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Application of electric arc furnace slag in building concrete: Environmental and structural characterization[☆]

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ABSTRACT

Recycling industrial by-products into sustainable building products is essential for advancing to a circular economy. These products must meet structural requirements and be environmentally safe and not contaminate soil or water through leaching. This study assesses the leaching of hazardous elements into the environmental and the structural performance of replacing natural aggregate with Electric Arc Furnace Slag (EAFS) in the manufacture of structural concretes ($F_{cu} \geq 25$ MPa). The study compares Slag Concrete (SC) and Limestone Concrete (LC) as a reference, throughout different stages of the life cycle of the material. A battery of harmonized leaching tests, including dynamic surface, granular compliance, percolation, and pH-dependence tests, were conducted alongside mechanistic modelling to evaluate the performance of slag-based concrete across different scenarios. The study found that among the trace elements, As, Ba, Cd, Cr, Cu, Hg, Mo, Ni, Pb, Sb, Se, V and Zn, determined in leachates, during the service life stage, only Vanadium (V) showed significant leaching differences in EAFS-based concrete, with diffusion being the main release mechanism. At the end-of-life stage, granular specimens met European inert material standards for secondary resource use. The pH-dependence test revealed varying leaching behaviours, likely due to surface adsorption with Fe oxides for Chromium (Cr) and solid solution formation with cement minerals for Molybdenum (Mo) and Vanadium (V). The results highlight the importance of multiple leaching tests for thorough environmental assessment during the product lifecycle. The results obtained can contribute to the basis of a pre-normative study for the use of EAFS aggregate in the manufacture of structural concretes and thus move towards sustainable building.

1. Introduction

Steel slags constitute a high-volume by-product stream from the steelmaking process. Each year, around 20 million tons of steelmaking slags are generated in the European Union, of which more than 30 % are produced in the electric arc furnace steelmaking route [1]. The electric arc furnace steel production uses scrap metal as feedstock and is expected to grow in developed countries due to

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lower operational costs than the Basic Oxygen Furnace integrated route [2]. According to the International Energy Agency [3], EAF steel production is projected to increase from 28 % in 2019 to approximately 38–40 % by 2050 globally, with developed countries expected to exceed 50 %. Hence, coupled with the increase in steel demand up to 2070 [3], the generation of electric arc furnace slags (EAFS) is likewise expected to be increased.

The valorisation of these slags in building materials as supplementary cementitious material or natural aggregate replacement has already been performed, which is a management option that reduces the embodied CO_2 of the secondary materials obtained, increases resource efficiency, and is in line with circular economy policies for the manufacture of building materials while mitigating the hazards associated with the disposal in landfills of slags [4,5]. Particularly, the incorporation of EAFS has been addressed in different kinds of construction materials such as road basements and asphalts [6,7], alkali-activated materials [8] and concrete [9–11]. The latter option is of particular interest, as concrete is the most employed construction material in the world [12,13]. EAFS have higher physical properties than natural and other waste derived aggregates [7], and when introduced in concrete mixes they reach similar compressive strength and durability compared to concrete made of natural aggregates [10,14–16], thus constituting a reliable way of valorisation of these type of steelmaking slags. The use of EAFS as an aggregate in concrete can be applied in structural elements of buildings, such as columns and slabs, where high strength and durability are required. However, while this application is viable, it is important to ensure that the CaO and MgO present in the slag have been properly inertized to prevent expansive reactions. Additionally, these concretes exhibit significantly higher density compared to conventional mixes, which may be a limiting factor in applications where weight reduction is critical.

However, the EAFS are characterized by the presence of inorganic elements of potential concern that can be released to the environment through leaching processes. Although they are listed in the European Waste Catalogue (code 10 02 02) as non-hazardous, it is particularly recommended to monitor the leaching of Cr, Mo, Se and V [17,18], since their incorporation to building materials does not ensure the absence of leachability of these elements [4]. Indeed, the European Construction Products Regulation [19] states that any marketable construction material should not entail any undesirable risk to the ecosystems and human health, and therefore the release of potentially hazardous elements to groundwater and soil should be assessed through leaching tests or methods for construction materials [20]. To address this question, two methods have been proposed in the European Union for characterizing the leaching behaviour of construction materials [21]; the Dynamic Surface Leaching Test (DSLT, CEN/TS 16637-2) [22] for monolithic building materials (e.g. concretes, bricks) and the up-flow percolation leaching test (CEN/TS 16637-3) [23] for granular building materials (e.g. aggregates). In other parts of the world, analogous tests to those proposed by the EU are used, although many of them are specifically developed for waste. The test methods employed in the USEPA standards are procedurally similar to those prepared by the CEN, with minor deviations such as time intervals and the number of steps, leading to equivalent results [24]. For instance, EN 16637-2 is similar to USEPA 1315, EN 16637-3 to USEPA 1314, and EN 12457 differs from the TCLP in the leachant used (deionized water versus acetic acid solution at pH 4.93) and contact time (24 h versus 18 h). Additionally, EN 16637-2 and EN 16637-3 are identical to NEN 7375 and NEN 7373, respectively. National standards include the Brazilian NBR 10005 and the Chinese HJ/T 299–2007 and HJ/T 300–2007.

The leaching assessment of concrete incorporating EAFS as aggregate has been addressed in a few studies, which consisted in performing a single leaching test either granular or monolithic. These studies typically compare the concentrations obtained in the leachates with reference materials, such as concrete with natural aggregates, or regulatory limits. Specifically, the single-stage leaching test for granular samples, EN 12457, proposed by the European Union, is applied, and the results are compared with the Italian limits for waste recycling [14,25]. Additionally, the dynamic surface leaching test (DSLT) for monolithic materials is employed, with results compared against the Dutch Soil Quality Decree [15] or reference concrete [26]. These findings demonstrate that concrete incorporating EAFS as aggregate meets environmental standards, highlighting its potential for sustainable construction practices.

Although necessary, this evaluation is insufficient and a thorough environmental assessment with a combination of leaching tests that reflect the intended use and possible exposure scenarios that the material may be subjected to should be conducted. This integrated approach allows to determine the characteristics that govern the release of substances and their controlling release mechanisms, and thus to predict the long-term leaching behaviour of the tested material in its entire lifecycle, which is essential in construction materials partial or entirely made of waste and industrial by-products to promote material circularity in the construction industry [27].

