Literature review: The impact of limestone and plastic additives on concrete

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Abstract

Even though concrete is one of the most universal construction materials, projections indicate that 5-9% of global greenhouse gas emissions result from the cement industry. This research summarizes available information regarding concrete mix designs that incorporate additives, repurpose waste plastic, and reduce carbon emissions. Some of the explored additives include limestone, a carbonate sedimentary rock, and plastic waste, a globally abundant product. The resulting concrete mixture will be "greener" since the lower cement required will lower carbon dioxide emissions. Both the inclusion of limestone and plastic attempt to create a more sustainable mixture, with limestone promoting hydration and plastic promoting recycling. The summarized literature compares the compressive and tensile strength of mixes with varying proportions of limestone, plastic fibers and cement. The inclusion of plastic fibers acts as a secondary layer of reinforcement and helps to reduce plastic shrinkage and settlement. Additional properties of limestone are examined such as slump, shrinkage, hydration, and packing density. The carbonization process is also examined with the goal of understanding how different methods evaluate carbon dioxide emissions. Literature review suggests limestone in concrete mixtures increases healing and physical strength, while decreasing alkali-silica reactions and drying shrinkage.

Keywords: Concrete; Sustainability; Plastic; Plastic Fibers; Limestone

1. Concrete in Construction

With the hopes of making the future a better and more sustainable place, the United Nations created 17 Sustainable Development Goals (SDGs) that are a call to action for all countries to achieve by 2030. These goals tackle relevant initiatives such as improving industry and infrastructure, encouraging responsible material consumption and production, decreasing carbon dioxide emissions and addressing climate change. Despite carbon dioxide emission intensity from cement production increasing by 0.3% annually from 2014 to 2017, a 0.7% annual decrease is necessary to achieve the aforementioned SDGs by 2030 [1]. In 2018, cement was the third largest contributor to annual global fossil emissions at 4% or 1.6 gigatonnes (Gt) of [2]. This critical time period calls even more attention to the existing statistics for the construction industry. It is anticipated that global construction will continue to increase, with a suggested 230 billion square meters of new floor area added by 2060 [3]. Substituting alternatives to Portland cement (PC) serves as a simple low-carbon solution, while options for meeting sustainability goals can also include regenerative design.

2. Material Replacements: Plastic & Limestone

To combat the emissions side effect of the concrete carbonation process, material replacements are used. In some concrete mixtures, supplementary cementitious materials (SCMs) are used in place of Portland cement. SCMs are either industrial byproducts or naturally occurring materials that mimic cementitious behavior when they are hydrated [4, 5]. Examples of SCMs are fly ash, silica fume, ground-granulated blast furnace slag (GGBFS), and limestone (LS) [5]. By using fly ash, the cement paste density increases which allows the water-to-cement (w/c) ratio to decrease. Silica fume is used as a PC replacement, yet it requires a higher w/c ratio and as a result, additional water reducing agents [5]. Slag cement works with PC to increase strength and reduce permeability [6]. The incorporation of coal ash as a replacement to PC can reduce at least 5.89 million tonnes of carbon dioxide emissions by 2035 [4].

2.1. Plastic

The 2018 Chinese import ban is suggested to displace 111 million metric tons of plastic waste by 2030, with 89% of exports containing polymer groups from single use food packaging sources [7]. In the United States, it is estimated that 35.4 million tons of plastics were generated in 2017, representing approximately 13.2% of total municipal solid waste (MSW) generation [8]. Only 3 million tons of plastic, or 8.4% of MSW, was recycled and 5.6 million tons of plastic, or 16.4% of MSW, was combusted for energy recovery, with landfills receiving 26.8 million tons of plastic, or 19.2% of MSW [8].

