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Phosphogypsum leachate cleaning waste as partial cement replacement in mortars

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ABSTRACT

This study presents the first valorisation of a waste generated from the cleaning process of phosphogypsum leachate. Leachate decontamination waste (LDW), which contains non-negligible concentrations of heavy metals, radionuclides and fluorine, was used as a partial cement replacement in mortars at varying proportions. New cements and mortars were produced from mixtures of a commercial Portland cement (CEM I 52.5) with different concentrations of this waste (2.5, 5, 7.5, 10, 25 and 35 wt.%).

The physical-chemical properties, mechanical behaviour, and environmental impact of these LDW-cement mortars were evaluated. The results revealed that incorporating LDW at levels of up to 10% not only maintained but even enhanced the mechanical properties compared to the reference mortar. Furthermore, the cement matrix effectively immobilized potential contaminants in all cases, contributing to environmental safety.

1. Introduction

Global waste generation was estimated at around $20{\cdot}10^9$ t in 2017, with projections indicating an increase to $28{\cdot}10^9$ t by 2030 and further to $46{\cdot}10^9$ t by 2050 [1]. The vast majority of these wastes are industrial solid wastes [2]. The management and reduction of their generation is necessary to avoid serious environmental impacts.

Phosphogypsum (PG) is an industrial by-product of the fertiliser industry derived from the production of phosphoric acid, producing about 300 million tons yr^{-1} [3,4]. PG is usually stacked worldwide in open stacks [5,6], generating leachates due to the exposure to weathering conditions. Phosphogypsum leachates (PGL) are characterised by high acidity ($pH < 2$) and high concentrations of phosphates, sulphates, fluorides, metalloids and radionuclides that exceed those found in seawater by several orders of magnitude, especially U-isotopes and ^{210}Po, whose activity concentrations are over 4 orders of magnitude higher than those found in unpolluted sea waters [7,8]. Thus, PGL can cause serious environmental pollution and ecological damage if it is not handled properly [9], even more than phosphogypsum itself [10].

In previous studies, we have conducted a treatment for the decontamination of PGL by using two alkaline reagents added in two sequential separate steps ($CaCO_3$ and $Ca(OH)_2$) [11,12]. An unpolluted liquid effluent and two solid wastes were obtained after decontamination (one waste per step). The first solid was composed of 37–55 % calcium fluoride (CaF_2), while the second solid was mainly hydroxyapatite (80 %) ($Ca_5(PO_4)_3OH$) [7].

This work is focused on the first waste generated in the PGL decontamination process: leachate decontamination waste (LDW). This solid contains non-negligible concentrations of potential hazardous substances such as heavy metals, fluoride and radionuclides [7]. Therefore, it is necessary to carry out a valorisation considering this information.

Sustainable stabilisation/solidification has been commonly adopted for hazardous waste control in recent years [13,14]. Ordinary Portland Cement (OPC) is one of the most popular binders used for this purpose, since toxic elements are often immobilised and settled in the cement hydration products in hydroxide or complex forms through reactions with cement [15,16].

Wastes materials such as phosphogypsum [17], urban wastes [18], thermoset plastic waste [19], used engine oil [20], fly ash [21] or red mud [22] as building materials replacement has been widely studied. Fluorine-containing sludge produced in the manufacturing of solar cells has been used as substitute of Portland cement [23]. Additionally,

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fluorapatite ($Ca_5(PO_4)_3F$), a mineral containing fluoride, has been analysed in several applications, as additive in temperature reduction in the manufacture of clinker [24,25], and its influence on the mineral formation and final composition of cement clinker has also been explored [26,27].

Unlike PG and other industrial residues, LDW is not generated directly by industrial activity, but rather results from the remediation of contaminated leachates originating from an industrial by-product. This distinction is essential, as the environmental behaviour and potential reuse applications of LDW remain unexplored in the scientific literature.

Taking into account the above mentioned, the main objective of this work is to assess the feasibility of incorporating LDW into cement mortars by analysing its performance from technological, mineralogical, and environmental perspectives. The central hypothesis proposed is that, despite its complex composition—including elevated levels of fluoride, trace elements and radionuclides—LDW can be effectively incorporated into cement matrices without compromising material integrity or environmental compliance.To the best of our knowledge, no previous studies have investigated a residue coming from the treatment of PGL as a separate waste potentially valorisable..