In absence of such a study, this work proposes the assessment of the leaching of hazardous elements into the environment of concretes, SC (Slag Concrete) made with EAFS as a natural aggregate replacement and Limestone Concrete (LC) as a reference during the entire life cycle of the material using a battery of harmonized leaching tests in order to correctly interpret complex leaching processes. The inorganic trace elements of potential concern As, Ba, Cd, Cr, Cu, Hg, Mo, Ni, Pb, Sb, Se, V, and Zn in leachates were determined through a scenario-based approach: (i) The EAFS was initially evaluated with the compliance leaching test (EN 12457-4) to identify the environmentally critical elements, (ii) the service life assessment is performed with the Dynamic Surface Leaching Test (DSLT, CEN/TS 16637-2) to monolithic concrete specimens and (iii) an end-of-life assessment of valorisation of the granular products for use as a secondary resource is conducted through the following leaching tests, compliance (EN 12457-4), Up-flow percolation (CEN/TS 16637-3) and pH-dependence (EN 14429). By completing this study, it can be shown that using EAFS instead of traditional aggregate in concrete can be a viable environmental choice in terms of material circularity.

2. Materials and methods

2.1. Dimensional, chemical and mineralogical properties of the EAFS

EAFS was supplied by a steelmaking facility in the north of Spain that produces high alloyed steel bars. The slag has been processed by quenching, primary metal separation, sieving, secondary metal separation, and ageing outdoors. The dimensional characterization of the aggregates has been determined in accordance with EN 933–1:2012. The chemical composition (as major oxides and trace elements content) of the slag was determined by lithium metaborate-tetraborate fusion and total digestion with inductively coupled plasma optical emission spectroscopy (ICP-OES) and Instrumental Neutron Activation Analysis (INAA) in the Actlabs facilities in Ancaster (Canada).

The X-ray diffractometry (XRD) analysis of the EAFS was performed in a Bruker D8 Advance. A Ni-filtered Cu Kα radiation was employed in a 2θ configuration, and 30 kV and 10 mA were the operating conditions. Samples were analysed in the 20 to 70° angle, with a step size of 1°/min. The X-ray powder diffraction pattern have been fitted using the pattern matching routine in the DIFFRACplus EVA program supplied by Bruker.

2.2. Concrete mix design and characteristics

Although limestone aggregate differs significantly from steel slag in terms of physical and chemical properties, it was selected as the reference material because it is the most commonly used aggregate in concrete production in Europe and the USA. Its widespread availability and frequent use in construction make it a relevant choice for comparative analysis.

The concrete mix proportions were calculated using the Fuller method, setting the same amount of cement and effective water/cement ratio (0.47) for the two mixes developed (the water from the absorption of the aggregates has been compensated). A superplasticizer (polycarboxylate ether) dosage of 1.2 % by weight of cement (wt% of CEM) has been established. Two concrete mixes have been developed (Table 1), a reference one using only natural limestone aggregate (LC) and another using a high replacement (68 %) of EAFS (SC) carrying out a replacement by volume. Both SC and LC mixtures are designed as structural concretes, meeting the necessary strength and durability criteria for use in load-bearing elements. The natural aggregate has an average density of 2.67 g/cm^3 and a water absorption of 1.5 % wt, while the EAFS have an average density of 3.83 g/cm^3 and a water absorption of 1.4 % wt. The fineness modulus of the coarse fractions of both natural aggregate and EAFS is around 6.9, while that of the 0/4 natural aggregate fraction is 2.6 and that of 0/4 EAFS is 3.5. As the fines content of the 0/4 fraction of EAFS is very low, this effect has been compensated by adding 0/4 mm limestone sand. The objective slump was 50 mm ± 20 mm, the binder used is a CEM I 52.5R and a 100 l capacity vertical axis planetary mixer has been used. The kneading methodology includes 3 min of initial kneading with the aggregates and water, 3 min of rest and 2 min of kneading incorporating the superplasticizer. The specimens were removed from the mould after 24 h and cured in a humidity chamber at 20±2 °C and 95 ± 5 % humidity for 28 days, according to EN 12390-2.

After a curing period (28 days), a physical-mechanical characterisation has been carried out, by tests that allow to determine the main properties of the structural concrete in hardened state: (i) apparent density, relative density, and saturated surface-dry density, according to the specifications of EN 12390-7: Tests on hardened concrete – Part 7: Density of hardened concrete; (ii) accessible porosity and water absorption coefficient, according to the procedures described in UNE 83980: Durability of concrete. Determination of water absorption, density, and accessible porosity in concrete; (iii) compressive strength according to EN 12390-3: Tests on hardened concrete – Part 3: Compressive strength of test specimens measured using cubic specimens of 100 mm [28]; (iv) indirect tensile strength according to EN 12390-6: Tests on hardened concrete – Part 6: Tensile splitting strength of test specimens, using standard cylindrical specimens; (v) modulus of elasticity according to EN 12390-13: Tests on hardened concrete – Part 13: Determination of secant modulus of elasticity in compression, performed on standardized cylindrical specimens; (vi) flexural strength determined using prismatic specimens measuring 100 × 100 × 400 mm; and (vii) water penetration under pressure according to EN 12390-8: Tests on hardened concrete – Part 8: Depth of penetration of water under pressure, conducted on standardized cylindrical specimens. All tests were conducted at a curing age of 28 days.

2.3. Leaching tests

Before executing the tests, all the equipment in contact with the leachant was rinsed with HNO_3 2 % wt. and washed with deionized water. Immediately after performing the leaching tests, pH and conductivity of obtained eluates were measured, filtered with a 0.45 μm membrane, acidified with analytical grade HNO_3 to pH < 2 if required, and stored at 4±2 °C for chemical analysis. Images of the different tests applied are shown in Fig. 1.

Table 1
Mix proportions of limestone concrete (LC) and EAFS-based concrete (SC).

Materials (kg·m^{-3})	LC	SC
Limestone sand 0/4 mm	943.5	774.3
Limestone coarse 6/12 mm	660.5	–
Limestone coarse 12/20 mm	283.1	–
EAFS sand 0/4 mm	–	433.8
EAFS coarse 4/8 mm	–	476.2
EAFS coarse 8/16 mm	–	716.1
Cement	342.1	342.1
Water	160.8	160.8
SP	3.4	3.4

2.3.1. Compliance leaching test (EN 12457)

Both, EAFS and the cured and moulded concrete samples were evaluated using this leaching test (see Fig. 2). The samples were crushed to a particle size lesser than 10 mm (EN 12457-4). 90 g of the material were introduced in a 1 L plastic bottle with 900 mL of deionized water, reaching a liquid-to-solid ratio (L/S) of 10 L/kg. Agitation to reach equilibrium between solid and liquid was performed during 24h in a Heidolph Reax 20 rotary shaker at 10 rpm. Tests were performed in triplicate [29].

