Use of reclaimed fly ash for the production of sustainable cementitious composites

Uso de cenizas volantes de depósito para la producción de compuestos cementicios sostenibles

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Abstract

This study analyzes the use of reclaimed fly ash (RFA) as an alternative cementitious material in engineered cementitious composites (ECC) mixtures. The performance of RFA-ECC is assessed by evaluating the tensile properties at 7 days, for two replacement levels of ordinary portland cement (OPC) (50% and 70%) and three polyethylene (PE) fiber contents (1.5%, 1.75%, and 2% by volume fraction). Results showed that RFA-based ECC can produce strain-hardening behavior and reach high tensile stress and strain. RFA-ECC with 50% OPC replacement and 1.5 vol.% PE fiber content exhibits average tensile strength and strain of 6.4 MPa and 8.9%, respectively. Furthermore, at 28 days, the ductile behavior remains. The study shows that RFA can be considered an alternative low carbon constituent to replace OPC in ECC mixtures, to reduce their carbon footprint, hence leading to a more sustainable cementitious composite.

Keywords: Reclaimed fly ash; ductility; engineered cementitious composites; tensile behavior; polyethylene (PE) fibers.

Resumen

Este estudio analiza el uso de cenizas volantes de déposito (reclaimed fly ash, RFA) como material alternativo al cemento en compuestos cementicios dúctiles (engineered cementitious composites, ECC). El desempeño de los RFA-ECC se evalúa por medio del comportamiento a cargas de tracción a los 7 días, para dos niveles de reemplazo de cemento portland (ordinary portland cement, OPC) (50% y 70%) y tres contenidos de fibra de polietileno (PE) (1,5%, 1,75% y 2% en volumen). Los resultados mostraron que ECCs confeccionados con RFA pueden producir un comportamiento de endurecimiento por deformación y alcanzar altas tensiones y deformaciones en tracción. Compuestos RFA-ECC, con 50% de RFA como reemplazo de OPC y 1,5% de contenido de fibra PE en volumen, exhiben una resistencia a la tracción y una deformación promedio de 6,4 MPa y 8,9%, respectivamente. Además, a los 28 días se mantiene el comportamiento dúctil. El estudio muestra que RFA puede considerarse un componente alternativo con bajas emisiones de carbono para reemplazar parcialmente el OPC en mezclas ECC. De este modo, se reduce su huella de carbono y, por lo tanto, se obtiene un compuesto cementicio más sostenible.

Keywords: Cenizas volantes de depósito; ductilidad; compuestos cementicios dúctiles; comportamiento a tracción; fibras de polietileno (PE).

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1. Introduction

Among the challenges faced by concrete construction in the 21st century are the higher expectations of service life, the reduction of the carbon emissions of concrete, and the improvement of the overall performance and sustainability of concrete infrastructure (Monteiro et al., 2017). As summarized by Swamy (Swamy, 2001), the design of sustainable concrete can be met by developing durable and functional concrete, using waste or recycling materials in concrete mixtures, and minimizing the environmental impact of concrete construction.

Engineered Cementitious Composites (ECC), also known as Strain-Hardening Cementitious Composites (SHCC), correspond to a type of material that can improve the functionality and durability of concrete infrastructure. The term ECC comprises a range of fiber-reinforced cementitious composites, with significantly higher ductility and outstanding crack control compared to conventional concrete. While normal concrete has an ultimate tensile strain of approximately 0.01% (Li, 2012), ECC typically exhibits tensile strain capacities beyond 2% (Li, 2019). This means that ECC can deform over 200 times more than standard concrete.

Unlike other cementitious-based materials, ECCs exhibit strain-hardening behavior. This means that, as ECC deforms, the material increases its tensile strength. To generate a strain-hardening behavior under tensile loads, two criteria must be met: the strength criterion and the energy criterion. These criteria depend on the properties of the fiber, the cementitious matrix, and the interaction between the matrix and the fiber, commonly described by the fiber-bridging capacity. The strength criterion indicates that the fiber-bridging capacity at the crack with the lowest capacity among the already-formed cracks must be greater than the tensile load required to generate a new crack (Yu et al., 2018). The energy criterion indicates that flat cracks should be generated where the crack opening remains relatively constant as the crack length increases (Li et al., 2001). It is expected that multiple small cracks will form, releasing energy progressively instead of forming a single large crack, as is the case with traditional concrete. For this to happen, the maximum complementary energy must be greater than the matrix toughness at the crack tip (Zhang et al., 2019). For the ECC to exhibit ductile behavior, both criteria must be met simultaneously.

