SUSTAINABLE CONCRETE PRODUCTION WITH RECYCLED CONCRETE WASH WATER BENEFICIATED WITH CO₂

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Abstract.

A significant amount of wastewater is generated through the cleaning of equipment utilized within the ready mixed concrete production cycle. The reuse of concrete wash water as mix water is limited by the negative material performance impacts associated with the suspended solids; the effects are exacerbated with increasing solids contents and water aging. A novel carbon dioxide treatment to allow the use of high solids wash water (specific gravity 1.10) as mix water was examined. Seven batches of concrete were produced and compared: a reference mix, two batches with untreated wash water and four batches with CO_2 treated wash water. The carbon dioxide treatment mineralized CO_2 at 28% by mass of the treated solids. Acceptable concrete was produced through adjusting admixtures for workability. The compressive strength at 1, 7, 28 and 56 days was improved relative to both the reference and the concrete produced with untreated wash water. The suspended solids containing mineralized CO_2 served as a viable cement replacement. The avoided cement and bound carbon dioxide served to lower the carbon impact of the concrete by about 14%. The approach allows three waste streams (CO_2 , wash water and wash water solids) to be reused to produce more sustainable concrete.

KEYWORDS: CO₂ utilization, concrete, sustainability.

1. INTRODUCTION

Awareness of the environmental impacts of the cement and concrete industries, combined with increasing global infrastructure demands, has resulted in a greater emphasis on improving concrete sustainability. Strategic roadmaps and technology assessments have recognized the shortcomings of traditional levers for reducing the carbon impact of the industries and the need for emerging technologies [1]. Action is require throughout the supply chain; cement producers, concrete producers, structural engineers and construction companies each have roles to play in the pursuit of a net zero carbon vision [2].

Within the context of emerging technologies lies the concept of CO_2 utilization [3]. In this approach, carbon dioxide is used directly or as a feedstock to produce valuable carbon-containing products. One use is for the manufacture of construction materials; in contrast to other utilization pathways, such as the production of fuels, the carbon dioxide is mineralized and permanently removed from the atmosphere [4, 5]. Construction has been identified as the largest market opportunity for the CO_2 utilization field with technologies in concrete and aggregate production offering a potential to utilize 5.0 Gt CO_2 and generate \$550B per year by 2030 [6].

The present work concerns an investigation into the use of carbon dioxide to beneficially treat concrete wash water for its reuse as mix water. The production of ready mixed concrete involves the washing of equipment whether it be a central mixer or individual trucks. After the concrete has been discharged the mixer may be washed out to prepare it for a new load of concrete. The water that is washed from the truck is laden with very fine sand and cementitious materials. The handling and disposal of this wastewater represents a significant operational, regulatory and logistical burden for the concrete producer. The most common approaches to address wash water are clarification and discharge, reclaimers, hydration stabilizing admixtures or no action at all.

Ideally the wash water can be recycled into new concrete but performance issues often arise when it is used as mix water in lieu of fresh water [7]. The approach can lead to concrete with increased water demand (and lower compressive strengths) and undesirable set acceleration. Both of these impacts worsen with the age of water meaning that the performance outcomes are sensitive to the "freshness" of the wash water. In practice, the solution has been to specify that mix water must have less than 50,000 ppm solids [8]. In cases where wash water is used in new concrete it is often merely diluted with fresh water to reduce the negative impacts. While the practice of clarifying water and disposing solids is the current status quo, the industry is being directed towards zero discharge practices and ready mixed producers are looking to decrease their water usage and carbon footprints [9].