2.3.2. Dynamic surface leaching test (DSLT, CEN/TS 16637-2)

Cubic mixtures of 100 mm were suspended in an inert nylon net and submerged in plastic vessels, and 4.8 L of deionized water as leachant were introduced, in a liquid to exposed sample surface ratio of 80 L/m^2, maintaining a separation of 20 mm from the walls of the vessel in all directions. Once the leachant was introduced and test was started, the vessel was covered in the upper part with a plastic lid to avoid external interferences. Then, replenishment of the leachant was performed 7 times from the start of the test at 0.25, 1, 2.25, 4, 9, 16 and 36 days, with a final extraction of the last eluate at 64 days since the start of the test. Tests were performed in triplicate.

2.3.3. Up-flow percolation leaching test (CEN/TS 16637-3)

Materials were crushed to a particle diameter lesser than 16 mm and compacted in a polycarbonate column of 300 mm in height and 50 mm diameter. A Watson-Marlow peristaltic pump was employed to force upwards the flow of leachant (Milli-Q water) through the voids of the compacted solid at a constant rate of 300 ± 10 mL/day. Test consists of 7 stages that corresponds to a L/S ratio fixed as a function of the material weight loaded into each column, 0.1, 0.1, 0.3, 0.5, 1, 3 and 5 L/kg, renewing the eluate collector once reached the volume required at each stage. Test was finished when the cumulative L/S ratio (i.e., sum of all eluates collected) of 10 L/kg in each column was completed. Tests were performed in duplicate.

2.3.4. pH-dependency leaching test (EN 14429)

Concrete samples were finely crushed with a jaw crusher to a particle diameter <1 mm to facilitate the chemical equilibrium state between leachant and solid. A set of pH values ranging from 2 to 12 were tested and a minimum of 8 pH values between this interval were determined for both the SC and LC mixtures. 30 g of sample were mixed with 300 mL of leachant in 500 mL plastic bottles. Immediately after, the bottles were then submitted to 48 h agitation in a Heidolph Reax 20 rotary shaker at 10 rpm.

2.4. Analytical determination of leachates

pH and conductivity of eluates were determined with a Crison GLP-22 pH-meter coupled with a Hach 50 21 T probe and XS Instruments Cond 61 conductivity meter, respectively. Concentrations of trace elements of potential concern in eluates were determined with a Thermo-Fisher Scientific Icap-Q Inductively Coupled Plasma Mass Spectrometer (ICP-MS) at the Institute of Marine Sciences of Andalusia (ICMAN-CSIC) (Spain), in accordance with ISO quality control standards. Detection limits of the apparatus were for each element ($\mu g \cdot L^{-1}$): As (0.02), Ba (0.04), Cd (0.001), Cr (0.02), Cu (0.05), Hg (0.01), Mo (0.01), Ni (0.01), Pb (0.01), Sb (0.01), Se (0.02), V (0.06) and Zn (0.26).

3. Results and discussions

3.1. Dimensional, chemical and mineralogical properties of the EAFS

The particle size distribution of EAFS (SA), as well as natural aggregates (NA), is shown in the 2.

Although there are differences induced by the crushing method itself or the differences in the nature of the aggregates, the dosage method will allow obtaining a very similar granulometric skeleton.

The chemical composition (as major oxides and trace elements content) of the slag is shown in Table 2.

The slag is mainly composed of iron, calcium, and silicate oxides, with a minor presence of magnesium, manganese and aluminium oxides. Content of Cr is particularly high, whilst other predominant trace elements are Ba, Mo, V, and Zn. The determined composition

Fig. 1. Images of the experimental developments of the leaching tests: a) Compliance leaching test (EN 12457) or pH-dependency leaching test (EN 14429), b) Dynamic Surface Leaching Test (DSLT, CEN/TS 16637-2) and c) Up-flow percolation leaching test (CEN/TS 16637-3).

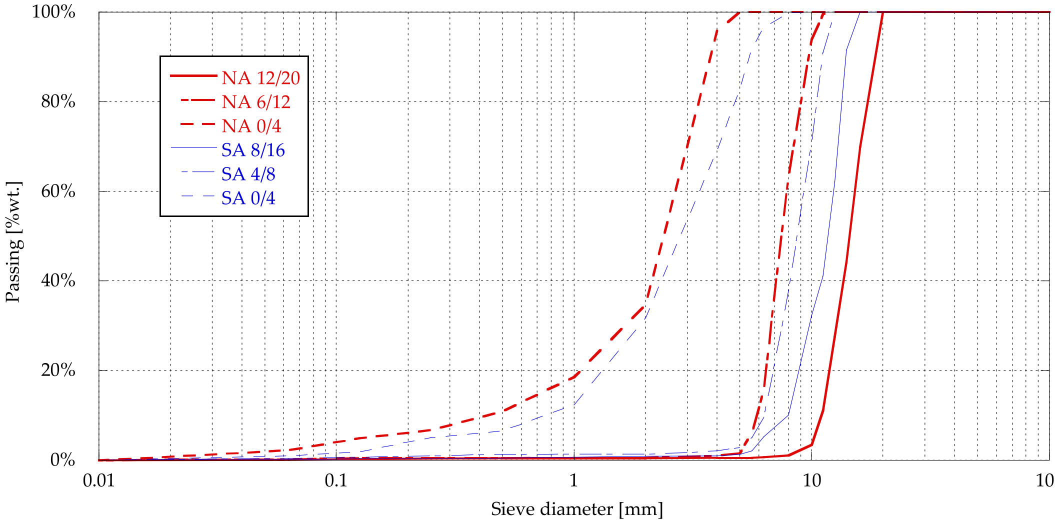

Fig. 2. Grading curves of the aggregates used.

is characteristic of EAFS from high alloyed steel production [30].

The X-ray diffractometry (XRD) analysis of the EAFS is presented in the Supplementary Material (Fig. S1). Main solid phases found in the slag are iron oxides, wustite (FeO) and magnetite (Fe_3O_4) mainly, and a high presence of magnesioferrite (Fe_2MgO_4). The presence of calcium silicates (Ca_2O_4Si), enstatite (MgO_3Si) and calcium aluminosilicate ($Ca_2Al_2SiO_7$) are also present. The magnesium oxides as haussmanite (Mn_3O_4) and chromium-rich compounds as eskolaite (Cr_2O_3) and chromite ($FeCr_2O_4$) also be found. The determined compositions and mineralogy are in agreement with those found in the literature for EAFS [31,32].

Leaching of the EAFS as an alternative aggregate is initially explored through a compliance leaching test (EN 12457-4) which describes its behaviour in chemical equilibrium with the leachant. This preliminary test is relevant to judge the release of potentially dangerous substances, especially for industrial byproducts that are characterized for the heterogeneity of its chemical composition. pH values, conductivity, and concentration of trace elements in leachates of the EAFS are shown in Table 3. A limestone used as aggregate is tested for comparison purposes, and obtained concentrations are evaluated with the regulatory thresholds of the European Waste Acceptance Criteria (WAC) for landfills specified for inert materials [33].

