# ASSESSMENT OF THE MECHANICAL AND ENVIRONMENTAL PROPERTIES OF CONCRETES WITH HIGH LEVELS OF LIME FILLER AND FLY ASH

## *AVALIAÇÃO DAS PROPRIEDADES MECÂNICAS E AMBIENTAIS DE CONCRETOS COM ALTOS TEORES DE FILER CALCÁRIO E CINZA VOLANTE*

## *EVALUACIÓN DE LAS PROPIEDADES MECÁNICAS Y AMBIENTALES DE HORMIGONES CON ALTO CONTENIDO DE POLVO DE CALIZO Y CENIZAS VOLANTES*

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### ABSTRACT

This study evaluated the possibility of producing high strength concrete (HSC) by replacing Portland cement (PC) with high levels of limestone filler (LF) in binary mixtures and ternary mixtures of PC, LF and fly ash (FA) in contents varying from 50 % to 80 %, with the objective of reducing CO2 emissions during manufacture. Parameters evaluated were axial compression strength, CO2 emissions of the constituent materials and cost per $m^3$ of concrete with focus on reductions in environmental impact. Results indicated the possibility of producing HSC with high LF and FA contents, reduced water/binder content (w/b = 0.25) and workability of 100 mm  20 mm through particle packaging and the use of a superplasticizer additive. The resulting concrete had axial compression strength of 51.8 MPa at 91 days with 77 kgco2/m3 of concrete for a mixture with 80 % of PC replaced with 70 % LF and 10 % FA. These amounts corresponded to the use of only 97 kg/m3 of PC (87 kg/m3 of clinker) and 104 L/m3 of water. Thus, it was demonstrated to be possible to obtain a HSC with fck of up to 80 MPa and low CO2 emissions through the use of high levels of mineral admixtures.

### KEYWORDS

Sustainability; High strength concrete; Limestone filler; Fly ash; high strength concrete.

### *RESUMO*

*O presente artigo avalia a possibilidade de produção de Concreto de Alta Resistência (CAR) com a substituição de cimento Portland (CP) por teores elevados de fíler calcário (FC), em misturas binárias e ternária com cinza volante (CV), em proporções de 50% a 80%, com o propósito de diminuir as emanações de CO2 durante sua fabricação. Foram estudadas a resistência à compressão axial, a emissão de CO2 dos materiais constituintes e o custo por m3 de concreto, com vistas à redução do impacto ambiental. Foi possível a elaboração de CAR com elevados teores de FC e CV e reduzidos teores água/ligante (a/l = 0,25), através do empacotamento de partículas e da utilização de aditivo superplastificante, com uma trabalhabilidade de 10020 mm. Obteve-se concretos com resistência à compressão axial de 51,8 MPa, a 91 dias, com 77 kgCO2/m3 de concreto, onde 80% do CP foi substituído por 70% de FC e 10% de CV,*

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*com o uso de apenas 97 kg/m3 de CP (87 kg.m-3 de clínquer) e 104 L/m3 de água. Este trabalho mostra que é possível obter-se CAR com fck de até 80 MPa e baixas emissões de CO2, com o emprego de elevados teores de adições minerais.*

## *PALAVRAS-CHAVE*

*Sustentabilidade; Fíler calcário; Cinza volante; Concreto de alta resistência.*

## *RESUMEN*

*Este trabajo evalúa la posibilidad de producir concreto de alta resistencia (CAR) reemplazando el cemento portland (CP) por altos niveles de polvo calizo (PC), en mezclas binarias y ternarias con cenizas volantes (CV), en proporciones del 50% al 80%, con el objetivo de reducir las emanaciones de CO2 durante la fabricación. Se estudió la resistencia a la compresión axial, la emisión de CO2 de los materiales que lo componen y el coste por m3 de hormigón, con el fin de reducir el impacto ambiental. se logró producir car con altos contenidos de PC y CV y contenidos reducidos de agua/conglomerante (a/l = 0,25), mediante empaquetamiento de partículas y el uso de un aditivo superplastificante, con una trabajabilidad de 100  20 mm. con resistencia a compresión axial de 51.8 MPa, a 91 días, con 77 kgco2/m3 de concreto, donde se reemplazó el 80% de CP por 70% de PC y 10% de CV, con el uso de solo 97 kg/m3 de CP (87 kg/m3 de clinker) y 104 L/m3 de agua. el trabajo demuestra que es posible obtener CAR con fck de hasta 80 MPa y bajas emisiones de CO2, utilizando altos niveles de adiciones minerales.*

## *PALABRAS CLAVE*

*Sostenibilidad; polvo de piedra caliza; ceniza volante; Concreto de alta resistencia.*

## 1. INTRODUCTION

Brazilian standard NBR 6118 (ABNT, 2023) classifies concrete strength in 2 categories: class I contains concretes with strengths between 20 MPa and 50 MPa and class II had strengths between 50 MPa and 90 MPa, the latter being considered high strength concretes (HSC). High strength concretes are achieved with reduced porosity through: (a) lower water/binder ratios (w/b); (b) decreased amounts of water used by cubic meter of concrete with plasticizer and superplasticizer additives and (c) optimized particle packing with the selection of coarse aggregates with lower characteristic dimensions and adequate fine aggregate granulometries though mineral admixture (MA). From a sustainability point of view, HSC has advantages related to longer durability, decreased use of raw materials and lower Portland cement (PC) consumption per MPa of compressive strength (De Matos et al., 2019; Yousuf et al., 2019).

An alternative to obtain sustainable HSCs consists of the replacement of PC with MA such as limestone filler (LF) and fly ash (FA). Portland cement clinker, while the most important component of concrete, contributes the most to the emission of greenhouse gasses (GHG), in particular CO2. This is a consequence of approximately 2/3 of clinker being composed of calcium carbonates which are decarbonized in ovens during manufacture and, added to fossil fuel combustion of industrial processes, are the primary sources of emissions. Clinker contributes to 85 % to 90 % of concrete CO2 emissions and 5 % of total global emissions (GCCA, 2017). Since PC is the most manufactured material worldwide by mass, it presents a contradictory challenge of supplying social economic development while maintaining desired environmental sustainability.

Amongst inert MAs, limestone filler (LF) is the most used due to its worldwide availability, ability to induce good rheological characteristics to fresh cement, low cost and low GHG emissions. International standards limit LF content in concrete to between 5 % and 35 % depending on region and technological know-how. A common procedure is its addition to clinker during grinding in the production of cement. However, recent studies made use of higher PC to LF substitution ratios and even ternary concrete mixtures with pozzolans such as FA. These increased substitutions sought to obtain a sustainable HSC with lower cost per m3 of concrete while maintaining its durability (PALM et al., 2016). Furthermore, the use of inert LF with an active mineral admixture could produce additional benefits through synergic effects, not only with respect to general concrete properties and sustainability but also with respect to normalized factors such as the amount (kg) of agglomerates needed for each MPa of concrete strength (ISAIA et al., 2003).

Increases in concrete axial compressive strength are commonly obtained from lower water/binder ratios (w/b), superplasticizer admixtures and PC replacement with MA such as pozzolans or FA. Overall, these procedures are employed to lower environmental impacts associated with the use of PC. This application of HSC with further particle packing techniques can produce structures with equivalent strength and performance to one constructed with conventional concrete albeit with a lower volume of concrete consumption (De Matos et al. 2019; Scrivener et al., 2018; Gartner and Hirao, 2015). Mehta and Monteiro (2014) noted the additional sustainability effect of HSC in the decreased consumption of raw materials.

Durability, while not addressed in this study, is known to be associated with the performance HSC with strengths above 50 MPa and w/b ratios of less or equal to 0.30. As porosity decreases, permeability and fluid diffusion decrease drastically due to pore sizes becoming smaller than the critical diameter. Malhotra et al. (2000) reported that mixtures with 150 kg/m3 of PC, 200 kg/m3 of FA, 102 L/m3 of water and w/b ratio of 0.29 presented good durability, especially with respect to factors related to reinforcement corrosion in in situ observations up to 10 years.