PlasticsEurope Market Research Group estimates that plastic production reached a global height of 360 million tonnes in 2018, with packaging compromising the largest production sector of 39.9%. Polypropylene, a resin that is used in the production of food packaging and microwave containers, accounts for 19.3% of plastics demand, while low density polyethylene (LDPE), a material that makes up reusable bags and packaging film, results in a 17.5% of plastics demand. Even though recycled plastic waste has doubled since 2006, 25% is still being sent to landfill sites. Switzerland, Austria, and the Netherlands send 100% of their plastic post-consumer waste to either recycling or energy recovery locations [9].

In recognition of the growing quantity of waste, the inclusion of plastic into different construction materials has been a growing interest. Because the incorporation of plastic fibers within concrete will improve mechanical performance, reinforcing waste plastic has been a rising topic of research since 2009 [10].

2.2. Limestone

Limestone is a naturally occurring rock that is found in abundance throughout the world. Thousands of years ago, the Romans used LS powder in their concrete structures which are still standing today. LS is one of the most available materials, accounting for approximately 5% of the Earth's crust [11]. When PC is mixed with this natural material, the carbon footprint of the PC is reduced [12]. Although Portland-limestone cement (PLC) has been heavily researched, the following section aims to examine its properties. By varying amounts of LS, an optimal ratio that improves sustainability without compromising strength can be determined in PLC.

When used in appropriate quantities, LS improves workability, reduces carbon footprint and does not compromise strength [13, 14, 15]. It is an accepted admixture used in concrete mixtures throughout the world, where it has even become part of European, Canadian, and British Standards [16]. European standards allow for a range between 6-20% LS addition, Canadian standards allow for up to 5% addition, and British standards allow no more than 20% addition [16]. These standards have increased the grounds for more sustainable concrete mixes and aid in the reduction of carbon dioxide emissions.

Due to its natural widespread availability, LS does not need to be transported long distances to be used. This reduction in greenhouse gas (GHG) emissions from transportation devices is another sustainable advantage that LS has over other SCMs. When used in concrete mixtures, LS reduces the environmental impact from the PC hydration process, the resulting emissions of , and has versatility when used for historical restoration purposes. The increased workability and lack of compromised strength are also benefits that increase its preferability across the industry [5, 17, 18].

3. Impact Due to Supplementary Cementitious Materials

3.1. Plastic

Similar to how the addition of SMCs in PC-based concrete has an effect on its strength, plastic fibers also have an impact on compressive and split tensile strength. Plastic has been shown to be a viable replacement material for sand in concrete mixtures. It is estimated that a 10% replacement of sand with plastic fibers has the potential to save 820 million tons of sand annually, which accounts for approximately 5% of the global use [19]. A large amount of research being has utilized standard compressive and split tensile testing.

3.1.1. Compressive Strength

A reduction in sand has an impact on the compressive strength of concrete. Research from the University of Salento in Italy concluded that the addition of 5% plastic by weight replacement of sand to a concrete mixture slightly reduced compressive strength, which is attributed to the decreased adhesive strength between the PC and the plastic compared to the sand [10, 20, 21, 22]. Similar research replaced sand with 10%, 15%, and 20% plastic fibers. This research found that compared to the control, the varying plastic addition mixes performed worse under compression testing, with the compressive strength decreasing approximately 5 MPa after 10 days of curing [22]. The decrease in compressive strength increases based on the increase of plastic percentage and curing age [22]. While strength decreases, it has been shown that the addition of plastic fibers reduces the severity of a concrete compressive failure and slightly increases the ductility of the concrete [21, 23]. When plastic was subjected to gamma irradiation, results found that it could partly retrieve some of the strength lost with substituting PC, leading to samples with improved compressive strength [24].

3.1.2. Split Tensile & Flexural

The split tensile test is used to measure tensile strength, which is relevant since concrete is weak in tension. Failure results show the weakest point and is indicative of the overall quality of a mix. The majority of research that has been considered concludes that plastic, which replaces sand in any form or quantity, results in little to no change in tensile strength when amounts less than 5% by volume of sand are replaced, yet the tensile strength drops when higher percent volumes are added [21, 22, 25, 26, 27].