Values of pH and conductivity are considerably higher in the EAFS with respect to the limestone aggregate. Increased conductivity and alkaline pH values are mainly because of the dissolution of free CaO and other metal oxides [32].

The mobility of trace elements Ba, Cr, Hg, Mo, Se and V is also higher in the EAFS than in the limestone aggregate. Nonetheless it should be noted that, for Ba, Cr, Mo and V, in relation to the chemical composition of the EAFS (Table 2), low levels of trace elements are mobilized due to the retention in mineral phases [31,34].

The comparison of these values with the WAC limit values for inert materials [33] indicates that the concentrations of Hg, Mo, Se and V in the EAFS leachate are exceeded. However, in some Spanish regions, such as Catalonia, Basque Country or Cantabria, there are specific regulations on slag valorisation, where the application as an aggregate in binders, like concrete or asphalts, is not subject to leaching limits for harmful elements. This is because specific regulations assume that the binder inertizes the slag, preventing any contaminant elements from leaching [35,36]. In order to assure that there is no potential environmental risk, the leaching behaviour of

Table 2
Chemical composition of the EAFS.

Major elements	% wt.	Trace elements*	mg·kg^{-1} dry matter
SiO_2	10.18 ± 0.47	As	<D.L.
Al_2O_3	5.98 ± 0.47	Ba	872.50 ± 20.5
Fe_2O_3	45.92 ± 1.24	Cd	<D.L.
MnO	6.28 ± 0.27	Cr	17,700 ± 1414.21
MgO	6.67 ± 0.22	Cu	104.50 ± 2.12
CaO	22.47 ± 1.48	Hg	<D.L.
Na_2O	0.05 ± 0.01	Mo	147 ± 9.89
TiO_2	0.41 ± 0.002	Ni	17.50 ± 2.12
P_2O_5	0.37 ± 0.01	Pb	17 ± 1.41
SO_3	0.08 ± 0.001	Sb	0.40 ± 0.001
Loss on ignition (750 °C)	−1.85	Se	<D.L.
		V	1092.50 ± 78.49
		Zn	197.50 ± 7.78

(*)Detection Limits (D.L.) (mg·kg^{-1}): As (2), Cd (0.50), Hg (1), Se (3).

Table 3
pH values, conductivity and concentrations of contaminants in leachate from compliance leaching test (EN 12457-4) of limestone and EAFS, and limit values of WAC for inert materials.

Elements ($mg \cdot kg^{-1}$)	Limestone	EAFS	Limit values
pH	8.78 ± 0.09	11.44 ± 0.05	–
Conductivity ($\mu S \cdot cm^{-1}$)	38.17 ± 0.78	305.67 ± 12.66	–
As	0.002 ± 0.0006	0.002 ± 5.54E-05	0.5
Ba	0.010 ± 0.001	1.040 ± 0.016	20
Cd	0.00007 ± 4.53E-05	0.00028 ± 5.84E-05	0.04
Cr	0.020 ± 0.0053	0.343 ± 0.0644	0.5
Cu	<DL*	<DL*	2
Hg	0.003 ± 0.0011	**0.011 ± 0.00051**	0.01
Mo	0.150 ± 0.0511	**0.778 ± 0.0801**	0.5
Ni	<DL*	<DL*	0.4
Pb	<DL*	<DL*	0.5
Sb	0.001 ± 0.0002	0.002 ± 0.0002	0.06
Se	0.040 ± 0.0071	**0.144 ± 0.0058**	0.1
V	0.019 ± 0.0044	**3.661 ± 0.043**	1.5**
Zn	0.019 ± 0.028	0.019 ± 0.0027	4

Bold values exceed the limit. (*) Under detection limit; (**) Adopted limit value (BOPV 64/2019).

the EAFS-based concrete during the entire life cycle is studied. Elements that surpass the limits and Cr as an element with a concentration close to the limit in the EAFS are the elements evaluated in the leaching characterization of the EAFS-based concrete.

3.2. Physical-mechanical characterisation of structural concrete

The physical and mechanical properties of the samples were evaluated through a series of standardized tests after 28 days of curing. The results of these tests are summarized in Table 4.

This data indicates that SC outperforms LC in terms of mechanical properties, with higher density, compressive strength, tensile strength, and modulus of elasticity. The SC sample density exceeds 2600 kg/m^3, which is the density from which high-density concrete is conventionally considered. The porosity of both dosages is similar, moving around 7 %, considering a good value any that is below 10 %. The 10 % porosity threshold is taken as a reference and is not regulated by any specific standard. While natural aggregates have a relatively uniform porosity, EAFS exhibits a higher density and a distinct internal structure, characterized by cavernous voids rather than interconnected pores. This results in a greater ratio of absorbed water to aggregate weight but does not necessarily imply higher permeability. These morphological differences must be considered when evaluating the material's durability and mechanical performance. A compressive strength of 89.5 ± 3.2 MPa, while the control LC 62.9 ± 6.2 MPa, and a 16 % increase in tensile strength, demonstrated the technical suitability of EAFS-based mixtures in scenarios that require a high-performance concrete (e.g., structural applications) [11].

Incorporating EAFS improves the mechanical properties of concrete, resulting in higher compressive strength. The incorporation of EAFS also improves durability due to its lower water absorption and higher density compared to natural aggregates. The reduced water absorption limits internal moisture variations, minimizing shrinkage and cracking, while the higher density enhances mechanical performance and resistance to external aggressive agents. These factors contribute to a more durable concrete with lower permeability and higher long-term stability. These aspects are crucial in building structures that must withstand significant loads throughout their service life. Furthermore, this enhanced structural performance improves the long-term performance of building structures, thereby reducing the need for maintenance and repairs.

3.3. Leaching assessment of monolithic products during their service life

The release as a function of time of trace elements and the release mechanisms of the monolithic EAFS-based concrete are

Table 4
Physical-mechanical characterisation of limestone concrete (LC) and EAFS-based concrete (SC) (mean values±standard deviations are represented).