Thus, the objective of this study was to test HSC mixtures with high PC substitution content that would lower environmental impact and achieve economic viability and acceptable performance with respect to standard values of axial compressive strength. The concrete mixtures of this study were binary of PC and LF and ternary with PC, LF and FA. Mixtures were prepared with particle packing techniques to minimize void spaces. Portland cement substitution ratios varied between 50 % and 80 %. Parameters evaluated were compressive strength, CO2 emissions and cost per m3 of concrete. Results showed that LF content can be comfortably increased beyond current limits set in standards. While not comprehensive, this study is part of a larger prospective study with the objective of decreasing clinker consumption to below 100 kg/m3.

## 2. LIMESTONE FILLER

Filler is any finely ground material of approximately the same or greater fineness than PC. Fillers imbue beneficial effects in some properties of concrete such as workability, permeability, capillarity, extrusion and fissuring potential. They also have very low reactivity and are considered chemically inert which, depending on hydraulic characteristics or absence of undesirable reactions with cement paste products, is not a disadvantageous property (NEVILLE, 2016).

Limestone fillers are found in nature as: (a) calcite (calcium carbonate) with crystalline trigonal rhombohedral geometry; (b) dolomite (calcium and magnesium carbonate) with predominantly rhombohedral crystal structures and (c) aragonite (calcium carbonate) with metastable orthorhombic crystals. Of these minerals, the first two are most commonly used (SAMPAIO; ALMEIDA, 2008).

The addition of limestone filler is an important technique in reducing CO2 emissions in the manufacturing process of cement. The main effect is a decrease in decarbonation and related decreases in fossil fuel combustion emission and electrical energy consumption during grinding (BATAGGIN, A. F.; SILVA, 2019)

## 2.1 Physio-chemical reactions of limestone filler on concrete

### *2.1.1 Physical effect*

Physical effects occur when the mere presence of the supplemental cementitious material or inert MA interferes with clinker hydration. This is due to surface electrical potential, also known as zeta potential (BERODIER and SCRIVENER, 2015; SCRIVENER et al. 2015),

### *2.1.2 Dilution effect*

Dilution effects occur when particles of a reactive material are replaced with less reactive or inert ones. This can bring negative effects on durability and mechanical performance (IRASSAR, 2009) due to lower PC content and corresponding increase in effective w/b ratio (IRASSAR et al. 2015). As a consequence, less hydrated products are generated and a decrease in compressive strength occurs across all ages.

### *2.1.3 Heterogenous nucleation effects*

The presence of LF alters the speed of reaction of PC, contributing to hydration at initial ages (KADRI and DUVAL, 2002). Heterogeneous nucleation occurs from LF due to the smaller particle size filling void spaces. Consequently, density and nucleation points for hydration products increase in the mixture. Thus, crystal growth occurs not only on the surface of PC grains but also on the surface of LF, with corresponding increases in reactions and types of hydration products being formed. The reduction in void spaces induces the formation of large number of small crystals in lieu of a few large ones (HEMALATHA and SANTHANAM, 2018; MEHDIPOUR et al. 2017).

### *2.1.4 Effect in hydration reactions*

Different from pozzolans and pozzolanic activity, the chemical effect of LF (CaCO3) occurs from its interaction with alumina present in PC, especially C3A. From this interaction, a new hydrated monocarboaluminate phase is formed. This phenomenon was stressed by Battagin (2017) and Bonavetti et al. (2003) as an additional moderate chemical reactivity of LF with PC concurrent to their physical interaction.

### *2.1.5 Granulometry effects*

Grain size and structure are defined from granulometric analysis and BET specific surface area. Their values drive LF reactivity in cementitious pastes or concretes of binary, ternary or quaternary composition. Espining (2008) noted the effects of granulometry as: (a) better physical nature of the packed material, whose higher density and better grain dispersal affected concrete flowability and compressive strength; (b) fineness and specific surface area increased nucleation, with effects on compressive strength depending on the affinity of LF with hydrated PC products; (c) higher BET area LF decreased concrete flowability and increased autogenous shrinking, with cascading effects of decreased evaporation, decreased plastic fissuring and increased compressive strength; (d) an BET area increase in the order of 1,000 $m^2/kg$ required a 0.8 % increase by mass in the amount of water to maintain concrete workability and decreased plastic shrinkage by 20 % and (e) the increase in water content due to BET area increase tended to increase fissuring and decrease mechanical strength.

## 2.2 Effect of high replacement content of limestone filler

Studies over the last 3 decades examined the performance of LF in PC concrete and denoted a rising substitution content trend. However, there were still diverging results with respect to rheological and mechanical behaviors and durability (Lollini et al., 2014). As LF content increased in the paste with disregard to granulometry and water content, the w/b ratio increased, and performance declined. This behavior could be controlled by lowering the w/b ratio, improving workability with superplasticizer admixtures and, critically, controlling particle dispersal to decrease void spaces. This would increase packing between PC and LF grains with the aggregates of the mixture. According to Fennis and Walraven (2012), it was possible to elaborate concretes with low CO2 emissions by replacing 50 % of PC with MA and optimizing mixtures with particle packing techniques.

John et al. (2018) proposed a new perspective on lowering clinker content with an innovative technique. Their methodology combined low clinker and high LF content with dispersants so that water content would be reduced to counter dilution. This technique has been successfully applied in related fields such as pre-manufactured and pre-cast concrete industries.

Similarly, Proske et al. (2013) reported that concrete with 20 % to 35 % LF replacement content with respect to PC by mass and common w/b ratios had a critical level of performance. Consequently, desired concrete durability could only be achieved with reductions in w/b ratio. Proske et al. (2013) further listed the main principles for the development of a low clinker (high LF) content concrete: the use of a high-performance superplasticizer admixture and optimized packing density. These two principles allowed a reduction in water content and simultaneously minimized PC clinker levels.

Palm et al. (2016) applied elevated LF replacement content (above 50 %) with respect to PC and evaluated mechanical properties, durability and sustainability characteristics of the concretes. It was concluded that: (a) concretes with 50 % PC and 50 % LF content by mass and a w/b ratio of 0.35 could have sufficient mechanical properties for practical construction applications if properly monitored; (b) the w/b ratio was the main parameter for high LF content concretes; (c) LF did not appear completely inert as its effect on compressive strength was substantial as clinker was further replaced with LF; (d) all concretes with 50 % PC and 50 % LF content by mass and a w/b ratio of 0.35 had compressive strength equal or higher than the reference mixture with a w/b ratio of 0.50. Thus, in concert with results from other studies over the past 2 decades, Palm et al. (2016) demonstrated that high LF content concretes had viable HSC mechanical properties with respect to compressive strength as well as sustainability.

## 2.3 Ternary mixtures with LF and FA

Ternary mixtures with LF and FA have to account for synergic effects since physical and chemical effects of both MA cannot be separated. This synergic effect was studied by Isaia (1995) with several MA mixtures. It was concluded that the reactivity of a MA was not limited to its amorphousness or crystallinity but also linked to physical and chemical effects of other active or inert additions.

Deschener et al. (2012) and De Weerdt et al. (2011) noted that synergy occurred between the carbonaceous materials of LF with FA. By itself, LF reacted with aluminates of hydrated PC to form carboaluminates. But this effect was enhanced with additional carboaluminates introduced in the mixture from the pozzolanic reaction of FA. De Weerdt et al. (2011) further noted that LF reaction with aluminates and FA contributed to decreasing concrete porosity.