The split tensile strength decreased at higher volumes. The severity of the concrete's failure decreases with the addition of plastic fibers, compared to a non-fiber reinforced concrete cylinder, since plastic fibers can absorb post-failure energy [20, 23]. Similar to compressive strength, the split tensile failures have been linked to the decreased adhesion between the PC and the plastic fibers, compared to the PC and the sand [10, 20, 21, 22]. While sand replacement has been frequently analysed, plastic replacement does not stop with sand. Research has been conducted where the PC binder is being replaced with 0%-0.6% plastic fibers. This experiment concluded that the flexural strength of the concrete increased by 16.5% with a 0.6% optimal PC replacement compared to the control sample [28].

Due to the compilation of physical strength research that has been reviewed, it can be concluded that the addition of plastic fibers as a sand replacement may be better suited for concrete that is not subject to heavy loads [19, 25, 29]. However, the inclusion of waste plastics in concrete could have potential benefits in precast concrete applications where early strength is necessary [27, 29, 30].

3.2. Limestone

SCMs are commonly used to improve the workability and environmental impacts of the carbonization process of PC, as well as impacting strength. The effectiveness of LS as an SCM has been heavily researched as a PC replacement material due to its availability and effectiveness. LS has an effect on slump (ASTM C143), shrinkage, hydration, packing density, compressive strength and split-tensile strength of concrete mixtures. Although PLC has been heavily researched, the following section aims to compile information regarding its properties. An optimal ratio of PC to LS can be determined to improve sustainability without compromising strength or workability.

3.2.1. Compressive Strength

Research from the Structural Engineering Department of the Ain Shams University in Cairo, Egypt concluded that PC with LS substitution decreased the compressive strength of concrete [14]. However, their research also concluded that this decrease is negligible until the replacement reached 10% LS [14]. This finding is supported by research done by *El-Moussaoui et al.* [31] which states that, with a consistent w/c ratio, the compressive strength of the concrete decreased with the increase of LS content [13, 31, 32]. The reason for the reduced strength beyond the optimal 5-10% replacement is due to the fine LS particles filling the voids created from the PC and aggregate, which compacts the mixture. However, once the voids are filled, the LS begins to take the place of the stronger PC as an aggregate. When this happens, the stiffness and integrity of the concrete begins to be compromised [13, 15, 33].

3.2.2. Split Tensile

Split tensile strength is an important parameter when discussing the strength of concrete. Concrete failures, even under compression, are due to tensile failure. The replacement of various amounts of PC will have an impact on the tensile strength of the mix as seen in the compressive strength [14, 32, 34]. Research conducted by Alexandria University in Egypt [14] found that the addition of LS powder influences both compressive and tensile strength at 28 days. While compressive strength was reduced with the inclusion of LS, split tensile strength was reduced of a greater magnitude, with a steep decline in testing results of approximately 0.30 MPa observed with mixtures containing 5% - 10% LS content compared to the control [14]. See Figure 1 for further detail. This notion is supported by research which describes the split tensile strength of PLC. Once the 5%-10% LS replacement of PC is achieved, further addition of LS powder decreases flexural strength [34].

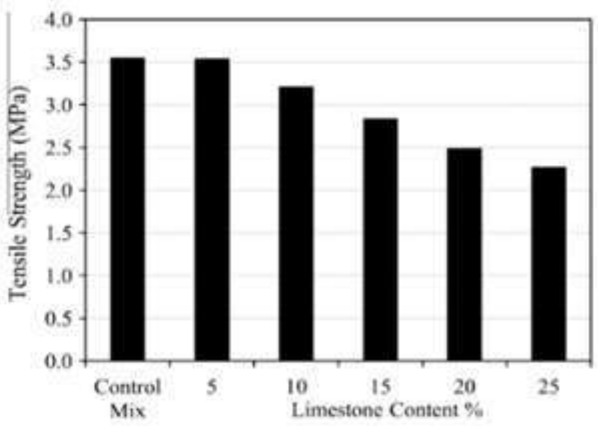

Figure 1. Limestone cement concrete splitting tensile strength at 28 days [14]