Properties	Units	LC	SC
Apparent density	Db [g/cm^3]	2.34 ± 0.03	2.93 ± 0.02
Relative density	Da [g/cm^3]	2.52 ± 0.03	3.14 ± 0.02
Saturated surface-dry density	Dssd [g/cm^3]	2.41 ± 0.03	3.05 ± 0.02
Accessible porosity	[% Vol.]	6.97 ± 0.28	6.83 ± 0.16
Water absorption coefficient,	[% wt.]	2.98 ± 0.14	2.33 ± 0.06
Compressive strength	fc [MPa]	62.90 ± 6.22	89.45 ± 3.15
Indirect tensile strength	fct [MPa]	3.54 ± 0.19	4.12 ± 0.16
Modulus of elasticity	E [GPa]	41.25 ± 0.31	53.48 ± 1.05
Flexural strength	σ [MPa]	9.47 ± 0.10	9.97 ± 0.22
Water penetration under pressure	KH_2O [mm]	41.70 ± 3.47	43.98 ± 3.36

characterized through the DSLT, CEN/TS 16637-2 [22], to simulate the scenario of exposure to water during the service life and to estimate the long-term leaching characteristics of the material. pH values, conductivity and cumulative release as a function of the exposed area (mg·m^{-2}) of concrete samples are depicted in Fig. 3.

As mentioned earlier, only elements that exceeded the limits in the EAF slag leachate, such as Hg, Mo, Se, and V, as well as Cr for being close to this limit, are studied.

pH values of both samples are similar between them. In the short-term stages (from 0.25 to 4 days) values are progressively increased due to longer contact time between leachant and concrete sample in each stage, whilst in the long-term the tendency is to stabilize around a mildly alkaline pH of approximately 8, probably due to CO_2 uptake with increased stagnant time of the leachates in the vessel during testing [37].

Conductivity is increased progressively with time for both samples, being the sharpest increment in the early stages. No major differences are found between LC and SC. Similar evolution of conductivity values have been reported in monolithic testing of concrete mixes from other works [15,38], and is mainly caused by the dissolution of Ca and alkali (i.e. Na, K) ions [39]. Stabilization of pH

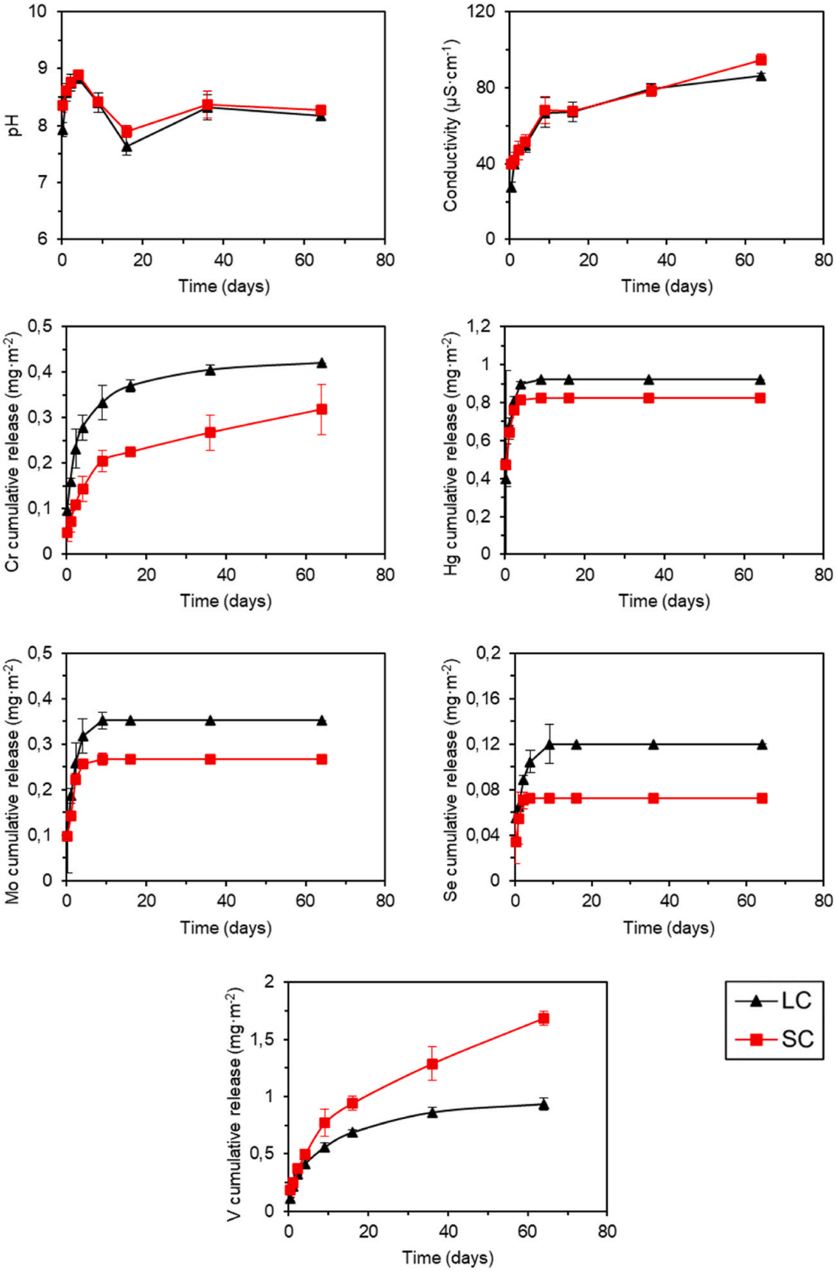

Fig. 3. Evolution of pH values, conductivity and cumulative area release over time of monolithic concretes in DSLT.

values and conductivity over time indicates that release of major chemical elements may continue in the long-term.

On the other hand, the release of trace elements has generally decreased over time. Cumulative release of Cr, Hg, Mo and Se shows increased concentrations in the short-term stages followed by stabilization in the long-term, meaning that little or no release occurs in the last stages. Same trends are found in LC and SC for these elements. Moreover, the LC release is slightly higher, and therefore in these elements there is not influence of the addition of EAFS as aggregate in concrete.

However, V release pattern in SC is different from the other trace elements, as it is not attenuated in the long-term and differs from the LC behaviour. Indeed, the released concentrations are the highest amongst the trace elements measured in SC. V is a pH-dependent oxyanion and increased concentrations in SC may be caused by the pH of the eluates [40], that promotes the mobility of V contained in the EAFS. In the mildly alkaline pH interval, between 8 and 10, a maximum on the leaching of V is found [41].

The controlling release mechanisms of monolithic concretes were derived from the experimental results and determined following the steps and calculations described in the Supplementary material of the CEN/TS 16637-2 procedure (Table 5). The method discerns two main or primary release mechanisms; diffusion or dissolution, and two secondary release mechanisms; surface wash-off and depletion, which are determined through a stepwise process, available as a flowchart in the Supplementary Material (Fig. S2).