2. Materials and methods

2.1. Raw materials

The raw materials used to prepare the cement mortars specimens were cement (CEM I 52.5), water, sand and LDW obtained from PGL collected at the phosphogypsum stockpiles located in Huelva (Southwestern Spain).

2.1.1. Cement – ordinary Portland cement (OPC)

In this study, CEM I 52.5 was used. This cement does not contain mineral additions. The water used was drinking water, and CEN Standard Sand was employed. The chemical composition of the cement used is shown in the Supplementary Material (Table S1).

2.1.2. Leachate decontamination waste (LDW)

The waste used in this study was the solid generated in the first step of the decontamination process of PGL by adding $CaCO_3$ up to pH = 3.5. This process is thoroughly detailed in our previous paper [7]. This cleaning process produces another waste consisting mainly of calcium phosphates, and we are currently studying its viability in the production of raw material for the manufacture of phosphoric acid.

The main components of LDW are F, P and Ca (Table 1). These elements constitute 64 % of the chemical composition of LDW, and they are in the form of fluorite (CaF_2) and amorphous phase [7]. LDW also contains potential contaminants such as heavy metals (As, Cd, Cr, Cu, Ni, Pb and Zn) and natural radionuclides (^{210}Po, ^{238}U,^{210}Pb) (Table 1). In addition, major and minor elements were compared with the composition of the continental crust [28] (Table 1), revealing that Ca, F, P, S, As, Cd, Cr, Cu, V, and Zn have higher concentrations (16–800 times more) than in the continental crust (CC). Furthermore, comparing the natural radionuclides (^{210}Po, ^{238}U,^{210}Pb) with an unperturbed soil (35 Bq kg^{-1} for ^{238}U and ^{232}Th in secular equilibrium with their daughters), it is observed that the activity concentrations are 140 times higher for ^{210}Po and ^{210}Pb and 225 times higher for ^{238}U [29]. It is important to note that LDW only contains 60 Bq kg^{-1} of ^{40}K, which is an activity concentration much lower than that found in unperturbed soil (500 Bq kg^{-1}) [29]. These concentrations were expected, since LDW comes from the treatment of leachates from a waste (phosphogypsum) of a NORM (Naturally Occurring Radioactive Material) industry [30].

The pozzolanic activity of LDW was evaluated by the Frattini test at 8 and 15 days according to EN 196–5:2011 [31]. This test consists in evaluating the amount of calcium ion capable of saturating a solution of the same alkalinity. As an additional method to Frattini, the compressive strength of the material (Strength Activity Index) has also been evaluated.

Table 1
Chemical properties of LDW and concentration coefficient between LDW and continental crust (CC) [28].

Properties	LDW	
Major elements	(%)	LDW/CC
Al	0.35 ± 0.01	0.041
Ca	42.4 ± 0.6	16.5
Cl	0.15 ± 0.01	4.05
F	12.9 ± 0.1	230
Fe	1.31 ± 0.01	0.33
K	0.23 ± 0.02	0.10
Mg	0.67 ± 0.04	0.45
Na	0.94 ± 0.19	0.39
P	9.08 ± 0.03	130
S	1.24 ± 0.09	200
Si	0.02 ± 0.01	$6.4 \cdot 10^{-4}$
Minor elements	(mg kg^{-1})	LDW/CC
As	320 ± 10	67
Cd	72 ± 4	800
Cr	4200 ± 80	46
Cu	460 ± 40	16
Ni	6 ± 1	0.13
Pb	44.1 ± 0.3	2.59
V	4100 ± 80	45
Zn	2600 ± 300	38.8
Radionuclides (Bq kg^{-1})		
^{210}Po	5120 ± 80	
^{238}U	7900 ± 600	
^{232}Th	< 0.4	
^{210}Pb	5000 ± 110	
^{226}Ra	39 ± 3	
^{228}Ra	< 20	
^{228}Th	< 5	
^{40}K	60 ± 10	

The material is considered to be pozzolanic when the concentration of calcium ions is represented below the $Ca(OH)_2$–[OH$^-$] saturation curve (solubility curve of CaO at 40 °C) (Fig. 1) confirming the presence of pozzolanic activity according to Frattini test. This experimental result constitutes a recognized normative criterion for classifying materials as pozzolanic in cement additions.