The present study concerns the CO_2 treatment of

Batch	Control	NT	NTA	CT	СТА	CTP	CTAP
WW Treatment	n/a	None	None	$\rm CO_2$	$\rm CO_2$	$\rm CO_2$	$\rm CO_2$
Cement (kg/m^3)	267	267	267	267	267	240	240
$Limestone(kg/m^3)$	63	33	33	30	30	57	57
WW solids (kg/m^3)	0	30	30	33	33	33	33
Total powder (kg/m^3)	330	330	330	330	330	330	330
PCE $(\%/b)$	0.34	0.34	0.34	0.34	0.34	1.15	0.77
Dispersant $(\%/b)$	0	0	0.66	0	0.74	0	0.74
Relative binder	100	91%	91%	90%	90%	90%	90%

TABLE 1. Mix design of concrete produced during test program. WW refers to wash water. WW solids are the suspended solids present in the wash water that are incorporated into the concrete mix. PCE refers to a polycarboxylate plasticizing admixture. Admixture dosages are in % with respect to the binder content.

high solids concrete wash water for reuse, without dilution, as concrete mix water. Carbon dioxide is readily mineralized into calcium carbonate upon reaction with tricalcium or dicalcium silicate [10, 11] or calcium hydroxide [12], Wherein the carbon dioxide is permanently removed from the atmosphere it was studied what impact the carbon dioxide would have on the use of wash water as mix water and whether performance-enhanced concrete could be produced.

2. EXPERIMENTAL

A simulated concrete wash water was produced by mixing 7.7 kg of cement with 18 litres of water. The slurry was mixed in a drum mixer at 24 rpm for 3 hours to simulate a production/delivery/washing cycle. The slurry was then added to a treatment vessel. The total water in the reactor vessel was brought up to 50 litres by adding 7 litres of water through wash out of the drum mixer and a final 25 litres of water. The slurry was designed to have a specific gravity of 1.10 or more than three times the nominal optional limit for concrete mix water that appears in many specifications such as ASTM [8].

The supply of slurry was maintained under agitation in a barrel adapted as a reactor vessel. An immersible pump served to circulate the slurry through a conduit that was routed up and out of the slurry before bending back down to the vessel bottom whereupon the pumped slurry was ejected. Wash water subjected to a CO_2 treatment received an injection of carbon dioxide gas at 600 litres per hour. The injection of CO_2 gas was integrated into the base of the conduit where it was connected to the exit of the pump. The conduit, approximately 1.2 meters in total length, served to increase the potential residence time of gas inside the slurry given that any bubbles would rise through the slurry via buoyancy. Three different batches of slurry were made (one untreated, two treated) and each batch of slurry was used to make two batches of concrete.

Both treated and untreated wash water slurries were kept agitated in a minimally rotating drum mixer through to 24 hours prior to use in concrete. Samples of the treated slurry were taken periodically throughout the treatment cycle which concluded when the pH had declined from an originally elevated level to a neutral level of about 7.0. The carbon uptake and bound water of the cement slurry solids was determined through thermogravimetric analysis. A second assessment of the bound carbon was completed using an ELTRA CS 800 carbon sulphur analyser (induction furnace).

A series of seven concrete mixes were produced and are summarized in Table 1. The batch size was 1 m^3 . The default (reference) mix design had a w/b of 0.55. The binder content totalled 330 kg/m^3 and was comprised of 81% cement and 19% limestone. Two batches were produced with untreated wash water comparing concrete excluding and including a retarder as a dispersant to improve slump (marked NT and NTA for not treated and not treated plus dispersant admix). Likewise, two analogous batches were produced with CO_2 -beneficiated wash water (marked CT and CTA for CO₂-treated and CO₂-treated plus dispersant admix). Lastly, an additional two batches were produced with CO₂-beneficiated wash water wherein the level of the superplasticizing admixture was increased and either excluded or included the dispersant addition (marked CTP and CTAP for CO₂treated plus plasticizer and CO₂-treated plus admix plus plasticizer).

Adjustments were made to the binder loadings of batches containing wash water in order to compensate for the additional solids present in the wash water slurry. The total powder content (cement + limestone + wash water solids) was maintained at 330 kg/m³. Reductions were made to the limestone in the first four cases (NT, NTA, CT, CTA) while a proportional reduction to both cement and limestone was investigated in the final case (CTP, CTAP). In the final case the admixture loading was changed with respect to the reference and the adjustments were determined with respect to what was required to achieve the target slump. The dispersant loadings were in proportion to the wash water solids.