Except for V, for the rest of the elements the primary release mechanisms have not been determined due to very low concentrations in the leachates (M1), very close to the detection limits of the ICP-MS equipment, or to unidentified control mechanisms (M4) in the case of Mo in the LC sample. A small difference in the Cr secondary release mechanism is observed, while in the LC sample a surface wash-off followed by low concentrations (M1.1) is determined, also found by Król [42], in the SC sample it is not observed. In the case of V, the determined primary release mechanism is diffusion for both concretes. However, the LC presents depletion as secondary mechanism while SC does not. This is clearly shown in Fig. 3.

The cumulative release values in DSLT at 64 days are evaluated through comparison with the Soil Quality Decree limits for moulded building materials (Table 6). In absence of harmonized criteria in the European Union, these values are used in the literature for evaluating the leaching of monolithic building materials [15].

Concentrations determined in SC are overall similar to the LC, except in V, as discussed above. It should be noted, however, that this difference is considered not significant because V and the rest of the elements remain below the limit values. Hence, the incorporation of EAFS to concrete as aggregate does not significantly influence the leaching characteristics in its monolithic form.

3.4. Leaching assessment of granular products: end-of-life compliance assessment

At the end of the service life, the demolished material can be either landfilled or reused in other applications as recycled aggregate. If disposal is the management option selected, leaching testing should be conducted to check the compliance of the granular EAFS-based concrete with the imposed leaching criteria. All leaching tests were performed on newly crushed concrete, without prior exposure to leachate, in order to simulate a worst case scenario for potential contaminant release. The compliance leaching test (EN 12457-4) is performed to the SC and LC (Table 7), and results are compared with the European Waste Acceptance Criteria limit values for disposal of inert materials.

pH values of granular concretes are strongly alkaline, and conductivity is slightly higher in SC compared to LC. Compared to the mobility of the EAFS (Table 3), the release of elements in SC, such as Mo and V is significantly reduced. All the trace elements comply with the limits considered, with the exception of Cr, which surpass by little the limit value of 0.5. However, the LC also exceeds the limit value, which confirms that the source of mobilized Cr is the cement and not the EAFS. These results are in agreement with the leaching behaviour of (granular) recycled concrete aggregates in the literature, that present concentrations of Cr close to the limit of 0.5 mg kg^{-1} [44,45].

3.5. Valorisation stage leaching assessment of granular products

On the other hand, the complexity of scenarios that the material can be subjected to in a material circularity context increases when

Table 5
Controlling release mechanisms identified of monolithic EAFS-based concrete (SC) and limestone concrete (LC) according to stepwise determination in CEN/TS 16637-2.

Monolith release mechanisms	LC					SC				
	Cr	Hg	Mo	Se	V	Cr	Hg	Mo	Se	V
M1 - Overall low concentrations	–	–	–	✓	–	✓	–	–	✓	–
M1.1 - Surface wash-off followed by low concentrations	✓	✓	–	–	–	–	✓	✓	–	–
M2 - Diffusion	–	–	–	–	–	–	–	–	–	✓
M2.1 - Surface wash-off preceding diffusion	–	–	–	–	–	–	–	–	–	–
M2.2 - Diffusion followed by depletion	–	–	–	–	✓	–	–	–	–	–
M3 - Dissolution	–	–	–	–	–	–	–	–	–	–
M4 - Unidentified controlling mechanism	–	–	–	–	–	–	–	–	–	–
M4.1 - Surface wash-off	–	–	–	–	–	–	–	–	–	–
M4.2 - Depletion	–	–	✓	–	–	–	–	–	–	–
Determined mechanism	M1.1	M1.1	M4.2	M1	M2.2	M1	M1.1	M1.1	M1	M2

Table 6

Cumulative release at 64 days of monolithic concrete samples and comparison with the Soil Quality Decree limit values.

Elements ($mg \cdot m^{-2}$)	Cr	Hg	Mo	Se	V
LC	0.42	0.92	0.35	0.12	0.93
SC	0.32	0.82	0.27	0.07	1.68
SQD limits	**120**	**1.4**	**144**	**4.8**	**320**

Table 7

pH values, conductivity and concentrations of contaminants in leachate from compliance leaching test (EN 12457-4) of limestone control concrete (LC) and EAFS-based concrete (SC), and limit values of the WAC for inert materials.

Elements ($mg \cdot kg^{-1}$)	LC	SC	Limit values
pH	12.63 ± 0.040	12.15 ± 0.064	–
Conductivity ($\mu S \cdot cm^{-1}$)	2856.67 ± 56.86	3370 ± 175.78	–
As	0.001 ± 0.00015	0.001 ± 6.7E-05	0.5
Ba	1.75 ± 0.042	3.394 ± 0.059	20
Cd	0.00007 ± 8.1E-06	0.00015 ± 4.95E-05	0.04
Cr	**0.616 ± 0.0056**	**0.515 ± 0.0083**	0.5
Cu	<DL*	<DL*	2
Hg	0.001 ± 9.47E-05	0.003 ± 0.000538	0.01
Mo	0.129 ± 0.029	0.339 ± 0.0309	0.5
Ni	<DL*	<DL*	0.4
Pb	0.003 ± 0.00066	0.004 ± 0.00051	0.5
Sb	0.003 ± 6.32E-05	0.002 ± 9.94E-05	0.06
Se	0.090 ± 0.011	0.096 ± 0.0042	0.1
V	0.041 ± 0.0035	0.068 ± 0.004	1.8**
Zn	0.026 ± 0.0022	0.023 ± 0.0051	4

Bold values exceed the limit. (*) Below detection limit (**) Adopted limit from Ref. [43].

the demolished material is employed as recycled aggregate in other applications (e.g. road bases, concrete mixes). Therefore, an evaluation of the factors that most affect the leaching characteristics of the recycled concrete is necessary. In this respect, the L/S ratio and pH are the most relevant parameters that influence the leaching of substances in granular materials [46].

3.5.1. Leaching as a function of liquid-solid ratio (percolation test)

The EAFS-based concrete as recycled aggregate could be subjected to water contact and infiltration by exposure to rainwater or seepage when left outdoors as a granular material. Characterization of the release of trace elements with the percolation leaching test (CEN/TS 16637-3) as a function of the percolated water through the material with a particle size <16 mm (L/S ratio) is useful to simulate this hypothetical scenario.

The pH values, conductivity, and cumulative release of trace elements in the up-flow percolation test are shown in Fig. 4. For comparison purposes, the behaviour of EAFS as a function of L/S is represented. Concentrations of the Equilibrium leaching test at L/S = 10 for EAFS and LC and SC are also depicted, as it is a conservative approximation to percolation testing results [47]. Although it should be pointed out that the procedure tests were carried out on materials with different particle sizes (<10 mm vs. < 16 mm) and different conditions of equilibrium.