High pozzolanicity values of LDW were observed at both 8 and 15 days. The high pozzolanicity of LDW was mainly due to its high Ca content. It was observed that LDW exhibited high pozzolanicity compared to common supplementary cementing materials, such as fly ash (FA) and silica fume (SF) (Fig. 1). Although the elemental chemical analysis (Table 1) shows a low overall content of Si and Al (0.37 %), compared to Si and Al contents of 25.7 % in FA samples and 35.7 % in SF samples [32], this parameter alone is not decisive for establishing reactivity [33]. Several studies have demonstrated that pozzolanic activity depends mainly on the amorphous/glassy fraction, which must be high—an aspect shown in previous studies carried out by the authors [7], where LDW presented 45 % amorphous phase content. In Table 1 it can be observed that the major components are Ca (42 %), P (12 %) and F (12.9 %), and a minority of Fe (1.31 %). These elements are not amorphous by itself, however, as discussed in the previous study [7], the Ca that is not in the form of CaF_2, which is highly crystalline, should be in the form of amorphous calcium phosphates among with Fe, Al, Na and Mg that are also in the amorphous fraction. In addition, pozzolanic activity is influenced by material fineness and the solubility of reactive compounds rather than by the total oxide content [34,35]. In this regard, even small amounts of amorphous silica or alumina can exhibit high reactivity with $Ca(OH)_2$ in an alkaline medium.

Regarding the presence of F and P (Table 1), previous authors have shown that a high percentage it limits the pozzolanic activation [36]. However, this reduction in pozzolanicity is observed in materials with a phosphorus content higher than 30 % [37], mainly affecting the internal microstructure at early ages; nevertheless, in the long term it can