## 3. METHODOLOGY AND MATERIALS

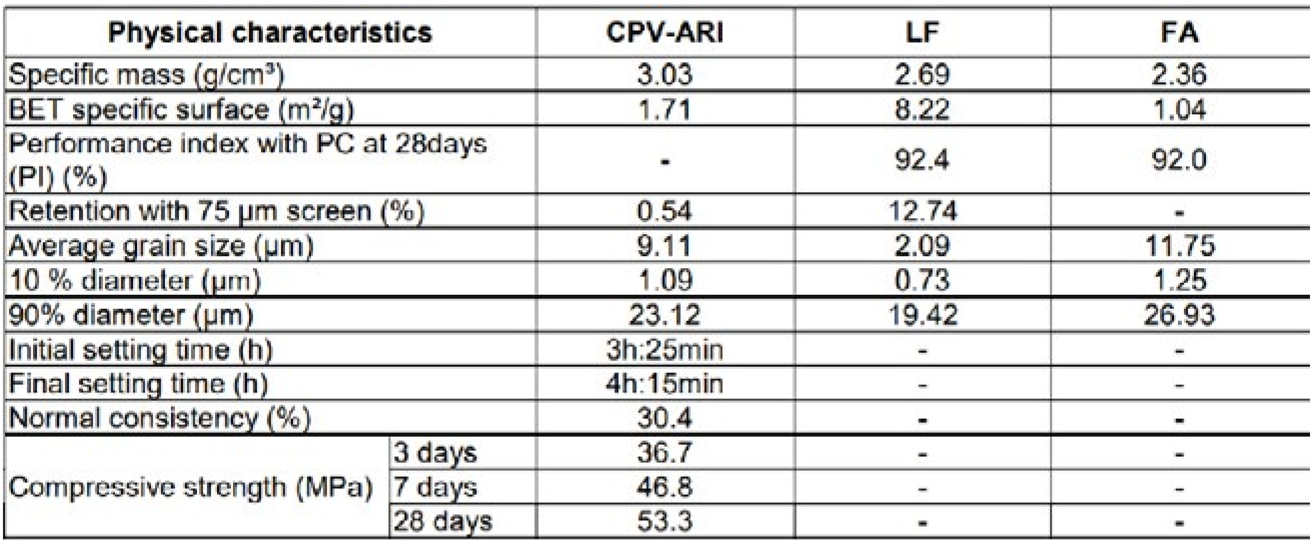
The objective of this study was to evaluate the potential of high strength concretes (HSC) with PC replaced with high contents of limestone filler (LF) and fly ash (FA). Two replacement ratios were tested: binary mixtures with 50 % and 60 % FC content and a ternary mixture with 70 % LF and 10 % FA for a total 80 % replacement of PC. All materials were characterized, and packing techniques evaluated to achieve the best concrete compaction. Results from previous studies were used to elaborate 4 concrete mixtures: (a) reference (REF) mixture with 100 % CP V-ARI cement; (b) 50 % LF and 50 % CP V-ARI cement (50LF); (c) 60 % LF and 40 % CP V-ARI cement (60LF) and (d) 70 % LF, 10 % FA and 20 % CP V-ARI cement (70LF10FA). All concretes were mixed with a w/b ratio of 0.25. After molding and manufacture of test bodies, axial compression strength, CO2 emissions and cost per m3 of concrete were evaluated.

### 3.1 Materials

The PC used in this study was CP V-ARI with high initial strength due to its low original LF content (up to 10 %) as produced nationally. A chemical analysis conducted by Brazilian Association of Portland Cement (ABCP) determined that this type of cement contained 7.2 % calcite, 2.5 % plaster and 0.56 % loss on ignition, which corresponded to an approximate clinker content of 90 %. The LF selected for this study was extracted from veins in Caçapava do Sul (RS) and was of dolomite-calcite rocks abundant in this region. The LF was ground in a ball mill for 180 min to reach adequate granulometry and fineness. Resulting BET surface area was 8.22 m²/g and PC performance index was 92.4 %, which allowed adequate behavior with respect to consistency in the fresh state and compressive strength in the hardened state. The FA was obtained from the thermoelectric power plant in Candiota (RS) and was also ground in a ball mill for 120 min to become an F class pozzolan. Table 1 presents the characteristics of PC, LF and FA. It should be noted that LF had a higher performance index than FA even though the former was theoretically considered inert while the latter was a chemically active pozzolan. This was likely due to the finer BET surface area of LF which allowed more intense and quicker chemical reactions and greater attraction from the zeta potential. The result was the quicker production of carboaluminates filling void spaces in the paste.

Two types of natural sand were used from quarries in Santa Maria (RS) with fine sand having a maximum diameter of1.2 mm and medium sand having a maximum diameter of 2.4 mm. The sand types were selected in order to allow a gradual granulometric transition from the fine PC grains to the MA and coarse aggregate. The coarse aggregate was gravel from crushed diabase rock from the city of Itaara (RS), classified as type 0 gravel with maximum diameter of 12.5 mm.

Superplasticizer admixture was used in amounts appropriate to the achieve desired rheological parameters of the concretes. The superplasticizer was polycarboxylate-based BASF Master Glenium 54 with average solid content of 40 % and density of 1.1 g/cm3. The compatibility of the superplasticizer with the PC was checked with the Marsh cone test and determined to be an excellent 1 %.



| Physical characteristics | | CPV-ARI | LF | FA |
|---|---|---|---|---|
| Specific mass (g/cm³) | | 3.03 | 2.69 | 2.36 |
| BET specific surface (m²/g) | | 1.71 | 8.22 | 1.04 |
| Performance index with PC at 28days (PI) (%) | | - | 92.4 | 92.0 |
| Retention with 75 µm screen (%) | | 0.54 | 12.74 | - |
| Average grain size (µm) | | 9.11 | 2.09 | 11.75 |
| 10 % diameter (µm) | | 1.09 | 0.73 | 1.25 |
| 90% diameter (µm) | | 23.12 | 19.42 | 26.93 |
| Initial setting time (h) | | 3h:25min | - | - |
| Final setting time (h) | | 4h:15min | - | - |
| Normal consistency (%) | | 30.4 | - | - |
| Compressive strength (MPa) | 3 days | 36.7 | - | - |
| | 7 days | 46.8 | - | - |
| | 28 days | 53.3 | - | - |

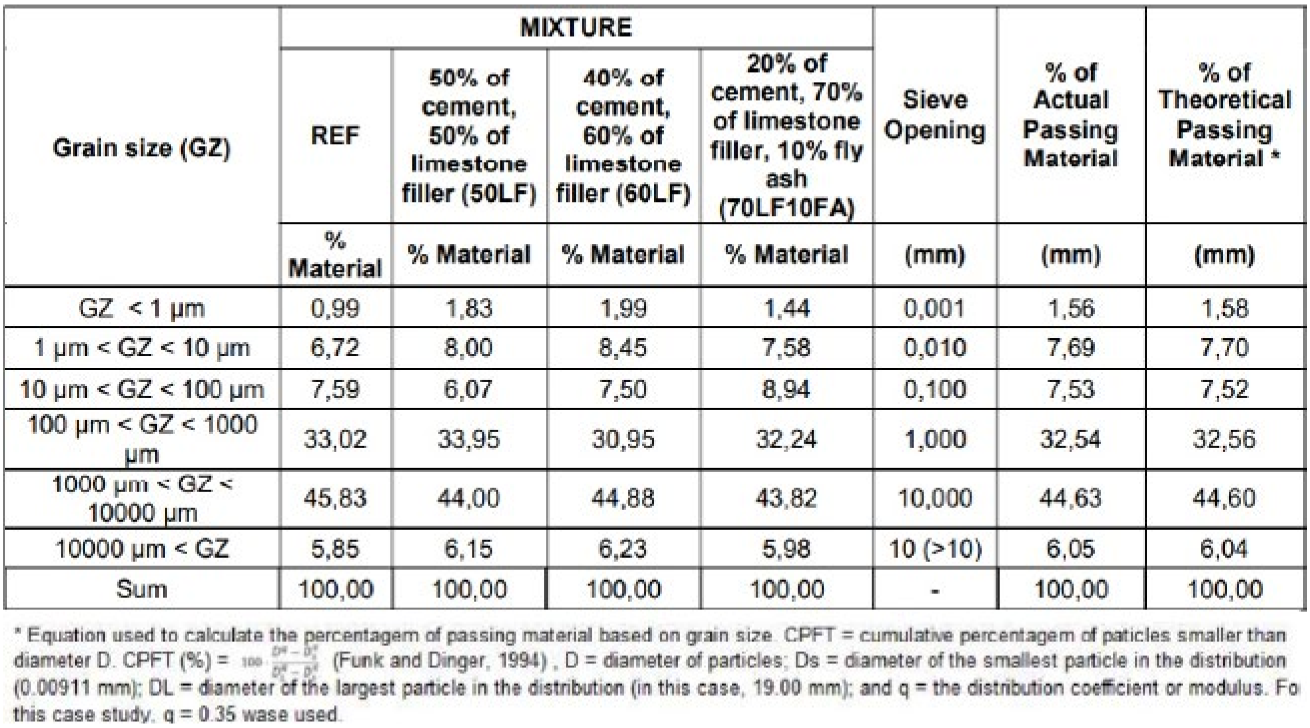
**Table 1:** Agglomerate characteristics
**Source:** the authors