Under strain-hardening behavior, ECC specimens exhibit multi-cracking behavior, where the cracks usually have openings between 0 to 100 μ m (Li, 2003). Due to the controlled crack widths developed by ECC under tensile stress, the composite can develop low permeability and enhanced durability at different service conditions (Liu et al., 2017). Traditionally, to lower the high cement content in ECCs, a high replacement of fly ash (FA) has been used. Recently, given the increasing concern about the reduction of the carbon footprint of the concrete industry and the expected scarcity of fly ash, low-carbon constituents of ECCs have been explored (Leon-Miquel et al., 2023; Shoji et al., 2022; Zhu et al., 2023). Among these, the incorporation of reclaimed fly ashes (RFA) into ECC mixtures has received little attention.

FAs are a by-product of carbon combustion in coal power plants. This combustion by-product is carried by exhaust gases and then captured by cyclones or an electrostatic precipitator to avoid the release of the material to the air (Norton et al., 1986); (Tosun-Felekoglu et al., 2017). Fly ash obtained from thermoelectric plants is one of the most commonly used supplementary cementitious materials (SCMs) in concrete and cementitious composites (Scrivener et al., 2018). However, in the last decade, the availability of FA to be used as SCM in concrete has decreased significantly, due to the emergence of cheaper alternatives of energy generation and stricter environmental requirements and emission standards. The operation of thermoelectric power plants poses a health risk for adjacent communities (Cortés et al., 2019), which is why many of them have been decommissioned in recent years. In Chile, a total shutdown of these plants is expected by 2040 (Ministerio de Energía, 2021). In the United States, no coal-fired power plants have been built since 2013, and plant retirements are expected to continue through 2040 (Al-Shmaisani et al., 2019). Due to the ongoing closure of thermoelectric plants, there is a need for new and abundant SCMs that can satisfy the demand for fly ash, providing a sustainable cementitious alternative to cement in mixtures.

Unlike regular fly ash, reclaimed fly ash (RFA) is stored in landfills, with direct exposure to the environment over time. As this wet-storage time increases, the fineness and loss on ignition (LOI) of the RFA increases (Al-Shmaisani et al., 2019); (McCarthy et al., 2017). Additionally, since reclaimed fly ash is exposed to the environment for extended periods, it is likely subjected to higher moisture, which can lead to physical and chemical changes during storage, causing variability in the ash and potentially affecting its reactivity (McCarthy et al., 2017).

This article assesses the feasibility of the use of RFA as a replacement for OPC to produce of sustainable ECC mixtures. The performance of the RFA-ECCs is evaluated by measurements of their tensile capacity: tensile strain and strength.

2. Materials and methods

2.1 Materials and mixture proportions

ECC mixtures were prepared using ordinary portland cement (ASTM C150 Type I (ASTM International, 2024), OPC) and different OPC replacement levels by reclaimed fly ash (RFA). (Table 1) shows the chemical composition of the cementitious materials, as obtained by X-ray fluorescence (XRF).

Material	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	Na ₂ O	MgO	CaO	K ₂ O	SO ₃	TiO ₂	P ₂ O ₅	L.O.I
OPC	21.6	5.0	3.4	0.3	1.2	62.9	0.6	3.2	0.2	0.2	1.1
RFA	59.0	20.9	6.9	2.3	1.9	1.9	1.5	1.0	0.6	0.2	3.3

Table 1. Chemical compositions of OPC and RFA.

The fine aggregate used in the ECC mixtures was silica sand. The sand was obtained from a riverbed quarry located in Valparaíso, Chile. The particle size of the sand ranged between 75 and 300 μ m (sieves #50 to #200), the specific weight was 2.7, and the absorption was 0.56%. The addition of a superplasticizer (SP) was considered for each ECC mixture. The SP was ViscoCrete-5100 CL, a polycarboxylate-based high-range water reducer with a specific weight of 1.085.