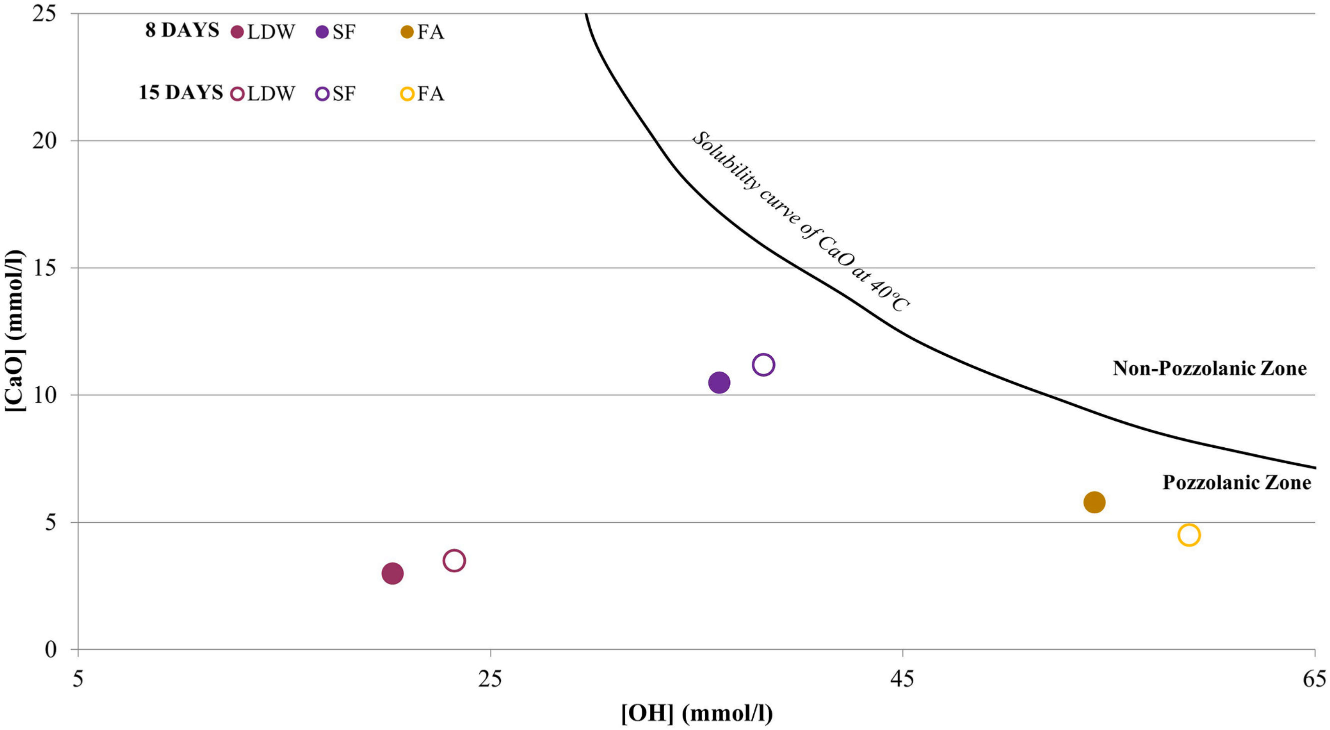

Fig. 1. Results of Frattini's test at 8 and 14 days. LDW: phosphogypsum leachate decontamination waste; SF: silica fume; FA: fly ash.

enhance the mechanical strength of the cement paste. Finally, the sulphur content in LDW (Table 1) could be beneficial in small amounts [38], but in excess it generates expansion and loss of durability due to ettringite formation. The sample showed values close to 4 % SO_3, a relatively low percentage that enhances the pozzolanicity of the material without affecting paste durability. With reduced SO_3 values, there is usually sufficient gypsum to adequately regulate initial ettringite formation and to increase the solubility of $Ca(OH)_2$, since the presence of sulphates promotes ionic mobility in the medium, which can accelerate the dissolution of the amorphous Si and Al rich phases of the cement. This improves the secondary pozzolanic reaction, which depends on the availability of $Ca(OH)_2$.

The release of aluminates, silicates and Ca^{2+} in hydration reactions in residues of similar origin to the analysed waste [22] showed the generation of a large amount of ettringite and the acceleration of pozzolanic reactions. The presence of these elements usually leads to the formation of C-(A)-S-H and ettringite, which produce a uniform and compact structure, thus improving strength. It was observed that the particle size distribution of LDW was similar to that of the cement used in the study, Ordinary Portland Cement, CEM I 52.5 (OPC). (Fig. 2).

In addition to the chemical composition of the material, which is essential to evaluate its pozzolanicity, we also considered the granulometry of the material (using a Mastersizer 2000). An adequate fineness leads to an improvement of the pozzolanic properties and to a higher specific surface area, thus increasing the specific surface area and, consequently, the pozzolanic activity [39,40]. The higher LDW

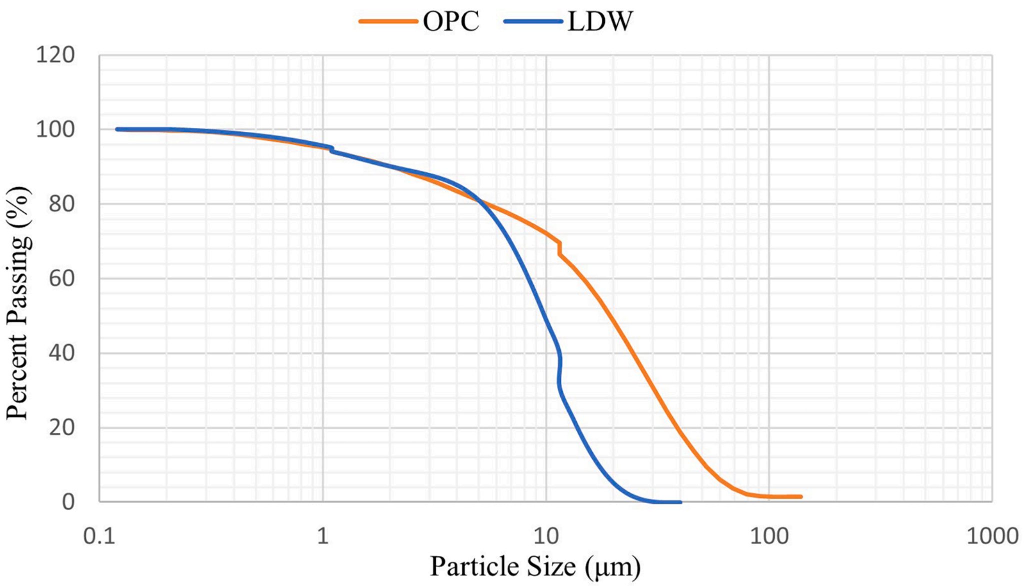

Fig. 2. Particle size distribution of LDW.

fineness resulted in higher pozzolanicity (Fig. 2). To understand the internal microstructure of the new cements manufactured, it would be useful to perform MIP or SEM tests to evaluate the formation of new gels and structures.