### 3.2 Particle packing

Particle packing between aggregates, PC and MA was evaluated with EMMA (Elkem Materials Mix Analyzer) software. This was conducted in order to determine a granulometric mix ratio based on the real grain size of the materials. For each mix ratio, material quantities, densities and granulometric curves were entered and, by varying the ratios, a mix ratio closest to the ideal mix ratio curve was obtained. The ideal curve followed the recommendations of Funk and Dinger (1992) which made use of the Andreassen mathematical model. This model was selected since it was the most suitable for mixtures with small-sized particles. Furthermore, a value of 0.35 was used for the "q" distribution coefficient as recommended by Funk and Dinger (1992) in order to improve packing as in mixtures with high workability. The particle size distribution of the concrete mixtures is shown in Table 02.

### 3.3 Concrete preparation

Preliminary studies indicated optimum values for the concrete: 53 % for mortar content by mass and 8.33 % for the water/dry agglomerate ratio. These values were constant for all mixtures used in this study. The mixtures were prepared in a high-power mixer (900 W) equipped with high efficiency paddles and metallic drum under high rotations (between 645 rpm and 1,400 rpm). This set up was used comply with high energy requirements in mixing due to the low 0.25 w/b ratio. Table 3 presents the mix ratios of all concretes as kg/m3 of concrete.

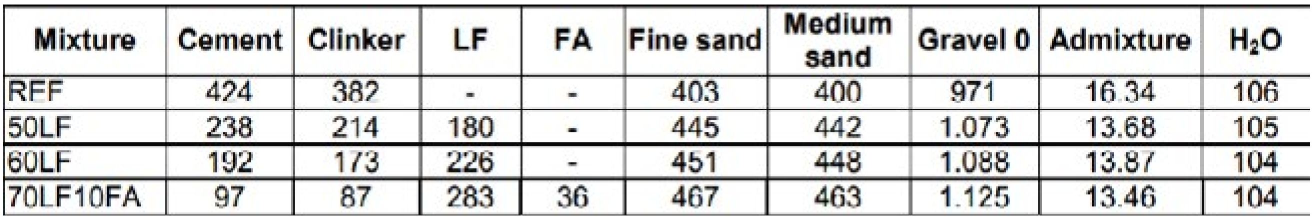


| Grain size (GZ) | MIXTURE | | | | Sieve Opening | % of Actual Passing Material | % of Theoretical Passing Material * |
|---|---|---|---|---|---|---|---|
| | REF | 50% of cement, 50% of limestone filler (50LF) | 40% of cement, 60% of limestone filler (60LF) | 20% of cement, 70% of limestone filler, 10% fly ash (70LF10FA) | | | |
| | % Material | % Material | % Material | % Material | (mm) | (mm) | (mm) |
| GZ < 1 µm | 0,99 | 1,83 | 1,99 | 1,44 | 0,001 | 1,56 | 1,58 |
| 1 µm < GZ < 10 µm | 6,72 | 8,00 | 8,45 | 7,58 | 0,010 | 7,69 | 7,70 |
| 10 µm < GZ < 100 µm | 7,59 | 6,07 | 7,50 | 8,94 | 0,100 | 7,53 | 7,52 |
| 100 µm < GZ < 1000 µm | 33,02 | 33,95 | 30,95 | 32,24 | 1,000 | 32,54 | 32,56 |
| 1000 µm < GZ < 10000 µm | 45,83 | 44,00 | 44,88 | 43,82 | 10,000 | 44,63 | 44,60 |
| 10000 µm < GZ | 5,85 | 6,15 | 6,23 | 5,98 | 10 (>10) | 6,05 | 6,04 |
| Sum | 100,00 | 100,00 | 100,00 | 100,00 | - | 100,00 | 100,00 |

* Equation used to calculate the percentagem of passing material based on grain size. CPFT = cumulative percentagem of paticles smaller than diameter D. CPFT (%) = $100 \cdot \frac{D^q - D_S^q}{D_L^q - D_S^q}$ (Funk and Dinger, 1994) , D = diameter of particles; Ds = diameter of the smallest particle in the distribution (0.00911 mm); DL = diameter of the largest particle in the distribution (in this case, 19.00 mm); and q = the distribution coefficient or modulus. Fo this case study, q = 0.35 wase used.

**Table 2:** Particle size distribution of the concrete mixtures.

**Source:** the authors



| Mixture | Cement | Clinker | LF | FA | Fine sand | Medium sand | Gravel 0 | Admixture | $H_2O$ |
|---|---|---|---|---|---|---|---|---|---|
| REF | 424 | 382 | - | - | 403 | 400 | 971 | 16.34 | 106 |
| 50LF | 238 | 214 | 180 | - | 445 | 442 | 1.073 | 13.68 | 105 |
| 60LF | 192 | 173 | 226 | - | 451 | 448 | 1.088 | 13.87 | 104 |
| 70LF10FA | 97 | 87 | 283 | 36 | 467 | 463 | 1.125 | 13.46 | 104 |

**Table 3:** Amounts of materials per kg/m3 of concrete.

**Source:** the authors

Materials were added to the mixture in the following order: (a) 100 % quantities of PC, MA, water and admixtures; (b) fine and medium sand and (c) gravel. Consistency was measured with a slump test and kept in the range of 100 mm ± 20 mm.

Sustainability was evaluated with relative scores for each parameter examined with the best performance receiving a score of 100. The highest axial compression strength was obtained by REF mixtures across all ages, so it received a score of 100 for this parameter. The lowest CO2 emissions and cost occurred for the 70LF10FA mixture, which then received a score of 100 for those parameters. It should be noted that CO2 emissions and cost were inversely proportional to axial compressive strength. The most sustainable mixture was then selected through a mathematical average of the three relevant parameters of this study between the ages of 28 days and 91 days.

### 3.4 Axial compressive strength

Axial compressive strength tests were conducted in accordance with the procedures of standard NBR 5739 (2018) on cylindrical test bodies 10 cm in diameter and 20 cm in height molded in accordance to standard NBR 5738 (2015). The cylindrical test bodies were cured in water with limestone until the age for the tests, at which moment they were polished and crushed in an Instron model HDX 1500 hydraulic press. Tests were conducted at ages of 28 days and 91 days with 4 test bodies for each mixture. Non-conformant results were discarded, and the remaining values averaged out for each set of test bodies. Once axial compressive strength was determined, the normalized strength with respect to m3 of concrete, clinker intensity and binder intensity were determined for each mixture.

### 3.5 CO2 emissions

Coefficients used to determine CO2 emissions from the materials were based on certified values of Isaia and Gastaldini (2004). The following CO2 emission coefficients (in kg.CO2/ton) used were: a) PC = 617 (GCCA, 2017); b) LF = 26 (Habert et al., 2013); c) FA = 10; d) sand = 3; e) gravel = 4; f) water = 5; g) chemical admixture = 94 (Isaia and Gastaldini, 2004). These coefficients refer to average values for each of the constituent materials.