The concrete was measured in terms of slump and compressive strength at 24 hours, 7 days, 28 days and



FIGURE 1. Carbon uptake in wash water solids and solution pH with treatment time of wash water slurry treated with CO_2 .

56 days with the exception of the batches containing increased plasticizer (PCE) which were assessed only at 7 and 28 days.

3. Results

3.1. CO_2 treatment of the slurry

The CO_2 uptake and pH data over time for the two treated slurries closely agreed. One case is presented in Figure 1 and discussed herein. The pH of the treated slurry was 12.7 after three hours of prehydration. The pH declined upon addition of the CO_2 and was only a modest decline at first, down to 12.1 after 60 minutes, before a more rapid drop leading to a pH of 7.1 at 150 minutes. The injection of the CO_2 into the agitated slurry functionally changed when the treatment neared completion; whereas the initial injection of carbon dioxide did not result in any bubbles of carbon dioxide gas reaching the surface of and exiting from the slurry, the slurry displayed extensive bubbling around the time of neutralization.

The carbon content of the slurry solids was observed to increase with treatment time in a broadly linear fashion over the first 120 minutes of treatment. Ultimately the treatment was complete after 150 minutes at which time the solids were 28.1% CO₂ by mass. The carbon contents, as presented in the figure, have been normalized to the same ignited mass to express the uptake as a (percentage) mass of carbon dioxide per mass of the original anhydrous cement. This allowed for a simple comparison against the amount of CO₂ delivered, which can be described in the same terms. The uptake at the conclusion of the treatment was 41.4% CO₂ by weight of cement under the TGA analysis and 37.4% with the induction furnace. The uptake can be used to calculate process efficiencies. The injection of CO_2 gas at 600 LPH with a density of 1.98 g/L over 2.5 hours means that 2.97 kg of CO_2 was delivered. Where the initial slurry contained 7.7 kg of cement, the uptake rate via the TGA translates into 3.19 kg of bound CO_2 for an efficiency of 107% (a more robust TGA analysis that includes unattributed weight losses and a true LOI would serve to decrease this calculated value). Using the uptake rate via induction furnace there were 2.88 kg of CO_2 mineralized for a conversion efficiency of 97%.

The bound water content was normalized in the same fashion as the bound carbon dioxide. It was observed that the bound water increased over the first 90 minutes of the treatment before declining. In particular the decline in pH may have changed the equilibrium of any hydrate phases. For example, ettringite is destabilized below a pH 10.7 while monosulphate disappears below 11.6 [13].

The change in rate of the uptake curve between 120 and 150 minutes attests to a slowing down of the reaction. The delivery of the carbon dioxide was not changed so at this time the entering carbon dioxide is tending to change the pH of the solution moreso than mineralize with the slurry solids.

3.2. Fresh properties

The slump measurements of the seven batches of concrete are presented in Figure 2. The slump of the control was 19 cm after the conclusion of mixing (5 minutes after the addition of water) and was used as a target in later batches that were adjusted for slump. The untreated wash water served to reduce the slump by about 25% while an addition of the dispersant in turn increased the slump by 10%. The slump of the batch with the CO_2 treated slurry had



FIGURE 2. Workability (slump) of concrete batches produced with wash water slurry. Admixture loadings of dispersant and PCE are indicated as symbols read from the secondary vertical axis.

Batch	Control	NT	NTA	CT	CTA	CTP	CTAP
24 h strength (MPa)	10.2	10.8	10.0	14.6	13.4	n/a	n/a
28 d strength (MPa)	25.1 32.6	$\frac{26.0}{32.3}$	$\frac{29.9}{35.8}$	$\frac{30.0}{36.0}$	31.0 36.9	$\frac{26.4}{33.0}$	$29.7 \\ 36.2$
56 d strength (MPa)	35.0	n/a	36.9	38.9	39.9	n/a	n/a
Relative strength 24 h	100%	106%	98 %	143%	131%	- 105 07	
Relative strength 7 d Relative strength 28 d	100% 100%	104% 99%	119% 110%	120% 110%	124% 113%	105% 101%	118% 111%
Relative strength 56 d	100%	_	105%	111%	114%	_	_

TABLE 2. Compressive strength of concrete mixes from 24 hours to 56 days.

a major reduction (75%) in workability and, as in the untreated slurry case, an improvement in slump was observed when the dispersant was used. However, in either case, it was observed that the use of the treated slurry resulted in a workability loss.