The fibers used in this study were polyethylene (PE) fibers. This type of fiber was selected because it has a lower embodied energy than other commonly used fibers in ECC mixtures (Shoji et al., 2022). The dimensions and properties of the PE fibers used in this study are shown in (Table 2).

Table 2. Physical and mechanical properties of PE fiber.

Fiber	Diameter	Length	Strength	E	ε _u	Density
	(µm)	(mm)	(MPa)	(GPa)	(%)	(g/cm³)
PE	18	12.7	2900	117	< 4	0.97

2.2 Preparation of ECC Mixtures

The feasibility of the production of RFA-based ECCs was assessed using 2 levels of OPC replacement (50% and 70%) and 3 fiber volume additions (1.5, 1.75, and 2.0% of the total volume of the ECC mixture).

The mixture designs of ECC are presented in (Table 3), where the nomenclature of each ECC mixture is denoted as "RFA-ECC-XX%-Y.Y", where "XX%" corresponds to the OPC mass replacement by RFA and "Y.Y" corresponds to the volume addition of PE fiber, as a percentage of the total volume.

Material	OPC	Sand	RFA	Water	SP	PE (vol.%)
RFA-ECC-50%-1.5	687	343	687	343	13.1	1.50
RFA-ECC-70%-1.5	399	200	998	349	8.8	1.50
RFA-ECC-70%-1.75	399	200	998	349	8.8	1.75
RFA-ECC-70%-2.0	399	200	998	349	14.0	2.00

Table 3	Mixture	design	of RFA-FCC	(kg/m^3)
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Mixing was performed in a 5-liter bowl. First, all solids (cement, reclaimed fly ash, and sand) were mixed for 2 minutes at 140 rpm. The water and the superplasticizer were mixed separately and added to the solids at a constant pace, while the equipment was mixing. After a total mixing time of 7 minutes, the corresponding dosage of PE fibers was slowly included. Once all fibers were introduced into the mixture, the speed of the equipment was increased to 285 rpm. After a total mixing time of 13 minutes, mixing was stopped.

Fresh ECC mixtures were molded into dogbone-shaped specimens. The mixture was evenly spread on steel molds, and a trowel was used to flatten the surface and remove any excess mixture. Finally, an acrylic plate was placed over each mold to keep a flat surface and avoid water evaporation before demolding.

Dogbone specimens were demolded after 24±0.5 h. After demolding, each specimen was wrapped with plastic bags, and kept in sealed conditions until testing, at a constant temperature of 22±2 °C. Tensile tests of the ECC specimens were performed at 7 days from mixing. Additionally, as the reactivity of the cementitious materials could compromise the ductility of ECC, samples of mixture RFA-ECC-50%-1.5 were also tested at 28 days.

2.3 Tensile Test

The uniaxial tensile test determines the stress-strain behavior of ECC. Tests were performed on a set of three dogbone specimens for each mixture. The tests were performed on a 100-kN Zwick-Roell universal testing machine, under displacement control at a rate of 0.5 mm/min, following the recommendations of the Japan Society of Civil Engineers (JSCE) for testing of high performance fiber reinforced cementitious composites with multiple fine cracks (JSCE, 2008). The tensile loads were recorded using a universal testing machine, while the strains were measured using two linear variable displacement transducers (LVDTs). The approximate gauge lengths were 100 mm. The dimensions of the dogbones and the experimental setup are shown in (Figure 1).



Figure 1. (a) Dimensions of the dogbone specimens and (b) experimental tensile test setup.

3. Results and analysis

3.1 Effect of RFA addition

(Figure 2) shows the results of the tensile test for ECC specimens with 50 and 70 wt.% OPC replacement, and 1.5 vol.% fiber content. The curves exhibit the traditional trend observed on ECC specimens undergoing strain-hardening behavior. After the formation of the first crack, the tensile stresses and strains increase progressively until failure. Each stress drop in the curves indicates the formation of a crack. This trend confirms the occurrence of a strain-hardening behavior of the RFA-ECCs.



Figure 2. Results of tensile tests for ECC samples using OPC replacement of 50% and 70% by RFA, and a 1.5 vol.% fiber content a) RFA-ECC-50%-1.5, b) RFA-ECC-70%-1.5.