2.2. Sample preparation

Samples were prepared by replacing cement (CEM I 52.5) with different concentrations of LDW (2.5, 5, 7.5, 10, 25 and 35 %) (Table S2). In this study, a control mortar made with CEM I 52.5 (C) and a reference mortar made with CEM II 42.5 A/L (R) were analysed. In order to compare the properties of these mortars with a mortar made without cement replacement, this reference sample was prepared with CEM II 42.5 A/L. To ensure a correct comparison, this cement was selected based on its clinker content (80–94 %). This proportion is similar to that of the cement manufactured with LDW.

Mortar mixtures were prepared using the optimum water/binder (w/b) ratio to obtain normal consistency and good workability, as established by the flow table test in accordance with UNE 83–811–92 [41]. The mixtures were then poured into steel moulds to obtain prismatic test specimens measuring 40 × 40 × 160 mm, following the procedure outlined in EN 196–1 [42].

2.3. Analytical methods

(1) Technological tests were conducted to evaluate the mechanical properties of the cement mortars: flexural and compressive strength. Both mechanical properties are considered quality indicators to evaluate the potential use of different waste types in new cements [43]. These tests were carried out at 1, 7 and 28 days of curing time following regulation UNE-EN 196–1. For the technological tests, a universal electromechanical testing machine (IBERTEST ELECTROTEST-300-MD2) was used, equipped with two load cells: 50 kN and 300 kN.

(2) A mineralogical study of the manufactured mixtures (curing time 28 days) was performed by X-ray diffraction (XRD) using a Bruker D8advance- A25. The XRD measurement conditions were: 2θ of 5–90°; step size = 0.03°; $t = 0.03$ s; tube conditions: 40KV and 40 mA. The XRD analysis was carried out using Diffrac.eva software.

(3) Environmental risks of the manufactured mortars were carried out with the monolithic leaching test, following the EN 16,637–2 procedure [44]. This test is used to determine, as a function of time, the release of substances from a monolithic product with a leaching liquid in contact with its surface. According to the harmonised standard, the specimens were subsequently leached for a total of 64 days. During these 64 days, 8 fractions took place (after 6 h (fraction 1), 1 d (fraction 2), 2 d 6 h (fraction 3), 4 d (fraction 4), 9 d (fraction 5), 16 d (fraction 6), 36 d (fraction 7), and 64 d (fraction 8)). Eluate samples were taken at each leaching step and consequently analysed on the constituents of interest. The leaching test was carried out at room temperature. Results were reported as normalised concentrations by dividing the eluate concentrations of each step by the averaged concentration of all steps. These normalized concentrations were plotted as a column diagram. The raw data can be found in the Supplementary Material (Table S3). The incremental releases in mg m^{-2} using Eq. (1) and cumulative release (ε_{64}) in mg m^{-2} were calculated following EN 16,637–2 (Eq. (2))

$$r_i = \frac{c_i \times V}{A} \times 0.001 \tag{1}$$

$$R_n = \sum_{i=1}^{n} r_i \tag{2}$$

Where r_i is the area-related release of a substance in period i ($i = 1$ to 8) in mg m^{-2}; c_i is the concentration of the substance in eluate I, in μg l^{-1}; V is the volume of the leachant, in L; and A is the area of the test portion, in m^2; R_n is the cumulative area-related release of the substance for period n, including fraction $i = 1$ to n, in mg m^{-2}.

For the calculation of the cumulative release, concentrations reported to be below the limit of detection (LOD) were taken as equal to LOD.

In addition, the influence of the leaching behaviour of the LDW with initial acid/base addition was carried out following the normative EN 14,429 [45]. The raw data can be found in the Supplementary Material (Table S4).

The concentration of heavy metals was determined by inductively coupled plasma mass spectrometry (ICP–MS) (Agilent 7700) in the CIDERTA laboratory at the University of Huelva. The anion concentration was measured by ion chromatography (IC) (883 Basic IC plus, Metrohm) in the CIDERTA laboratory at the University of Huelva.

For the measurements of alpha emitters, alpha-particle spectrometry was conducted using an EG&ORTEC system equipped with an integrated Octete PC PLUS. Data acquisition and analysis were carried out using AlphaAnalyst software. The tributyl phosphate (TBP) sequential separation method was used [46]. Subsequently, Po was self-deposited onto silver discs for 6 h at room temperature, and U and Th isotopes were electrodeposited onto stainless steel discs at pH = 2, for 1 h and also at room temperature [47]. The samples were measured for 48 h to obtain sufficient counts. This method was validated in a previous work [3].