3.2.3. Slump

The workability of concrete is an essential part of its effectiveness as a construction material. Slump testing following the ASTM C143 procedure is used as a preemptive means of checking that the correct w/c ratio has been implemented in the concrete mix. The w/c ratio and the amount of aggregate used can have a profound impact on the workability of the concrete mixture. PC alone has low workability compared to PC that is supplemented with various amounts of SCMs [35].

LS supplementation has been shown to improve workability in many cases. When working with an optimized concrete that contains 5% LS, the workability can increase compared to the same cement mixture without the LS addition [15, 35]. LS can accomplish this when it is finely ground by filling the gaps between the clinker particles, reducing the water demand and densifying the structure of the hardened cement paste [13, 35, 36]. In a study from the Laboratoire de Recherche de Génie Civil (LRGC) [15], slump tests were compared between a control concrete sample and LS concrete sample. When the w/c ratio is held constant and the aggregate content is increased, the workability is slightly decreased. However, when a portion of the cement is replaced with LS powder and both the w/c ratio remains constant and the aggregate content is increased, the resulting workability increases [15].

3.2.4. Shrinkage

Shrinkage in concrete is a result of the loss of capillary water during the curing process. Shrinkage can lead to cracking, external deflection, and an increase in tensile stress and strain, before the concrete is subjected to any external load [37]. This greatly weakens the concrete and shortens the lifespan of the structure. Cement content, water content, aggregate type, aggregate content, chemical composition, temperature, and humidity all play a role in shrinkage.

Dimensional variations, such as shrinkage, were evaluated by the group from LRGC [15]. In their research, they studied the known relationship between increased PC amounts and increased shrinkage, to that of increased PLC and shrinkage. Shrinkage measures were carried out on 7x7x28 cm prismatic samples. Shrinkage in micrometers was measured using a digital dial. Results showed that shrinkage decreased for samples with low contents of filler (limestone). While 15% of filler content will return close to the control samples shrinkage measurements, any filler content above 5% showed increased shrinkage [15]. Multiple research findings indicate that any percentage replacement higher than 15% will increase shrinkage past the control value [15, 18, 37]. Based on the size of the particle filler, it is possible that a higher quantity of filler will be required, resulting in a more optimal mixture [38].

3.2.5. Hydration

The hardening of a concrete mixture is known as the hydration process. Driven by water and the chemical reaction with the PC binder, hydration is the process of these two bonding together with the inert aggregate to form a hardened concrete. This is an exothermic reaction and relates to shrinkage if the reaction becomes too hot, for example in high strength concrete on a hot day. Hydration can occur at varying rates based on the chemical reaction between the binder, water, and most importantly, the w/c ratio.

It is well known that LS as calcium carbonate in various amounts has an effect on the hydration process of PC mixtures. It has been shown that the addition of LS in any amount will variably decrease the hydration time for a concrete mixture. Compared to a control sample, a concrete mixture with LS in a 1:1 ratio against PC had a higher heat development over the span of approximately 900 minutes [18]. This extra heat causes the hydration process to speed up and shortens the amount of time available to work with the material. Increasing the rate of hydration also leads to the potential for increased cracking. The accelerated hydration process also reduces the workability of the product. It is shown that calcium carbonate has an accelerating effect on calcium trisilicate and cement hydration, and leads to the precipitation of some calcium carbon silicate hydrate [37, 39, 40].

3.2.6. Packing Density

LS is softer than PC, which allows the fine LS particles to grind preferentially and concrete samples to be made with an improved particle size distribution. The LS itself requires less processing energy to produce. These filler particles fill the gaps created by the PC and aggregate which compacts the mix [15, 38].