The RFA-ECC with a 50% replacement of OPC and 1.5 vol.% PE fiber reaches maximum tensile strain values over 10%, 1000 times higher than conventional concrete. On the other hand, when the replacement level increases to 70%, the ductile performance of the RFA-ECC worsens; the strength at the first crack, the maximum tensile stress, and the strains were lower compared to RFA-ECC-50%-1.5. An increase in the replacement of fly ash produces a decrease in the mechanical properties of concrete at early-ages. In ECC, the decrease in the mechanical properties of the matrix can explain the lower stress at the first crack. (Yang et al., 2007) showed that an increase in regular fly ash replacement decreases the maximum tensile strain, tested at 3 days. At replacement levels between 55% and 85%, the ductility decreased from 4.6% to 3.8%. They showed that an increase in the FA to cement ratio leads to an increase in the interface bond between fibers and the cementitious matrix and a decrease in matrix toughness, which should facilitate the fulfillment of the strength and energy criteria. However, the results of the RFA-ECC do not clearly indicate that the criteria will be met at higher replacements. The increase in RFA replacement reduces the ductility of ECC and negatively affects ductile behavior. Overall, based on the results, RFA-ECC can be produced with replacements of 50% and 70%.

3.2 Effect of fiber addition

(Figure 2b) and (Figure 3) show the tensile test results of ECC mixtures with 50 wt.% OPC replacement and 1.5, 1.75, and 2 vol.% fiber contents. The results indicate that the addition of a higher vol.% of fiber content leads to a reduction in tensile strain. Previous studies (Said and Razak, 2015); (Wang et al., 2020) analyzed the effect of PE fiber content in a FA-ECC sample. It was found that increasing the fiber volume addition leads

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to higher number of cracks, of reduced widths and spacing, and lower drops of tensile stress. Consequently, higher tensile stress and strain were measured on FA-ECC specimens. The results of RFA-ECC specimens under tensile loading do not coincide with the performance of the FA-ECC. Wang et al. (Wang et al., 2020) showed that an increase in vol.% of fiber content decreases the workability of the mixture, which negatively impacts the dispersion and orientation of the fibers. When using RFA, higher OPC replacement produced ECC harder to mix and with a higher number of fiber lumps. The mixture RFA-ECC-70%-2.0, with the highest fiber content of this study, showed an extremely poor workability during mixing.





3.3 Evolution of tensile properties

The results of RFA-ECC-50%-1.5 specimens after 28 curing days are shown in (Figure 4). At 28 days, the specimens achieve lower tensile strains, but higher tensile stresses compared with measurements at 7 days. With longer curing times, the cementitious matrix develops higher strength through the formation of a stronger molecular and fiber-matrix bond, as a result of a more complete hydration process (Li, 2019). As hydration increases, the toughness of the matrix also increases, leading to a reduction in the ductility of the specimen (Lepech & Li, 2006). This phenomenon could also explain the performance of RFA-ECCs; as the matrix continues to hydrate over time, the composite becomes stronger. Consequently, the tensile load required to generate a new crack increases. This load will exceed the fiber bridging capacity, preventing the strength criteria from

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being met and causing the mixture to lose its strain hardening behavior. As a result, the strain capacity decreases compared to the mixture at 7 days.



Figure 4. 28 curing day tensile tests for PE-1.5-50 specimen.

The average tensile strength (σ_{max}) and ultimate tensile strain (ε_u) of RFA-ECCs are included in (Table 4).

Curing days	Material	σ_{max} (MPa)	ε _u (%)
	RFA-ECC-50%-1.5	6.4 ± 0.8	8.9 ± 3.0
7.1	RFA-ECC-70%-1.5	3.7 ± 0.6	7.7 ± 2.9
/ days	RFA-ECC-70%-1.75	3.6 ± 0.2	5.4 ± 1.4
	RFA-ECC-70%-2.0	2.7 ± 0.2	2.2 ± 1.2
28 days	RFA-ECC-50%-1.5	7.8 ± 0.6	5.9 ± 1.3

Table 4. Average tensile stress	(σ	and strain cana	acity (<i>s</i>) in FCC mixtures
Table - Average tensile stress	(Umax/	and strain cape		u in LCC mixtures.