Multiplying the CO2 emission coefficients by the material consumption per cubic meter of concrete shown in Table 2 allows for calculating the total CO2 emitted by each of the concrete mixtures.

### 3.6 Cost per m3 of concrete

The cost per m3 of concrete of each mixture was calculated based on the amount of materials shown in Table 03. The amounts were linked to the TCPO (Tabela de Composições de Preços para Orçamentos, in portuguese) provided by PINI and prices per kg of material were sourced from the July, 2023 SINAPI (Sistema Nacional de Pesquisa de Custos e Índices da Construção Civil, in portuguese) reference table.

## 4. RESULTS

### 4.1 Axial compressive strength

Table 04 presents results for axial compressive strength, standard deviation and cost per m3 of concrete.

Figure 01 presents the relation of axial compressive strength at 28 days and 91 days with respect to LF substitution ratio. Overall, all concrete mixtures with MA had decreasing axial compressive strength as LF content increased. No mixture with MA substitution matched or exceeded the reference REF mixture strength. Sharp drops in strength were observed for LF contents above 50 % and ternary 70LF10FA mixture at 28 days. However, mixture 70LF10FA presented a considerable recovery in strength at 91 days with respect to its performance at 28 days. This was attributed to physical and chemical effects that occurred in between these 2 ages. The results of this study differed from Daminelli (2013) which observed an increase in compressive strength as LF content increased for a fixed w/b ratio.

Results of this study matched Madani et al. (2016), Zhao et al. (2015) and Dhir et al. (2007) with decreasing compressive strength across all ages as LF content increased for a fixed w/b ratio. Dhir et al. (2007) regarded the decrease in strength due to the low PC content as LF substitution increased. The relative increase in strength at 91 days when compared to 28 days was explained by Courard et al. (2018). Cementitious mixtures with LF form ettringite at first from monosulfate consumption. After monosulfate has been exhausted, monocarboaluminate was formed from hemicarboaluminate consumption. Once this latter phase was exhausted, the reactions cease leaving added calcium carbonate as a stable phase. Ettringite and monocarboaluminate filled pores as they formed, decreasing porosity and increasing compressive strength. On the other hand, leftover added carbonates tended to increase the number of pores which limited the increase in strength.

In the mixtures of this study, the minimum LF replacement content was 50 %. This content resulted in substantial carbonate formation which could explain the increase in porosity and decrease in axial compressive strength. Additionally, high BET specific surface could have resulted in particle coalescence and prevented water access to all grains. This limited LF activity, lowered carboaluminate generation, increased porosity and decreased strength. As noted by Perlot et al. (2013), these mechanisms could be further intensified by low w/b ratios (such as the 0.25 of this study). Another factor contributing to the decrease in strength as LF content increased would be the dilution effect as reactive particles were substituted with less reactive or inert ones. This dilution decreased hydration products and impacted strength development. In the case of the ternary LF7010FA mixture, pozzolanic effects increased strength due to the chemical effect of smaller pore size from secondary C-S-H formation and physical effects of particle packing (Ramezanianpour and Hooton, 2014; Tsivilis, 2010; Espining, 2008; Lothenbach et al., 2008).

Mixture 70LF10FA yielded 51.8 MPa of compressive strength at 91 days, which represented an 85 % increase with respect to the 28.0 MPa measured at 28 days. This was attributed to the synergic effect between LF and FA as noted by Deschner et al. (2012) and De Weerdt et al. (2011). This synergy was the result of the interaction between LF and aluminates from pozzolanic reactions of FA. This led to carboaluminate formation which decreased porosity and increased strength. This phenomenon was not observed at 28 days since, as noted by Detwiler and Mehta (1989), physical effects dominated in the first 7 days and synergy of physical and chemical effects became noticeable only after 28 days. Isaia et al. (2003) postulated that the increase in compressive strength in ternary mixture containing pozzolans was a hybrid effect: a combined synergy between PC hydration, FA pozzolanic reaction and LF physical effects. Thus, mixture LF70FA10, despite its low PC content (20 %), benefited from this exact synergy and achieved an optimum bond between PC and MA particles as important as the amount of hydration products formed.

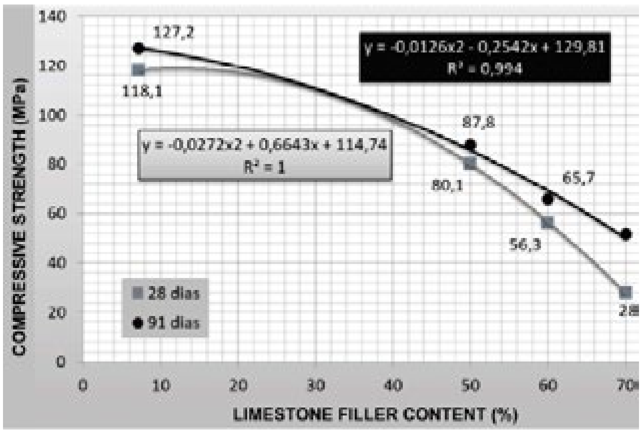
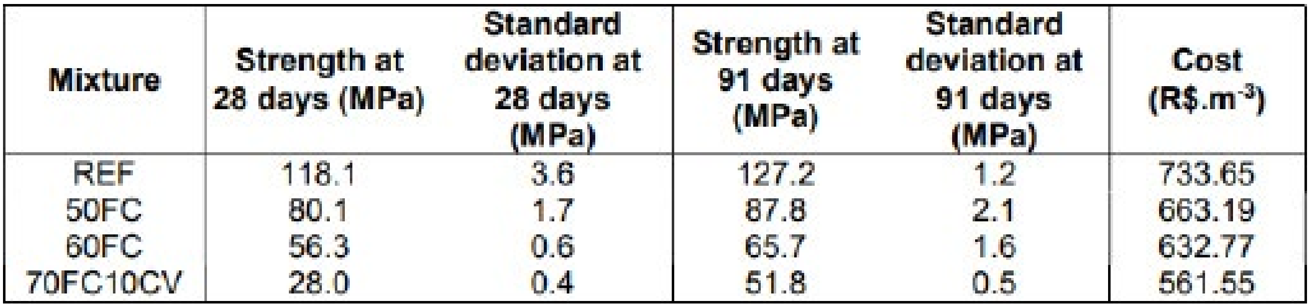


**Table 3:** Amounts of materials per kg/m3 of concrete.

**Source:** the authors



| Mixture | Strength at 28 days (MPa) | Standard deviation at 28 days (MPa) | Strength at 91 days (MPa) | Standard deviation at 91 days (MPa) | Cost (R$.m$^{-3}$) |
|---|---|---|---|---|---|
| REF | 118.1 | 3.6 | 127.2 | 1.2 | 733.65 |
| 50FC | 80.1 | 1.7 | 87.8 | 2.1 | 663.19 |
| 60FC | 56.3 | 0.6 | 65.7 | 1.6 | 632.77 |
| 70FC10CV | 28.0 | 0.4 | 51.8 | 0.5 | 561.55 |

**Table 4:** Average compressive strength, standard deviation and cost per m3 of concrete of the mixtures of this study

**Source:** the authors

### 4.1.1 Clinker intensity (Ick)

Clinker intensity (Ick) is a ratio that indicates the mass of clinker necessary to obtain one unit of axial compressive strength in MPa. Thus, lower Ick corresponds to better performance of the mixture.

Table 5 presents Ick for all mixtures of this study. Results show that all mixtures with MA presented lower Ick than the reference REF mixture. This could be attributed to decreased clinker consumption from the low w/b ratio of 0.25. For low w/b ratios (below 0.4), part of the cement remained un-hydrated since there was no physical space for hydrated products, especially Portlandite. Consequently, this un-hydrated portion could be replaced with more cost-effective materials such as LF (BENTZ, 2006; BONAVETTI et al., 2003). Thus, as w/b ratio decreased, axial compressive strength increased and Ick decreased.