The highly alkaline pH values in SC remain constant throughout the test. On the contrary, conductivity is exponentially decreased, indicating a stabilization of the release of major constituents with increased L/S ratio.

Cr release is slightly lower in SC than in EAFS. This slight difference can be attributed to the contribution of chromium from the cement. Hg and Mo release is also lower in comparison, with the exception of Se, increased at a longer L/S ratio. Small differences for these elements between SC and EAFS may be explained by the contribution from other components of the granular slag-based concrete such as cement or fine limestone aggregates and can also be attributed to the difference of leachates pH values, order of 2 units. Lastly, V release is severely reduced in the SC, mainly due to high pH values.

In general, except for Se, trace elements release is lower in SC compared to the EAFS, and released trace elements are similar or lower in the percolation test compared to the compliance leaching tests performed to EAFS and SC. This is due to the fact that in each test different objectives are targeted, in the compliance test equilibrium and in the column test percolation.

The cumulative release obtained at L/S ratio of 10 is evaluated with the SQD limits for granular building materials. In the EAFS, the vanadium is the only element that surpass the limits for column leaching tests at L/S = 10. All the elements in the SC remain below the limits, being considered environmentally acceptable to be employed as granular building material.

On the other hand, the release mechanisms derived from the procedure in CEN/TS 16637-3 are collected in Table 8.

It is identified the same mechanisms for EAFS and SC, except for V. For Hg and Se, the mechanism cannot be determined due to overall low concentrations (M1) (i.e. close to the detection limit) in eluates. In Cr and Mo apparent depletion (M4) is established. This means that concentrations as a function of the L/S ratio are decreasing rapidly in the long-term, but not sufficiently enough to confirm that substances will not leach out at longer L/S ratios, i.e. the wash-out (depletion) mechanism (M3) occurs. In the case of V, however, significant differences of SC compared to EAFS, lead to changes in the determined mechanism. In EAFS, apparent depletion (M4) is

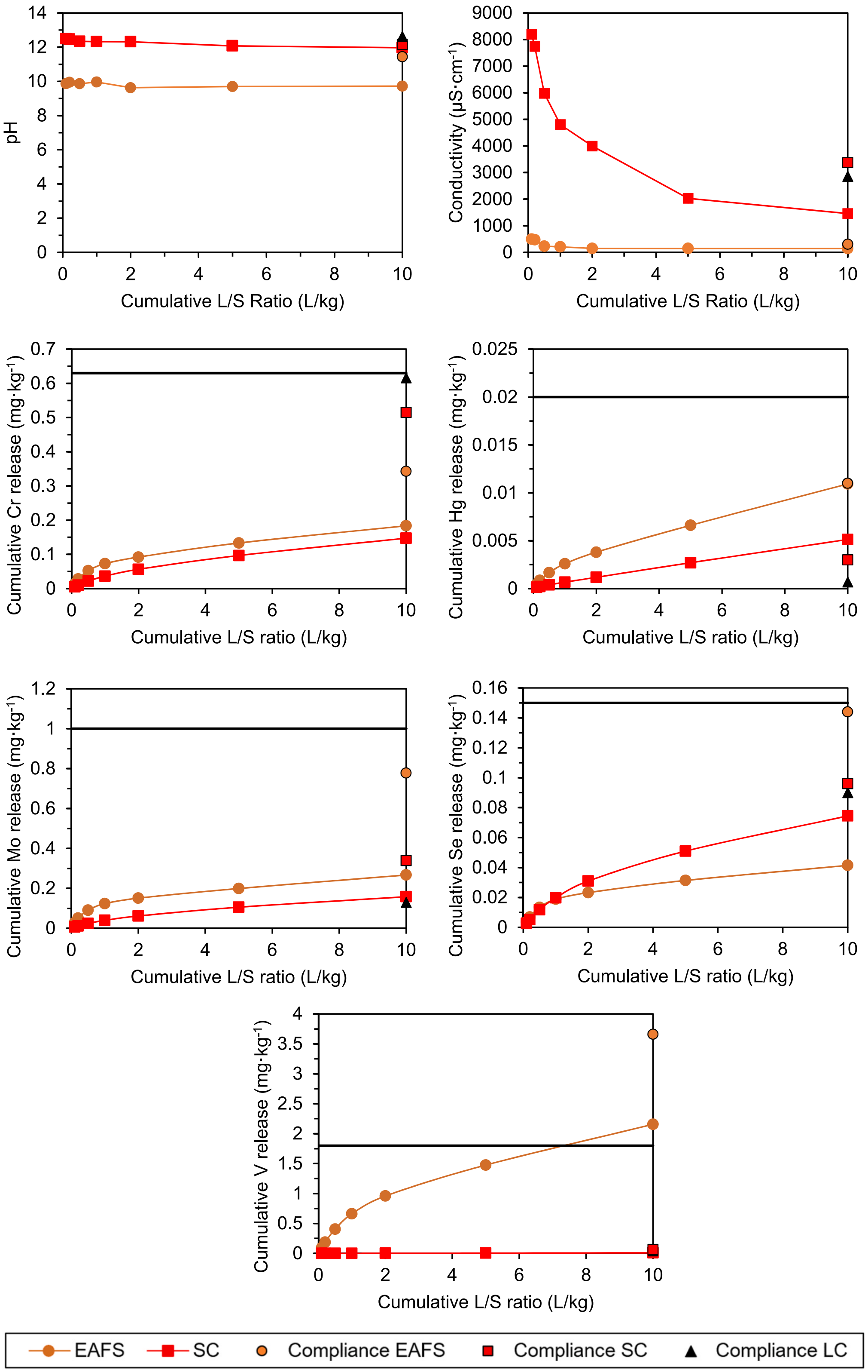

Fig. 4. Evolution of pH values, conductivity and cumulative area release over L/S ratio of SC in up-flow percolation leaching test. Solid line represents the SQD limit for granular materials.

Table 8
Controlling release mechanisms from granular EAFS-based (SC) and limestone (LC) concrete according to the calculations and procedures in CEN/TS 16637-3.