When the addition of a superplasticizer was explored the slump was improved from 9.5 cm to 18.5 cm to effectively match the target. The addition of the dispersant before the suplerplasticizer allowed the same flowability to be achieved albeit with 33% less PCE. The workability issues can be solved through the use of conventional admixtures.

3.3. Compressive strength

The summary of the compressive strength data is presented in Table 2. Reported compressive strengths are averages of three specimens. The untreated wash water had a small positive effect at the two earliest ages but was neutral at 28 days. The addition of the dispersant resulted in improved performance at ages of 7 days and beyond, albeit diminishing relative to the control from 19% better at 7 days to 5% better at 56 days.

The carbonation treatment served to improve the concrete strength at all ages of batches so produced. The benefit was greatest at the earliest age with a

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43% benefit at 24 hours. As with the untreated slurry, the relative strength benefit declined with increasing age but remained at 11% at 56 days. The batch that included the dispersant was not quite as strong at 24 hours (a 31% benefit) but showed higher strengths than the batch without the dispersant at all subsequent ages concluding at a 14% benefit at 28 days. While the batches with the treated slurry were stronger than the control they also showed a reduced workability that would render them unviable.

The batches with the treated slurry and the PCE had a 10% lower cement content than the control. Even so, the 7 and 28 day strengths were acceptable and slightly above the control. As with the other examples of using the dispersant, the strength was increased further with an 11% improvement observed at 28 days, despite the cement reduction.

4. DISCUSSION

The compressive strength results are best compared in terms of the binder efficiency. The comparisons between the control and the batches with acceptable workability and using the treated slurry are outlined in Table 3. The cementing efficiency was calculated in terms of the cement clinker (assuming a clinker factor in the cement of 92% as per the Cembureau EPD

Batch	Control	CTP	CTAP
Clinker Efficiency 28d (kg clinker $m^{-3} MPa^{-1}$)	7.5	6.7	6.1
Cement Efficiency 28d (kg cement $m^{-3} MPa^{-1}$)	8.2	7.3	6.6
Binder Efficiency 28d (kg binder $m^{-3} MPa^{-1}$)	10.1	9.0	8.2
Powder efficiency 28d (kg powder $m^{-3} MPa^{-1}$)	10.1	10.0	9.1
Carbon efficiency (kg $CO_2 m^{-3} MPa^{-1}$)	7.4	6.3	5.7
Anticipated strength via cement loading (MPa)	32.6	29.3	29.3
Actual strength (MPa)	32.6	33.0	36.2
Change in strength (MPa)	n/a	3.7	6.9
Equivalent units of cement (kg)	n/a	30.0	56.2
Actual units of WW solids (kg)	n/a	33	33
Units WW solids/unit cement	n/a	0.91	1.71

TABLE 3. Cementing efficiency summary of control and batches with treated slurry.

[14]), the cement content, the overall binder content (cement + limestone), the overall powder content (cement + limestone + wash water solids) and the carbon impact [15] (relevant to the binder mixture of cement + limestone + wash water solids). The carbon intensity of the cement is taken to be 0.898 t CO_2/t tonne cement [14] while that of the limestone is taken as 0.008 t CO_2/t cement [16].

It was observed that the clinker, cement and binder efficiencies of the batches produced with wash water were improved 11% over control in the CTP batch. Whereas the control produced concrete with a 28 day strength of 32.6 MPa and 267 kg cement/ m^3 concrete, the CTP batch was 33.0 MPa with 240 kg of cement. If the cement was to provide strength with the same efficiency in the latter as it had in the former then the expected strength was 29.3 MPa. The difference between the actual strength and the expected, +3.7MPa, can be attributed to the wash water since the overall powder loading was unchanged. Whereas 3.7 MPa of strength would be associated with 30 kg/m^3 of cement the CTP batch contained 33 kg of wash water solids. The powder efficiency was unchanged in comparison to the control while the carbon the efficiency was improved 15%.