4. Sustainability & Carbon Emissions

It is important to consider a variety of different data with the vast quantity and quality of resources available for measuring carbon emissions. Different initiatives have been launched, such as the “Getting the Numbers Right " program by the World Business Council for Sustainable Development (WBCSD), a global organization which combines CEOs from all over to promote sustainability. WBCSD collects relevant cement data from different parts of the world. However, some organizations that use this or the UN Framework Convention of Climate Change (UNFCC), an agreement that attempts to stabilize GHGs, have seen outdated numbers. The Global Carbon Project aims to annually publish and maintain carbon models for estimating accurate emission databases [41].

The Global Carbon Project, an organization established in an attempt to quantify global greenhouse gas emissions with accurate data, published startling results in 2019. The report indicates that global fossil carbon dioxide emissions have steadily increased by almost 27 Gt throughout the last five decades, with little promise of slowing down soon [2]. Projections in 2018 indicate that the United States has the highest annual fossil carbon dioxide emissions per capita of 16.6 tonnes/person. Overall, China annually contributes the highest emission totals with 10.06 Gt or 27.5% while the United States is the second highest contributor at 5.42 Gt or 14.8% [2].

Talaei [42], who performed a case study in Canada, uses a bottom up energy model and scenario analysis to assess greenhouse gas emissions from the cement industry. Through the Long-range Energy Alternative Planning model, 20 different scenarios were created with the goal of reducing greenhouse gas emissions with analyzed factors such as cost of energy saved, GHG reduction potential and carbon abatement. Results indicate that 70% of emissions can be reduced without negative cost to the industry through strategies such as implementing energy management systems, fuel switching, and indirect firing for clinkers, resulting in mitigated carbon dioxide emissions of 27 million tonnes by 2030 and 59 million tonnes by 2050 [42].

In other studies, the Green Concrete Life-Cycle Assessment Tool is used to understand unit processes involved with the creation of concrete [43]. This tool uses factors of energy consumption related to material production such as lead and carbon dioxide emissions, electricity and fuel, and material usage. The user can input different characteristics specific to the concrete mix design including anticipated volume quantities, materials used, and method of aggregate transportation. Results show that global warming potential and carbon dioxide emissions are reduced when creating cement mixes that reduce Portland cement content, and that using dichotomous earth in areas where the product is abundant (Western US, China, Turkey) serves as a mix design advantage [43].

Research published by *Flower* [44], used an Australian Greenhouse Office Factors and Methods workbook to calculate carbon dioxide emissions from multiple energy sources throughout Melbourne, Australia. Data collected from a life cycle analysis reviews two coarse aggregate quarries, one fine aggregate quarry, six concrete batching plants, and other sources. Results indicate that Portland cement is the main source of carbon dioxide emissions within a concrete mixture, responsible for 74% to 81% of total emissions [44].

5. Carbonation

Carbonation occurs within concrete because of the reaction of calcium hydroxide with the carbon dioxide of the cement paste. This reaction, which creates calcium carbonate, is often concerning because of its pH, which is known for corroding steel reinforcement bar, and its harmful greenhouse gas emissions [45]. As a result, improving the resistance of concrete carbonation is a widely researched topic.

Some methods used to evaluate carbonation profiles include Thermogravimetric Analysis (TGA), chemical analysis, gammadensimetry, pH change, Energy Dispersive X-Ray Fluorescence Spectrometer (EDX), Fourier-Transform Infrared Spectroscopy (FTIR), and Laser Diffraction Particle Size Analyzer [45, 46]. TGA finds that using the temperature range of 530-950 ˚C is the best way to measure decomposition due to carbonation, in conjunction with a chemical analysis correcting factor that accounts for tracer cement materials [45, 46].