Regarding the measurements of gamma emitters, an extended range (XtRa) high-purity germanium detector (model GX3519, Canberra) was used (energy range: 0 to 3 MeV). The relative efficiency was 38.4 % at 1332 keV (60Co) in relation to a 3″ × 3″ NaI (Tl) detector, a full width at half maximum (FWHM) of 1.74 keV and 0.88 keV at 1332 keV (60Co) and 122 keV (57Co), respectively, and a peak-to-Compton ratio of 67.5:1. All samples were analysed for 48 h.

For both spectrometric techniques, blanks, replicas and CRMs were used to make a quality control of the results. The radiological implications were studied according to index *I*.

Standard deviations were determined for all analyses. Additionally, the error bars shown in the relevant figures represent the standard error of the mean, calculated as the standard deviation divided by the square root of the sample size. For the statistical significance analysis, Student's *t*-test was used.

3. Results and discussion

3.1. Technological test. compressive and flexural strength

Fig. 3 shows the compressive and flexural strength results of the formulated mortars and reference cement. Both compressive and flexural strengths increased with curing time. From 2.5 % to 10 % replacement, the flexural strength of the formulated mortars showed no significant differences to that of the reference cement (R) (values from 8.4 to 9.1 MPa at 28 days). Also from 2.5 % to 10 % replacement, the compressive strength of the formulated mortars was about 30 % higher than that of the reference cement (R). Among the tested samples, LDW7.5 and LDW35 showed the highest and lowest flexural and compressive strengths, with compressive strength values of 58.6 MPa and 25.7 MPa, respectively, representing a decrease of 56 %.

As shown in Fig. 3, replacements of up to 10 % cement with LDW led to an increase in compressive strength of 19–23 % compared to the reference cement (R) at all ages tested. From 10 % replacement, the flexural and compressive strength values decreased progressively, reducing to values lower than those obtained with the reference cement (R). Specifically, compressive strength values were 14 % and 43 % lower for LDW25 and LDW35, respectively. The compressive strength values showed no significant differences compared to those of the control cement up to a 10 % replacement, remaining above the requirements