The performance of RFA-ECC specimens after 28 curing days can be compared with results of FA-ECC in other studies. For instance, Wang et al. (Wang et al., 2020) developed ECC mixtures with 30 wt.% fly ash replacement and 1.5 vol.% PE fiber content. They obtained a maximum tensile strength and strain of 8.2 MPa and 9.4%, respectively. The RFA-ECC-50%-1.5 mixture, with an OPC replacement 20% higher than the study performed by Wang et al., reaches tensile strengths and strains 5% and 37% lower, respectively. Another study conducted by Said and Abdul (Said and Razak, 2015) used a OPC replacement of 25% with fly ash cenospheres and 1.5 vol.% PE fiber content. They measured a peak tensile strength of 4.0 MPa and a tensile strain of 2.5%. In comparison, a 95% and 136% higher tensile strength and strain is achieved in the present study, respectively.

Leon-Miquel et al. (Leon-Miquel et al., 2023) showed that cement is the constituent of ECC mixtures with highest contribution to ECC's CO_2 emissions. They replaced 10% to 50% of OPC by natural pozzolans in ECC mixtures. At the highest replacement, the CO_2 emissions of the mixture were reduced by 39%. Thus, the replacement of OPC with low-carbon cementitious materials is one of the most effective ways to reduce the carbon footprint of ECC. Additionally, the superior performance of ECC is capable of providing enhanced durability to concrete structures. (Hou et al., 2024) performed a life-cycle analysis on a bridge deck system and found that the use of ECC to replace joints in link slabs can reduce carbon emissions by half compared to traditional concrete. The high replacements of RFA reached in this study suggest that sustainable ECC can effectively be produced using abundant reclaimed fly ashes.

4. Conclusions

This study analyzes the use of reclaimed fly ash (RFA) to produce sustainable ECC mixtures. To assess the feasibility of the development of RFAbased ECC, two high OPC replacements by RFA (50% and 70%) and three fiber volume additions (1.5%, 1.75%, and 2.0%) are evaluated. The dogbone-shaped specimens produced from the RFA-based ECC mixtures were measured under tensile testing at 7 days from mixing. Additionally, the effect of the hydration process on the evolution of ductility was analyzed by comparing the tensile behavior of a selected mixture at 7 and 28 days from mixing.

Based on the results, the following conclusions were drawn:

- Mixture RFA-ECC-50%-1.5 exhibited the highest ductility (maximum tensile strain) and tensile strength (maximum tensile stress) among the samples tested at 7 days. The increase of OPC replacement from 50% to 70% showed a negative effect on ductility.

- The fiber content used in RFA-based ECCs has a significant impact on their tensile behavior. The increase of the fiber volume fraction from 1.5% to 2.0% produced a reduction in the ductility and tensile strength of the samples.

- The tensile behavior of RFA-based ECC with 50 wt.% OPC replacement and 1.5 vol.% PE fiber changes between testing at 7 and 28 days from mixing. The ductility decreased and the tensile strength increased, results that can be attributed to the evolution of the hydration of the cementitious materials.

Overall, the results of this study indicate that the replacement of OPC by RFA on ECC mixtures allows for the development of ductile composites. Furthermore, considering that OPC has a high impact on the carbon footprint of ECC, the use of a low-carbon cementitious material has a beneficial effect on the sustainability of the material.

Even though the results achieved by RFA-ECC samples presented in this study show a good tensile behavior, a more in-depth analysis is recommended to accurately determine the effects of increasing the RFA replacement level and the fiber content. This can be achieved by performing a micromechanical analysis, in which the parameters included in the strength and energy criteria can be measured or calculated. The determination of the micromechanical parameters is commonly done by the toughness test, single fiber pullout test, and single crack test (also called the notched dogbone test). Toughness tests are important for quantifying the matrix toughness which is essential for determining if the energy criteria is met or not. These parameters are important to correctly assess the replacement level of a selected RFA. An optimal mix design could lead to the replacement of the novel RFA in higher ratios than the ones analyzed in this study, hence reducing the carbon footprint of the RFA-ECC mixtures. A more detailed micromechanical analysis may also indicate why some of the samples analyzed in this study exhibited reduced tensile stresses and strains at higher RFA replacement and fiber content.

Finally, a life cycle analysis is suggested to assess the actual environmental impact of the OPC replacement and quantify the benefits of the implementation of RFA-based mixtures on concrete infrastructure.

5. Notes on Contributors

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