When experimenting with concrete mix designs, a variety of different additives and SCMs are explored. Research in Brazil [47], that created concrete samples of varying strengths to be crushed and used as recycled concrete aggregates (RCA) in new samples of 32.5 MPa concrete, used compression and carbonation testing to understand the durability of concrete with recycled aggregates. Results indicate that after accelerated carbonation, which took place in conditions of 1% and 70% relative humidity (RH) across 147 days, it was found that concrete with RCA from 18 MPa and higher porosity resulted in a carbonation depth equal to 50% of reference mixes, while concrete with RCA from 37 and 50 MPa and equal or higher porosities resulted in similar carbonation depth to reference mixes [47].

Other anticipated influences on carbonation results include aggregate replacement content and type, material size and microcracking. Using aggregate replacement from RCA is one option that displayed carbonation depth results similar to the reference concrete samples [47].

Limestone powder has a fineness which creates nuclei locations for calcium carbonate precipitation, and it is of comparative size with cement particles. Thus, PLC mixtures result in high carbon reactivity and early strength gain [46]. The use of finer materials promotes higher carbonation reactivity and early strength, while the occurrence of microcracking along the carbonation front causes cement degradation, decreased mechanical strength and increased permeability [46, 47, 48].

When considering the effect of the addition of SCMs on carbonation, such as silica fume and low/high calcium fly ash, results indicate that carbonation depth decreases when SCMs replace aggregates and carbonation depth increases when SCMs replace cement [49]. Other research considers the effects on carbonation due to the inclusion of limestone in concrete. One study found that despite the inclusion of 40% calcium carbonate, 15% limestone additives, and 25% carbonation induced calcium carbonate, results indicate that PLC can be used to effectively replace Portland cement, especially for precast concrete products where early strength is necessary [46]. Portland limestone concrete mixes that are designed on the basis of equal strength, rather than equal w/c ratio and exposed to continuous moist curing, show comparable carbonation resistance with control mixes. However, the inclusion of SCMs such as fly ash, limestone, and slag tend to create concrete mixes that carbonate at higher rates than control groups [16]. When limestone was used as an embedding agent and subjected to carbonation from brine, no evidence of degradation or permeability was present in the concrete [50].

However, research conducted in Italy [51], that aims to explore the w/c ratio, cement content, curing time, and other performances of the effect of ground limestone (15/30% replacement to PC) in concrete, show opposing results indicating that mechanical properties (compressive strength) and resistance to penetration are reduced with the increase of limestone [51].

With the growing interest of plastics, this material has also been evaluated for its effect on carbonation. Concrete, that was made with 10% replacement of sand by volume with graded polyethylene terephthalate (PET) plastic, was tested for carbonation at 4% and 55-65% RH [52]. Results indicate that while plastic concrete mix had a higher initial carbonation rate at 14 and 28 days, the results from 180-

day testing indicate that an acceptable carbonation depth was only 0.5 mm deeper, or 5% higher than the reference concrete [52].

6. Conclusion

Based on the compiled research, it can be concluded that introducing certain amounts of supplementary cementitious materials into Portland-limestone cement mixes can have countless benefits. Limestone and recycled plastic fibers have been shown to improve the sustainability of concrete, without comprising strength or workability, by reducing the carbon dioxide emissions and providing a use for recycled plastic. It is anticipated that a small percentage (0.5% by weight) of plastic is desired, as large percentages can create oxygen that hinder binding abilities. The inclusion of waste plastics in concrete could have potential benefits in precast concrete applications where early strength is necessary [27, 29, 30]. Meanwhile, a percentage (5-10% by weight) of limestone is desired, as it fills voids that hinder compacting abilities. Results indicate that Portland cement is the main source of carbon dioxide within a concrete mixture, responsible for 74% to 81% of total emissions [44]. When combined in the aforementioned proportions, Portland-limestone cement mixtures will have comparable qualities to a control sample. These proposed proportions fill the voids between research that solely tested limestone or solely tested plastic fibers and verifies results from combined mixtures.

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