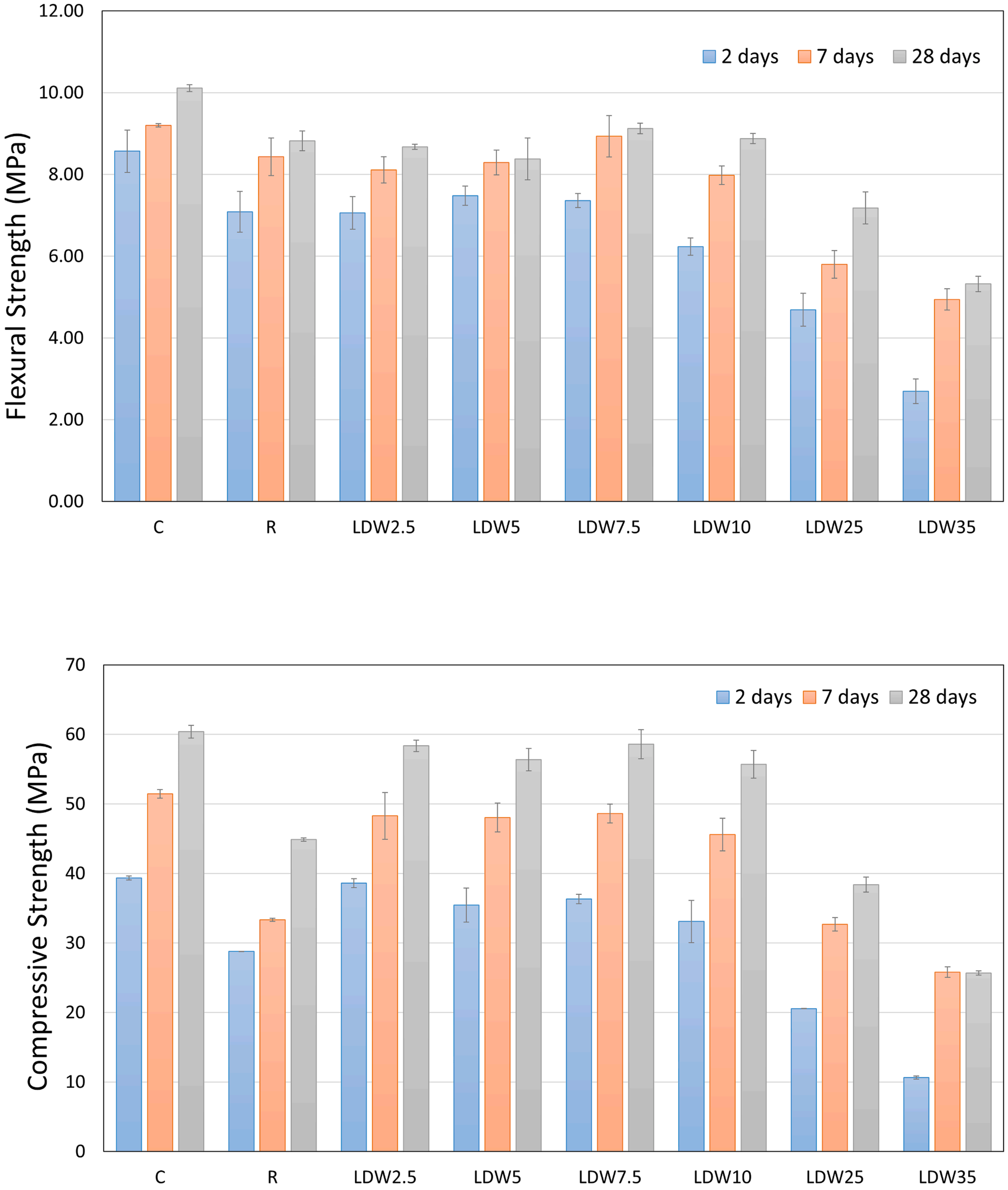

Fig. 3. Flexural and Compressive Strength (MPa) of the formulated mortars and control (C) and reference cement (R).

established by the EN 196–1 standard for 52.5 cement: ≥ 30.0 MPa at 2 days, and ≥ 52.5 MPa at 28 days.

Based on the obtained mechanical properties, LDW could be used as cement replacement, reducing the use of raw materials by up to 10 %.

The combination of the use of PG with $CaCO_3$ has been previously studied by other authors. Lin et al. [48] showed that the combination of PG with $CaCO_3$ increased the compressive strength by 15 % over a control mortar. One reason for the increase in the compressive strength of the combination of both materials can be the reduction of the porosity of the matrix [49]. The formation of microcrystals takes place, which accelerates the hydration processes and improves the mechanical properties of cement mortars [50,51].

LDW showed a high calcium fluoride content (Table 1). The feasibility of using this component as a partial cement substitute has been proved, with previous studies corroborating the obtained results. For instance, Lin, W. T [52]. showed that it was possible to use up to 10 % of calcium fluoride sludge as cement substitute, as it led to an improvement in mechanical properties and durability due to the formation of denser microstructures.

3.2. Mineralogical characterisation

Fig. 4 shows the diffractograms of LDW-cement mortars. The crystalline phases identified were quartz (SiO_2), calcite ($CaCO_3$), portlandite ($Ca(OH)_2$, ettringite ($Ca_6Al_2(SO_4)_3(OH)_{12}\cdot 26H_2O$) and phurcalite ($Ca_2(UO_2)_3O_2(PO_4)_2\cdot 7H_2O$). Quartz, portlandite, ettringite and calcite are common phases present in cement mortars. In the case of phurcalite, this was expected, considering the calcium, phosphorus, and uranium composition of the residue. The presence of phosphorus in the material (Table 1) suggests that hydration reactions may lead to the formation of $Ca_3(PO_4)_2$. This effect is commonly associated with a delay in setting time and a reduction in compressive strength development. However, XRD analyses indicated that phosphorus in LDW is present in a more complex compound (phurcalite). Fig. 4 shows that the phurcalite peak appears as the proportion of replaced residue rises. The high quartz content is due to the presence of standardised sand in the manufacture of the mortars. As can be seen in Fig. 4, the presence of quartz is similar in all mortars, so the LDW content applied does not influence it. There are no visible peaks of C-S-H or C-A-H which typically contribute to the development of mechanical strength. However, the presence of ettringite suggests that its crystallization may contribute to the increase in mechanical strength, due to the volume of this compound.