Granular release mechanisms	EAFS					SC				
	Cr	Hg	Mo	Se	V	Cr	Hg	Mo	Se	V
M1 – Overall low concentrations	–	✓	–	✓	–	–	✓	–	✓	✓
M2 – Solubility	–	–	–	–	–	–	–	–	–	–
M3 – Wash-out (Depletion)	–	–	–	–	–	–	–	–	–	–
M4 – Apparent depletion	✓	–	✓	–	✓	✓	–	✓	–	–
M5 – Unidentified mechanism	–	–	–	–	–	–	–	–	–	–
Determined mechanism	M4	M1	M4	M1	M4	M4	M1	M4	M1	M1

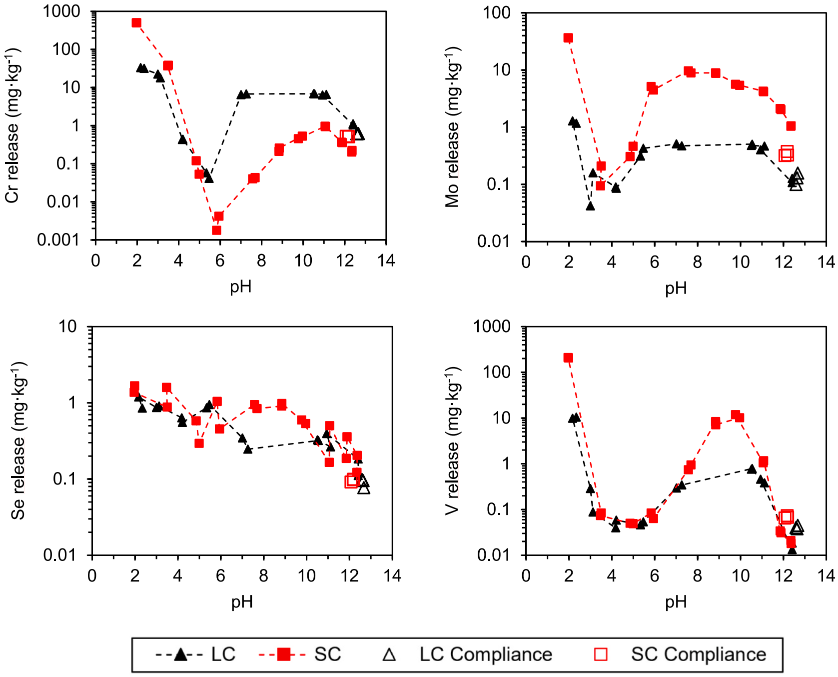

Fig. 5. Release of Cr, Mo, Se and V as a function of pH of SC and LC samples.

identified, while in SC concentrations in eluates are low enough to establish overall low concentrations (M1).

3.5.2. Leaching as a function of pH (pH dependence test)

In a context of material circularity and possibility of various reuse or recycling scenarios, knowing how the material releases substances as a function of pH is essential. Since small changes in pH values in the environment can lead to drastic changes in leaching of trace elements, which is critical when considering the long-term behaviour of construction materials. The release of trace elements through the pH-dependence test (EN 14429) [48] is depicted in Fig. 5. Compliance leaching test results are also depicted. Hg was not represented because concentrations were below detection limits of the equipment.

The release of Cr is minimal in the pH value around 6, following a "V-shaped" curve for both concrete samples. In the range of acid pH the release is slightly higher in SC, probably due to dissolution of Cr-spinels from the EAFS [34], while in the range between 6 and 12 the release of Cr in LC is higher. It can be explained by surface adsorption complexation with Fe oxides due to the high iron content in the slag, which is well established in the literature [49]. Mo presents an oxyanionic species behaviour, with a minimum pH value around 4 and a high overall release in the neutral and alkaline regions [50]. Se mobility, as opposed to the other elements, does not exhibit a strong pH-dependence and is slightly decreases towards the alkaline pH values, with no significant differences between LC and SC. The V release trend is similar on both types of concretes, both describing two minimums, in the pH range 3–6 and at high alkaline values. However, V release from SC is notably higher in the alkaline region, with a maximum around at pH above 10.

Considering that in many cases, the precipitation of oxyanions as pure metalates does not offer a satisfactory explanation for the leaching behaviour of oxyanions. It is very probably that surface adsorption and the formation of solids with common minerals in cement reduce the mobility considerably. While Cr is dominated by complexation by surface adsorption with amorphous Fe oxides (HFO), for the other oxyanions it is their incorporation into ettringite, monosulphate and hydrocalumite. It is very likely that these solid phases control the leaching of Mo, Se and V. This could explain the results obtained (Fig. 5), where in a given pH range, the leaching of these oxyanions is lower in the concrete sample with limestone as aggregate (LC).

4. Conclusions

Environmental and structural assessment of building concrete with EAFS as a replacement for traditional aggregates has been carried out by a physical-mechanical characterisation and a set of leaching tests that assess leaching at various stages of the lifecycle. Leaching as a function of time, liquid/solid ratio and pH was determined by monolithic and granular tests to form the basis for long-term term leaching behaviour at the service and potentially valorisation stages.

The environmental assessment of steel aggregates, EAFS, according to European regulations, identifies Cr, Hg, Mo, Se and V, as trace elements of potential hazard for use as a secondary resource. In the service life stage, the results from the monolith leaching test (DSLT) show that the use of EAFS as aggregate does not have a significant impact based on cumulative release of trace elements, except for V, with the "diffusion" as the controlling release mechanism. In a potential second life, the application of crushed concrete as a granular construction material was assessed. Firstly, by the Compliance leaching test, which showed that both concretes (LC and SC) behave inertly according to the European regulation for use as a secondary resource. Secondly, by the percolation leaching test, which showed an overall much lower release of trace elements in SC compared to EAFS, with "apparent depletion" predominating as a control mechanism. The pH-dependent leaching patterns of the studied oxyanions are similar for both concretes, but a significant difference in mobility is observed in the determined pH range. This is likely due to the fact that surface adsorption with Fe oxides reduces the Cr release in the EAFS-based concrete (SC), while the solid solution formation with the major minerals of the cement matrix reduces the Mo and V release in the limestone concrete (LC).

It should be noted that the release of all elements of environmental concern from both concretes tested met the leaching criteria proposed by the Dutch SQD for unrestricted use of construction products, below 10 % and 30 % of the limit values for monolithic and granular forms, respectively. Thus, the replacement of traditional aggregate by EAFS is a valid environmental option from a materials circularity perspective.

In a context of circularity, where construction products can be subject to physical and chemical changes and exposed to different environments throughout their life cycle, it is recommended that a battery of leaching tests should be used for environmental assessment. In addition to the technical advantages, using EAFS as a substitute for natural aggregates in building construction can reduce costs, contribute to sustainable construction and also contribute to the basis of a pre-normative study for the manufacture of structural concretes with high percentages of steel aggregates.

CRediT authorship contribution statement

I. Salas: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **P. Tamayo:** Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **E. Cifrián:** Writing – review & editing, Supervision, Project administration, Methodology, Investigation, Formal analysis, Conceptualization. **C. Thomas:** Writing – review & editing, Supervision, Project administration, Investigation, Formal analysis, Conceptualization. **A. Andrés:** Writing – review & editing, Resources, Project administration, Methodology, Investigation, Formal analysis, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jobe.2025.112993.

Data availability

Data will be made available on request.

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