There appeared to be a synergy upon adding both the dispersant and the PCE (batch CTAP). All of the clinker, cement, and binder efficiencies were improved 19% as compared to the control. In this instance the actual strength was +6.9 MPa greater than the expected strength as per the cement efficiency of the control and cement loading of the batch. The strength difference is equivalent to 56.2 kg of cement. The incorporation of 33 kg of wash water solids meant that the solids had a cement replacement efficiency of 1.7 units of cement per unit of solids. The powder efficiency was improved 10% in comparison to the control while the carbon the efficiency was improved 22%.

The sustainability impacts can be achieved with respect to the mineralized carbon dioxide in the wash water solids, the avoided cement as part of the mix design modification, and recycling of waste cement and avoided potable mix water. The direct CO_2 impacts are estimated from the uptake of carbon dioxide into the wash water. The uptake of 28% CO₂ by weight of solids determined that the 33 kg of wash water solids/ m^3 concrete contained 9.2 kg of mineralized CO_2 . In accordance with the Cembureau EPD for Portland cement, the 27 kg of cement avoided were associated with 24.2 kg of carbon dioxide. The combined carbon reduction was 35.4 kg of CO_2 of which the avoided was 75% and the mineralized was 25%. The carbon impact of the binder was reduced 14% from 240.3 kg CO_2/m^3 concrete in the reference to 206.8 kg CO_2 m³ concrete in the two batches with CO_2 treated slurry. The replacement of the fresh mix water meant that $182 \text{ kg fresh water/m}^3$ concrete was saved.

The untreated wash water produced acceptable concrete with workability better than in the CO_2 treated case and acceptable strength albeit not as high as in the CO_2 treated case. The performance of the concrete produced with the untreated wash water would be expected to vary with age of the wash water. It would be less predictable and burdensome to the producer given the evolving properties and condition of the wash water slurry itself. On the other hand, the treatment of the slurry with CO_2 not only mineralizes CO_2 permanently into waste cement, but it stabilizes the wash water such that its performance is consistent and predictable. Thermal analysis data characterizing % bound CO₂ and % bound water showed that for an untreated slurry the total mass loss (LOI) was increased from 1.2% in the analydrous case to 3.9% for the slurry hydrated 3 hours to 22.6% for the slurry hydrated 26 hours. Conversely, a test of slurry solids treated to an uptake of 27.4% by mass CO₂ and 8.9%by mass bound water (an LOI of 36.3%) at the conclusion of the treatment was observed to be 27.9% CO_2 and 7.7% H₂O (LOI of 35.6%) after holding for an additional 24 hours.

A successful adoption of the approach could realize several operational benefits. The operating expense for solids disposal could be greatly reduced or eliminated. Both costs related to neutralization of wash water prior to discharging and permitting costs associated with discharging could be avoided. The process would come with costs associated with the merchant carbon dioxide and increased admixture usage but these costs could be offset by savings of any admixtures used in an incumbent wash water management approach and through cement savings. The future may bring monetizable carbon offsets for CO_2 utilization that would improve the economic motivation to implement the approach.

5. CONCLUSIONS

The use of carbon dioxide to treat concrete wash water allowed the treated slurry to be used as mix water in the creation of performance enhanced concrete. A high specific gravity wash water slurry was shown to bind CO_2 at around 40% by weight of cement. The complete replacement of potable water with treated slurry lead to concrete with a reduced workability, but modification of the admixture loading addressed the issue. A mix design adjustment to reduce the binder loading in direct proportion to the suspended solids contained within the slurry did not compromise the compressive strength performance. The wash water solids contributed to the performance of the binder and thereby displayed a latent cementitious property.