At 28 days, mixture 50LF presented the lowest Ick, below 3 kgck/m3/MPa. This value was considered highly relevant in this area of study at this age. Further analyses showed that Ick at 91 days, compared to 28 days, were lower for all mixtures. This behavior was the result of clinker amount (kgck/m3) being fixed with respect to age while compressive strength increased over time.

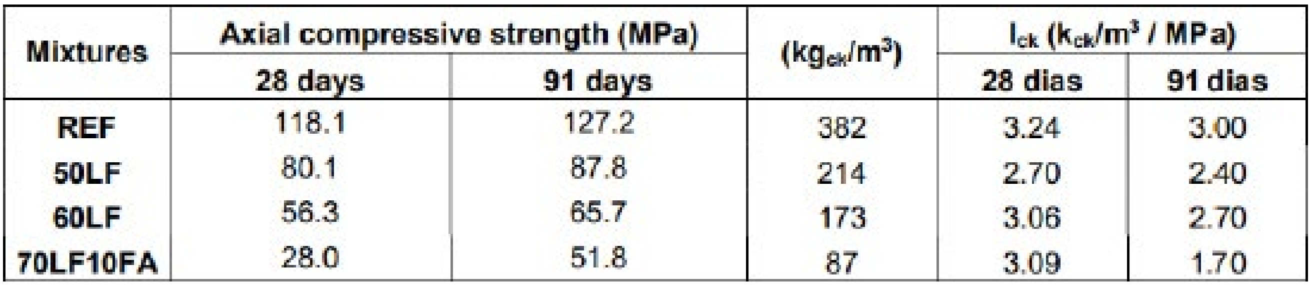
At 91 days, Ick presented the same trend of decreasing Ick as LF content increased. In this case, this behavior was related to the decrease in agglomerates as LF content increased. As compressive strength increased from 28 days to 91 days, Ick decreased forming the trend.

### 4.1.2 Binder intensity (Ib)

Damineli (2013) proposed a binder intensity index (Ib) to relate the consumption of agglomerates with respect to the compressive strength of concrete. This was represented by Ib (kg/m3/MPa) and consisted of the total amount of binders consumed in kg/m3 divided by the axial compressive strength in MPa at 28 days. From this definition, Damineli (2013) proceeded to compile the Ib of 156 national and international studies.

When calculating Ib, LF amounts were not included since it was considered inert. For the purposes of the index, PC was considered to have a reactivity factor (k) of 1 while remaining reactive MA materials, such as FA, were attributed factors in this scale based on their relative reactivity (HABERT, 2012). In this study, the definition of clinker intensity (Ick – kgck/m3/MPa) was based on the Ib concept of Damineli (2013).

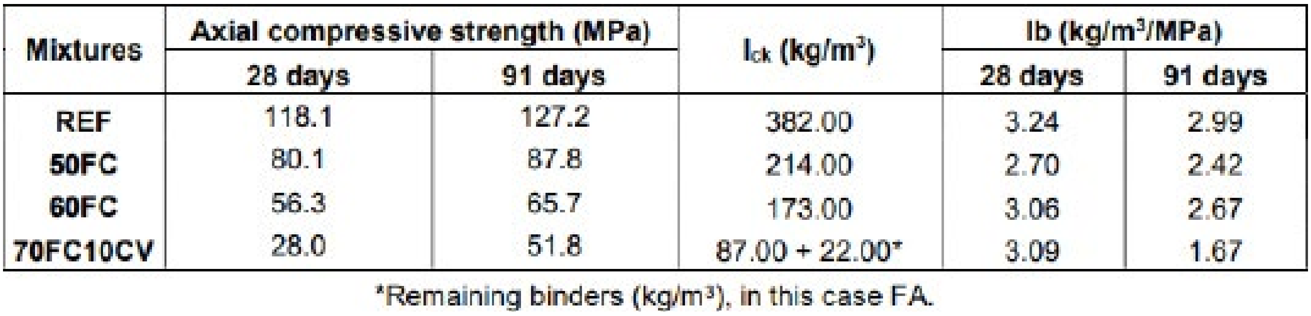
Table 06 presents Ib from converted Ick values for all mixtures of this study. Damineli (2013) noted that the minimum value for Ib was 5 kg/m3/MPa for concretes with compressive strength above 50 MPa. For concretes with strength below 50 MPa, Ib tended to be larger than this minimum value. Scrivener et al. (2018) expanded the concept of Ib and proposed a minimum Ib of 5 kg/m3/MPa for strengths above 50 MPa, 8 kg/m3/MPa for 30 MPa and a global average of 12 kg/m3/MPa for all variations in strength.



| Mixtures | Axial compressive strength (MPa) | | (kg$_{ck}$/m$^3$) | I$_{ck}$ (k$_{ck}$/m$^3$ / MPa) | |
|---|---|---|---|---|---|
| | 28 days | 91 days | | 28 dias | 91 dias |
| **REF** | 118.1 | 127.2 | 382 | 3.24 | 3.00 |
| **50LF** | 80.1 | 87.8 | 214 | 2.70 | 2.40 |
| **60LF** | 56.3 | 65.7 | 173 | 3.06 | 2.70 |
| **70LF10FA** | 28.0 | 51.8 | 87 | 3.09 | 1.70 |

**Table 5:** Clinker intensity (Ick) at 28 days and 91 days for the mixtures of this study.

**Source:** the authors



| Mixtures | Axial compressive strength (MPa) | | I$_{ck}$ (kg/m$^3$) | Ib (kg/m$^3$/MPa) | |
|---|---|---|---|---|---|
| | 28 days | 91 days | | 28 days | 91 days |
| **REF** | 118.1 | 127.2 | 382.00 | 3.24 | 2.99 |
| **50FC** | 80.1 | 87.8 | 214.00 | 2.70 | 2.42 |
| **60FC** | 56.3 | 65.7 | 173.00 | 3.06 | 2.67 |
| **70FC10CV** | 28.0 | 51.8 | 87.00 + 22.00* | 3.09 | 1.67 |

*Remaining binders (kg/m$^3$), in this case FA.

**Table 6:** Clinker intensity (Ick) at 28 days and 91 days for the mixtures of this study

**Source:** the authors

The concrete evaluated by Damineli (2013) was an ultra-high performance type (UHPC). It was produced from high pressure packing, thermal curing and consumption of special aggregates of 1,194.5 kg/m3. Resulting compressive strength was 800 MPa for an Ib of 1.49 kg/m3/MPa – high performance parameters but for an unconventional concrete.

In the case of this study, the binary 50LF mixture yielded an Ib of 2.42 kg/m3, which was lower than Damineli (2013) and others such as the 3 kg/m3 of Wongkeo et al. (2014). Thus, the 50LF mixture presented one of the lowest Ib registered so far and demonstrated the good performance of a concrete with high LF replacement content with respect to binding agents.

### 4.1.3 Normalized axial compressive strength (fcnorm)

Table 07 presents results of normalized compressive strength (fcnorm) for all mixtures of this study. This value was obtained by dividing the compressive strength by the amount of PC consumed (C) in kgck/m3.

As seen in Table 07, the fcnorm of all MA replacement mixtures at 28 days were greater than the reference REF mixture. This was the result of the definition of fcnorm accounting for the combined physical, chemical and synergic effects so that, with respect to the amount of clinker present, strength was greater for high LF replacement content.

