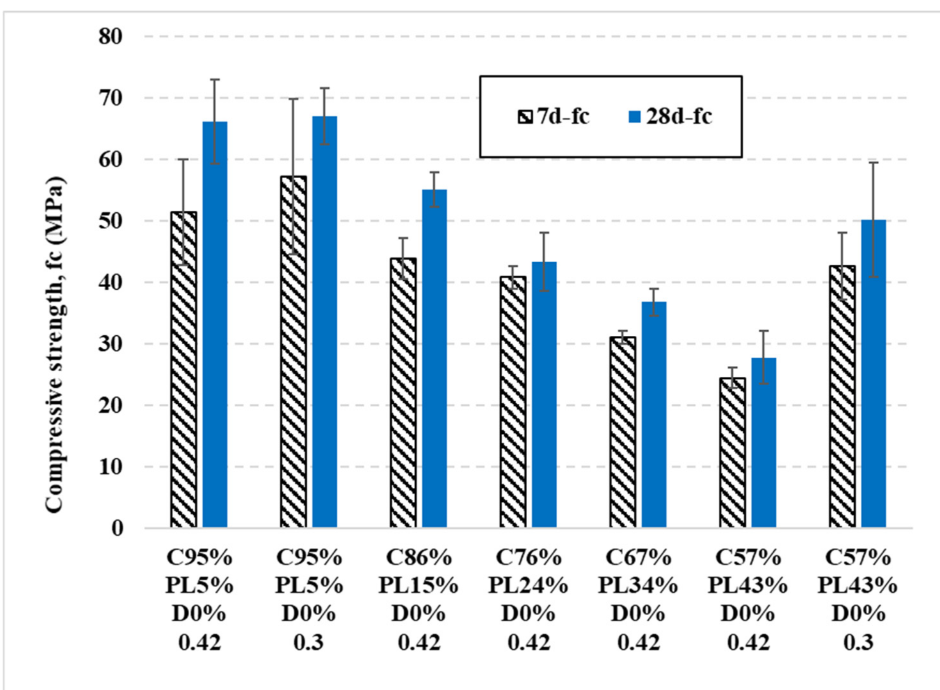


Figure 3: 7d-fc and 28d-fc for control specimens and specimens with wt. of PL

As shown in Figure 4, seven different mixes with w/b ratios of 0.3 and 0.42, and wt. of PL, 14%, 24%, 28%, 29%, 33%, 41%, and 43%, were tested, as well as their standard deviations are plotted on the bar chart. It was found that adding 5% wt. of Duraflex to the mixes with w/b 0.42 had no effect on or provided lower 7d-fc but yielded slightly higher 28d-fc, when compared to samples without Duraflex. However, the strength significantly improved when the w/b ratio was reduced to 0.3 and approximately 2% wt. of Duraflex was added to the mix with approximately 30% wt. of PL. For instance, the 7d-fc of C69%-PL29%-D2%-0.3 and C70%-PL28%-D1%-0.3 were statistically similar when the strengths were compared to the reference strength of the control specimens, while their 28d-fc were lower by 5% and 11% (see Table 3).