The approach aligns with industry sustainability goals to reduce the clinker content in concrete mixes, reduced wastes generated by concrete plants, to increase recycled materials usage and to reduce the water impacts of concrete production. An environmental benefit is realized through both mineralized CO_2 and avoided CO_2 from replaced cement and amounted to a 14% reduction of the overall binder carbon impacts. The proposed approach represents a significant operational, resource efficiency, cost saving, regulatory, and logistical opportunity for sustainable concrete production.

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References

- IEA and WBCSD. Technology Roadmap: Low-Carbon Transition in the Cement Industry, Paris and Geneva: OECD/IEA and WBCSD, 2018.
- [2] A. Favier, K. Scrivener, G. Habert. Decarbonizing the cement and concrete sector: integration of the full value chain to reach net zero emissions in Europe. *IOP Conference Series: Earth and Environmental Science* **225**, 2019.
- https://doi.org/10.1088/1755-1315/225/1/012009.

- [3] C. Hepburn, E. Adlen, J. Beddington, et al. The technological and economic prospects for CO2 utilization and removal. *Nature* 575(7781):87-97, 2019. https://doi.org/10.1038/s41586-019-1681-6.
- [4] W. Ashraf. Carbonation of cement-based materials: Challenges and opportunities. Construction and Building Materials 120:558-70, 2016. https: //doi.org/10.1016/j.conbuildmat.2016.05.080.
- [5] D. Zhang, Z. Ghouleh, Y. Shao. Review on carbonation curing of cement-based materials. *Journal* of CO2 Utilization 21:119-31, 2017. https://doi.org/10.1016/j.jcou.2017.07.003.

[6] CO2 SCIENCES THE GLOBAL CO2 INITIATIVE. Global Roadmap for Implementing CO₂ Utilization. Ann Arbor, MI: University of Michigan. Available online at: https://deepblue.lib.umich.edu/bitstream/handl

e/2027.42/150624/CO2U_Roadmap_FINAL_2016_12_07

- [7] C. Lobo, G. Mullings. Recycled water in ready mixed concrete operations. *Concrete InFOCUS* 2(1):17-26, 2003.
- [8] ASTM C1602M-12. Standard Specification for Mixing Water Used in the Production of Hydraulic Cement Concrete, 2012.
- [9] RMC Research and Education Foundation.
 Sustainable Concrete Plant Guidelines Version 1.1, 2011. https://rmc-foundation.org/wp-content/upl oads/2017/07/SCP-Guidelines-Version-1.1.pdf.
- [10] R. L. Berger, J. F. Young, K. Leung. Acceleration of Hydration of Calcium Silicates by Carbon Dioxide Treatment. *Nature Physical Science* 240(97):16-8, 1972. https://doi.org/10.1038/physci240016a0.
- [11] C. J. Goodbrake, J. F. Young, R. L. Berger. Reaction of Beta-Dicalcium Silicate and Tricalcium Silicate with Carbon Dioxide and Water Vapor. *Journal of the American Ceramic Society* 62(3-4):168-71, 1979. https: //doi.org/10.1111/j.1151-2916.1979.tb19046.x.
- [12] D. R. Moorehead. Cementation by the carbonation of hydrated lime. Cement and Concrete Research 16(5):700-8, 1986.
 https://doi.org/10.1016/0008-8846(86)90044-x.
- [13] A. Gabrisová, J. Havlica, S. Sahu. Stability of calcium sulphoaluminate hydrates in water solutions with various pH values. *Cement and Concrete Research* 21(6):1023-7, 1991.

https://doi.org/10.1016/0008-8846(91)90062-m.

- [14] CEMBUREAU 2015 Environmental Product Declaration - Portland Cement (CEM I) produced in Europe.
- [15] B. L. Damineli, F. M. Kemeid, P. S. Aguiar, et al. Measuring the eco-efficiency of cement use. *Cement and Concrete Composites* 32(8):555-62, 2010. https: //doi.org/10.1016/j.cemconcomp.2010.07.009.

//doi.org/10.1016/j.cemconres.2017.08.026.