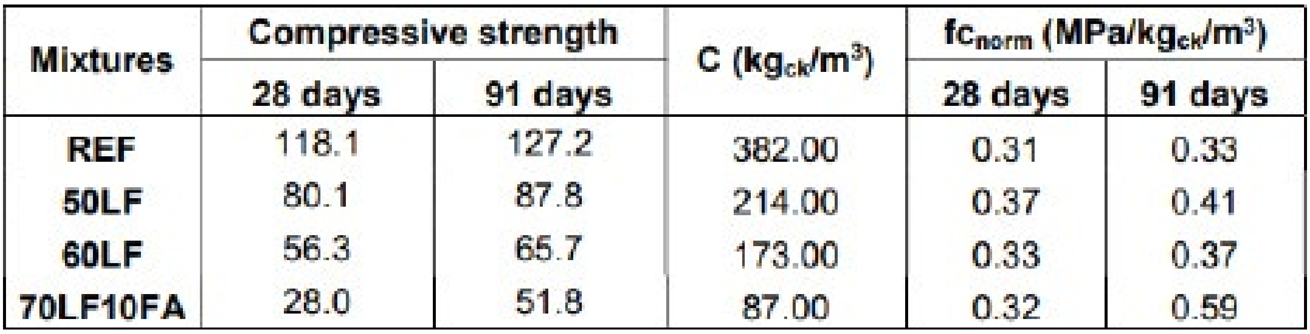
The increase in fcnorm could be the result of a “filler effect” in which LF interfered with PC hydration reactions through surface electrical potential (zeta potential) as noted by Scrivener et al. (2015). Of interest was the fcnorm of 0.32 MPa/kgcq/m3 for mixture 70LF10FA, which was only 3 % lower than the 0.33 MPa/kgcq/m3 of mixture 60LF despite having 50 % less clinker. This notable performance was attributed to synergic effects of LF and FA.

It was expected that aluminum oxides in FA would contribute to the added aluminate content in pozzolanic reactions. This would intensify LF interaction and increase carboaluminate production, leading to decreased porosity and increased normalized concrete strength (DESCHENER et al., 2012; DE WEERDT et al., 2011). However, it became apparent that FA did not fully realize its effect on compressive strength at 28 days (MEHTA; MONTEIRO, 2014). This was likely due to the low w/b ratio of 0.25 so that, at 28 days, FA contribution was mostly physical rather than pozzolanic (MINDESS et al., 2003).

Su and Miao (2003) suggested fcnorm between 0.11 MPa/kgck/m3 and 0.14 MPa/kgck/m3 for medium strength concretes with low PC content. On the other hand, Yu et al. (2015) cited values between 0.16 MPa/kgck/m3 and 0.19 MPa/kgck/m3 for high performance or eco-friendly concretes. In this study, the average fcnorm was of 0.33 MPa/kgck/m3 over all mixtures, which was 73.7 % higher than the upper limit proposed by Yu et al. (2015). Overall, it could be said that the mixtures of this study achieved a good performance with respect to fcnorm as they presented values higher than recommended in reference studies.

Binary mixtures 50LF and 60 LF presented small 9.76 % average increases in fcnorm over the tested ages. This was a reflection of the small increase of 9.96 % in compressive strength for both mixtures over the same time period. This indicated that fcnorm variation was proportional to the average variation in compressive strength. This was especially apparent at 28 days due to the small average increase in strength at this age.

Mixture 70LF10FA, as noted previously, had a considerable evolution in strength over time. Consequently, fcnorm performance was equally notable: at 91 days compressive strength was 51.8 MPa for a clinker consumption of 87 kgck/m3. This produced an fcnorm of 0.59 MPa/kgck/m3, the highest value of this study,



| Mixtures | Compressive strength | | C ($kg_{ck}/m^3$) | $fc_{norm}$ ($MPa/kg_{ck}/m^3$) | |
|---|---|---|---|---|---|
| | 28 days | 91 days | | 28 days | 91 days |
| REF | 118.1 | 127.2 | 382.00 | 0.31 | 0.33 |
| 50LF | 80.1 | 87.8 | 214.00 | 0.37 | 0.41 |
| 60LF | 56.3 | 65.7 | 173.00 | 0.33 | 0.37 |
| 70LF10FA | 28.0 | 51.8 | 87.00 | 0.32 | 0.59 |

**Table 7:** Normalized compressive strength (fcnorm) at 28 days and 91 days.

**Source:** the authors

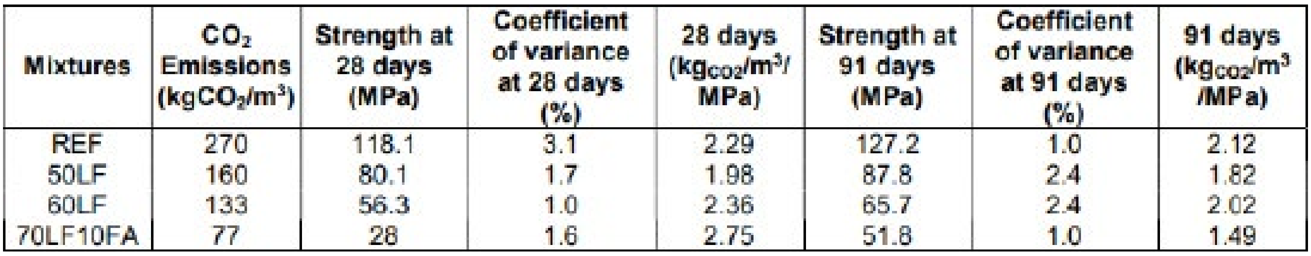
The good performance of mixture 70LF10FA, in addition to previously discussed factors related to LF, also benefitted from FA. As noted by Isaia et al. (2003), fcnorm increased considerably with the addition of FA due to the combined synergic effects of PC hydration, pozzolanic reactions of FA and physical effects of LF.

Over the ages tested, the average fcnorm over all mixtures was 0.42 MPa/kgck/m3, This value was higher than both Su and Miao (2003) and Yu et al. (2015). In particular, the fcnorm of 0.59 MPa/kgck/m3 of mixture 70LF10FA was over 3x greater than the upper limit proposed by Yu et al. (2015), making it a notable high performance eco-friendly concrete.

### 4.2 CO2 emissions per m3 of concrete

Table 08 presents CO2 emissions per m3 of concrete and its relation to the axial compressive strength. Values were determined with the following CO2 emission coefficients (in kgCO2/ton): (a) 617 for cement (GCCA, 2017); (b) 26 for LF (Habert et al., 2013); (c) 10 for FA; (d) 3 for sand; (e) 4 for gravel; (f) 5 for water and (g) 94 for the additive (Isaia e Gastaldini, 2004). These coefficients were average values for each material.

Table 08 shows that CO2 emissions were directly proportional to clinker content and axial compressive strength (Tables 03 and 04) and inversely proportional to LF content (Figure 02). The 50 % substitution of mixture 50LF presented a decrease in CO2 emissions of 40.7 % with respect to REF. Higher replacement of 60 % in mixture 60LF presented a 50.7 % decrease in CO2 emissions while the 70% replacement of 70LF10FA produced a corresponding reduction of 71.5 %. The direct relation between CO2 emissions and compressive strength were attributed to the need of increasing PC content to yield higher strengths as w/b ratio decreased. Kjellsen et al. (2005) noted that PC clinker was responsible for over 91 % of CO2 emissions of concrete. Consequently, any decrease in clinker content was a recommended option to decrease GHG emissions and increase global sustainability. Results of this study presented CO2 emissions from PC clinker of 87 % for the REF mixture, 82 % for 50LF, 80 % for 60LF and 70 % for 70LF10FA, which were proportional decreases in CO2 emissions as clinker content decreased.



| Mixtures | $CO_2$ Emissions ($kgCO_2/m^3$) | Strength at 28 days (MPa) | Coefficient of variance at 28 days (%) | 28 days ($kg_{CO2}/m^3$/ MPa) | Strength at 91 days (MPa) | Coefficient of variance at 91 days (%) | 91 days ($kg_{CO2}/m^3$ /MPa) |
|---|---|---|---|---|---|---|---|
| REF | 270 | 118.1 | 3.1 | 2.29 | 127.2 | 1.0 | 2.12 |
| 50LF | 160 | 80.1 | 1.7 | 1.98 | 87.8 | 2.4 | 1.82 |
| 60LF | 133 | 56.3 | 1.0 | 2.36 | 65.7 | 2.4 | 2.02 |
| 70LF10FA | 77 | 28 | 1.6 | 2.75 | 51.8 | 1.0 | 1.49 |