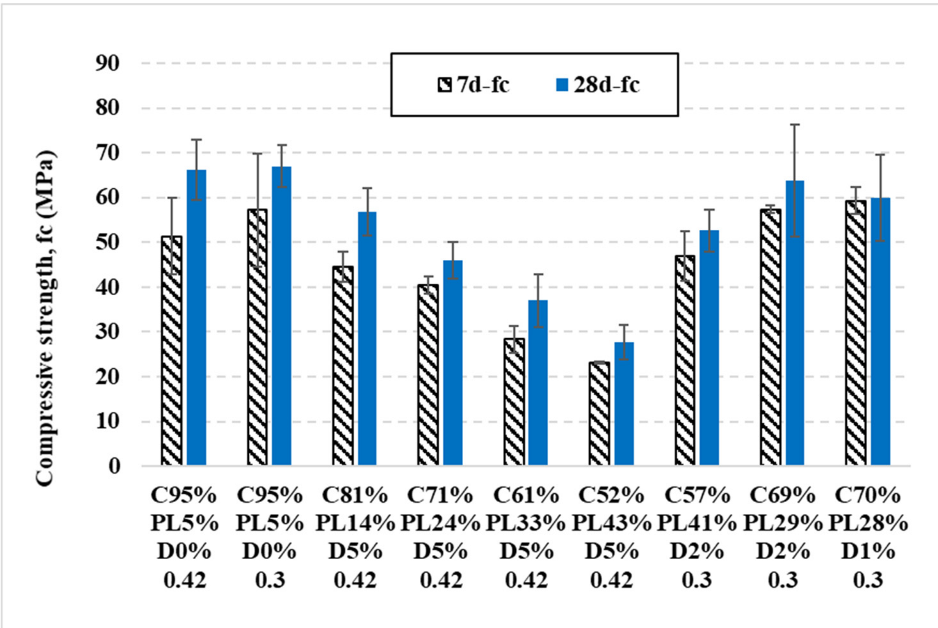


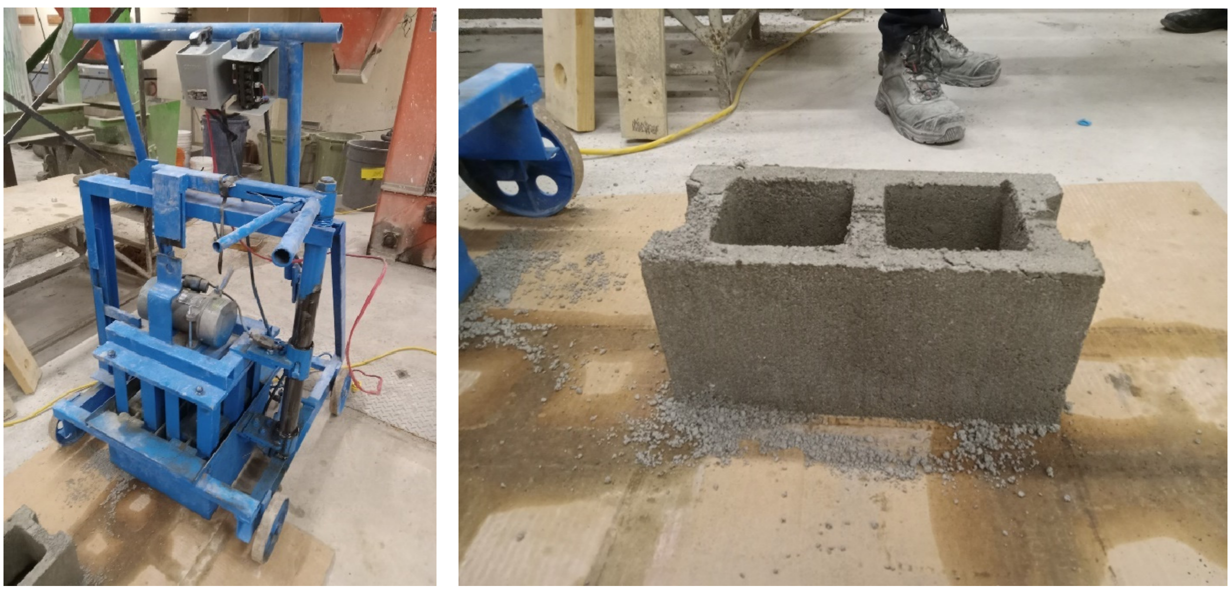
Figure 4: 7d-fc and 28d-fc for control specimens and specimens with wt. of Duraflex and PL

Table 3: 28d-fc T-test for C69%-PL29%-D2%-0.3, C70%-PL28%-D1%-0.3, and C95%-PL5%-D0%-0.3

Mix A			Mix B			t-value	critical t	Check	Interpretation
name	mean	S.D.	name	mean	S.D.				
C69% PL29% D2% 0.3	63.9	12.5	C70% PL28% D1% 0.3	60.0	9.7	0.427	2.776	Accept Null hypothesis	No difference
C69% PL29% D2% 0.3	63.9	12.5	C95% PL5% D0% 0.3	67.0	4.6	-0.410	2.776	Accept Null hypothesis	No difference
C70% PL28% D1% 0.3	60.0	9.79	C95% PL5% D0% 0.3	67.0	4.6	-1.143	2.776	Accept Null hypothesis	No difference

Conclusion and Recommendations

The work so far to produce "eco-friendly" concrete block mixes has led to two approaches that will have differing costs in Canada depending on location and thus the cost of the input materials. The first approach is nearing transfer to the industry while the second shows distinct promise but needs to shift from the basic stage to producing a concrete mix that can be used to create concrete blocks. Indeed, a concrete mix using the first approach and aggregate (recombined into a slightly different sieve distribution) from the local concrete block plant (Expocrete) has been used in our concrete block machine (Figure 5 (a)) to produce concrete block (Figure 5 (b)). Testing the concrete block has shown it to be weaker than the concrete itself and the face-shells to have different strengths. This is probably due to the weaker capability of the small block machine to impart enough energy to the concrete in the mold to achieve proper compaction as would be achieved in a machine in a block plant. Thus, units from this block machine have not been used to build and test concrete blockwork, but merely to show that the mix is viable, and can soon be transferred to industry.



(a) **(b)**

Figure 5: (a) The laboratory concrete block machine, and (b) a concrete block fabricated with it

The results of the preliminary work of the second approach show that it is possible to reduce the amount of clinker in a cement beyond the 15% from studies to date utilizing PL and Duraflex and maintaining strength. Duraflex is a mix of minerals that need to be mixed in a certain proportion – the GHG associated with this product is small compared to firing a kiln to produce PC clinker. Flowability can also be maintained, so use of a new "cement" containing Duraflex might also apply to the mortar used by masons. Thus, concrete blockwork does have the potential to be considerably more environmentally friendly than straight concrete. Details for approach 1 are provided elsewhere in these proceedings, but for approach 2, the following conclusions may be drawn:

- For flowability, it was found that adding 5% wt. of Duraflex, PL or both improves the flowability of the specimens, but adding 2% wt. of Duraflex to those mixes decreases the flowability.

- The optimal balance between the addition of Duraflex and the w/b ratio was found to be important for maximizing strength performance and clinker reduction. At a w/b ratio of 0.3, 2% wt. of Duraflex provided the highest strength, while at a w/b ratio of 0.42, 5% wt. of Duraflex was necessary to achieve significant performance gains. This could be attributed to the role of Duraflex in enhancing mix cohesion, increasing density, and reducing porosity, particularly in mixtures with higher water content, which may have contributed to improved overall performance despite clinker reduction.
- The addition of 15%-43% PL diluted the specimens and yielded lower strengths. However, the addition of 2% wt. of Duraflex to these mixes causes a significant improvement in compressive strength: specimens with approximately 30% wt. of PL and 2% wt. of Duraflex, C69%-PL29%-D2%-0.3 and C70%-PL28%-D1%-0.3, have similar strengths to those of the control specimens, C95%-PL5%-D0%-0.3.
- As the current product of Portland Limestone Cement with 15% PL reduces GHG emissions by approximately 10% but with lower compressive strength [9], the current study finds that 2% wt. of Duraflex has a potential to reduce approximately 20% GHG emissions, when it is mixed with approximately 30% wt. of PL, and yields similar strengths.

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