**Table 8:** Relationship between CO2 emissions and compressive strength.

**Source:** the authors

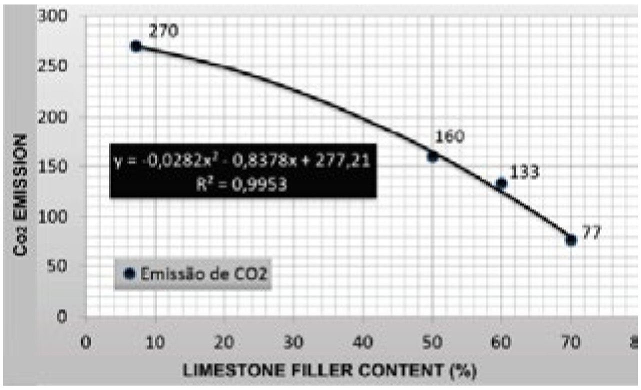


**Figure 2:** LF content (%) and CO2 emission (kgCO2.m-3)

**Source:** the authors

Costa (2012) used CP V-ARI cement to obtain a concrete with compressive strength of 50 MPa. This required a PC consumption of 487 kg/m3 and resulted in 485 kgCO2/m3 of emissions. In comparison, mixture 70LF10FA of this study had a strength of 51.8 MPa at 91 days from a PC consumption of 97 kg/m3 (equivalent to 87 kg/m3 of clinker) and emissions of 77 kgCO2/m3. Compared to the results of Costa (2012), this represented a 390 kg/m3 savings in PC and 408 kg/m3 less CO2 emissions, corresponding to decreases of 80 % and 84 %, respectively per $m^3$ of concrete with essentially the same compressive strength.

As noted by Damineli (2013), the higher the compressive strength of concrete, the less CO2 emissions in kgCO2/m3/MPa. This can be seen in Table 08 as emissions at 28 days were in agreement with Damineli (2013) except for REF mixture. At 91 days, binary mixtures presented decreases in CO2 emission index as compressive strength increased. Notably, the ternary 70LF10FA mixture, which had the lowest measured strength at 51.8 MPa, had a sharp decrease in emission index due to synergy and pozzolanic reactions. This was reflected in the 85 % increase in compressive strength from 28 days to 91 days while CO2 emissions remained constant.

Damineli (2013) obtained CO2 emission indices between 1.5 kgCO2/m3/MPa and 2 kgCO2/m3/MPa at 28 days, which were amongst the lowest reported in national and international studies. In this study, Table 08 presents an average emission index of 2.34 kgCO2/m3/MPa, which was 17 % higher than the cited upper limit. Damineli (2013) also noted that the smallest possible emission index for concrete production without clinker replacement was 4 kgCO2/m3/MPa at 28 days. In this study, mixture REF with no clinker replacement presented an emission index of 2.29 kgCO2/m3/MPa, or 42.8 % smaller than the proposed limit. These results demonstrated the good performance of the HSC of this study with respect to CO2 emissions per m3 of concrete and further signaled the positive impact in GHG emissions when PC was replaced with LF and FA.

### 4.3 Cost per m3 of concrete

Table 04 shows that cost was directly proportional to compressive strength and CO2 emissions while also inversely proportional to LF content. The first two factors were related to the PC content of each mixture, which was also the costlier material and higher GHG emitter per m3 of concrete. In contrast, excluding water, LF was one of the cheaper materials and its increasing replacement ratio contributed to the reduction of cost per m3 of concrete.

### 4.4 Susteinability comparisons

Results of Table 08 showed that binary and ternary mixtures containing LF had higher averages, with average global index of 63.2 % and coefficient of variance of 2.7 % at 28 days. Mixture 70LF10FA had the lowest CO2 emissions and cost but also low axial compressive strength at 28 days. It should be noted that all three replacement mixtures had similar cost at 28 days due to the elevated consumption of admixture. Nonetheless, at this age, mixture 50LF was considered the more sustainable option since it produced an fc28 of 80.1 MPa. In contrast, mixture 7LF10FA had the lowest performance at the same age with an fc28 of only 28.0 MPa despite its 80 % clinker substitution ratio. At 91 days, mixture 70LF10FA presented the highest average index of 74.1 %, followed by mixtures 50LF and 60LF with average indices of 64.9 %. The turnaround between 28 days and 91 days of mixture 70LF10FA was due to the 85 % increase in axial compressive strength to an fc91 of 51,8 MPa (the best performance at this age) while emissions and cost remained fixed. In comparison, mixtures 50LF and 60LF improved strength by only 9.6 % and 16.7 % from 28 days to 91 days, respectively. The relative increase in compressive strength as PC content decreased could be attributed to more available space in the mixtures for hydration reactions to occur. As noted by Isaia et al. (2003), special consideration must be given to aging since MA actions, in particular pozzolans, evolved over longer and longer times as synergic effects increased. Consequently, it should be stressed that sustainability did not depend solely on PC replacement with MA but also with ages in which concrete properties and cost were evaluated. Additives also contributed significantly to cost since polycarboxylate-based latest generation types tended to be expensive.

Results also demonstrated the possibility of producing structural concrete with PC and LF. At 28 days, mixture 50LF had PC content in the order of 238 kg/$m^3$ and fc28 of 80 MPa while mixture 60LF had 192 kg/$m^3$ and 56.6 MPa, respectively. Mixture 70LF10FA at 91 days had an fc91 of 51.8 MPa with only 97 kg/$m^3$ of PC content (87 kg/$m^3$ of clinker) and 104 L/$m^3$ of water. These values were equivalent to a clinker intensity of only 1.68 kgck/MPa (2.32 kg/MPa of binder intensity) and were one of the smallest values reported in this area. These results denoted an exceptional sustainability capacity of this mixture and confirmed other similar results such as Damineli (2013).

The statistical analysis of Isaia et al. (2012) gathered 7,308 results on microstructural and durability factors. It was shown that, in the case of dependent variables in general multiple linear regression models, MA content and w/b ratio had the most statistical significance. Regarding durability, Isaia et al. (2012) reported the best performance for a concrete with 70 % MA content and w/b ratio of 0.35, for a compressive strength of 50 MPa. This performance was attributed to physical, chemical and synergic effects that developed over time. Similar results were observed in this study with the high MA replacement content of mixture 70LF10FA and w/b ratio of 0.25, which corresponded to a water consumption of around 100 L/m3.

## 5. CONCLUSIONS

This study presented HSC of binary mixtures with 50 % and 60 % LF content and ternary mixtures with 70 % LF and 10 % FA content. The concretes were produced with reduced water content, in the order of 100 L/m$^3$, and particle packing techniques.

Resulting compressive strengths were in the order of 50 MPa to 80 MPa at 28 days for the binary mixtures and 50 MPa at 91 days for the ternary mixture.

In terms of sustainability parameters, a concrete was produced with a 51.8 MPa compressive strength at 91 days and 77 kgCO2/m3 of emissions in a mixture in which 80 % of PC was replaced by 70 % LF and 10 % FA. The resulting eco-friendly concrete had low clinker (87 kg/m3) and water (104 L/m3) content. This demonstrated that a sustainable HSC with low carbon emissions could be obtained with high LF content and low clinker content of only 1,68 kgck/MPa. The combination of high LF content and low w/b ratio was possible due to controlled granulometry through particle packing and the resultant concrete had lower GHG emissions, lower cost and more global sustainability potential.

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SSS: conceptualization, data curation, formal analysis, investigation, validation, writing - original draft and writing - review & editing.

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GAI: investigation and writing - original draft.

ABSSN: supervision, visualization and writing - review and editing.
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