3.3. Environmental risk

3.3.1. Leaching tests

One of the main objectives of this study was to evaluate the mobility of the pollutants contained in the mortars manufactured with LDW.

When cement is mixed with water, the pH increases from 6–7 to around 12. These changes in the pH can affect the release of pollutants from LDW. Therefore, a LDW leaching test with varying pH was carried out to analyse the behaviour of the potential contaminants with the influence of pH, following regulation UNE-EN 14,429 [45].

Fig. 5 shows the leaching effect of pH in the behaviour of potential contaminants present in LDW (Table 1). In general, the concentrations of the elements decrease as the pH increases, indicating that their mobility is pH-dependent. This suggests that, when the waste is incorporated into the cement mix, leaching is expected to be minimal, due to the high pH (~12) of the matrix, which promotes the precipitation and immobilisation of most contaminants.

However, in the case of V and As, leaching persists at high pH values due to the formation of soluble species (HVO_4^{2-}, AsO_4^{3-}, $HAsO_4^{2-}$) under alkaline conditions [53,54]. This means that these elements could still be present in the leachate of the produced mortar, highlighting the need for further evaluation of their long-term stability.

Based on the concentrations of the analysed anions, a clear dependence of their concentration on pH was observed. Specifically, phosphate and fluoride exhibited higher concentrations at lower pH levels, whereas sulphate concentrations increased as pH rose. The concentrations of phosphate and sulphate were in the range of 50–800 mg l^{-1}, while fluoride levels were in the range of 5–50 mg l^{-1}. These values fall within a range that required further research in subsequent phases of the study to evaluate potential environmental risks.

To complement these findings, a leaching test on the mortars was conducted following the EN 16,637–2 standard. This test provides further insight into the actual release behaviour of contaminants from the cementitious matrix, ensuring a more comprehensive assessment of the material's environmental performance. Additionally, prediction of the long-term release of dangerous substances is important with respect to the evaluation of the release behaviour of a construction product during its service life in a construction. In order to be able to predict the long-term release by extrapolation, it is necessary to determine its release mechanism [44]. These tests were conducted for LDW10, LDW25, and LDW35, as they contain the highest waste replacement. These replacement levels were explored to demonstrate the potential of these mortars for sustainable construction applications, offering a significant reduction in cement use and environmental impact while maintaining acceptable mechanical performance.

The results showed that most of the trace elements (As, Ni, Cd, Cu, Pb, Th and U) were under the limit of detection (< 0.04 mg m^{-2}) in all

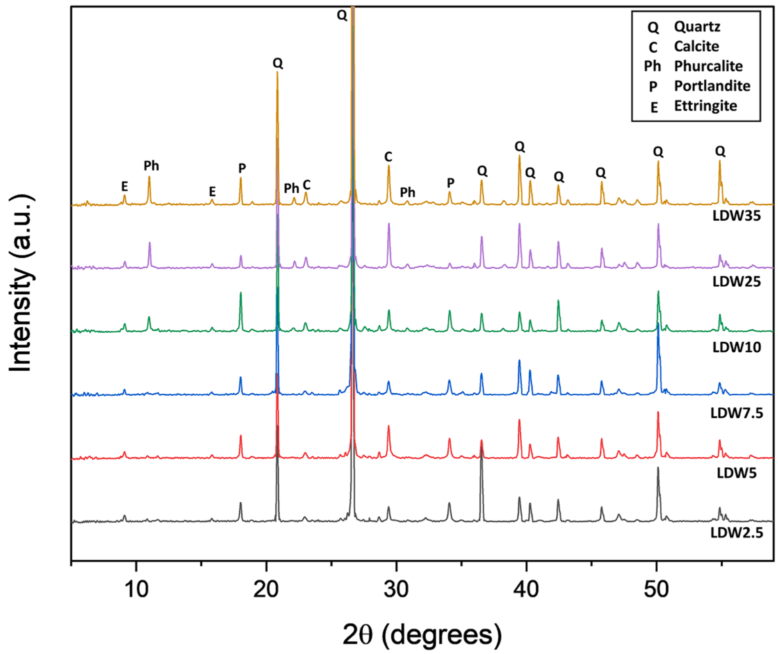

Fig. 4. X-Ray diffractograms of the manufactured cement mortars at 28 days.

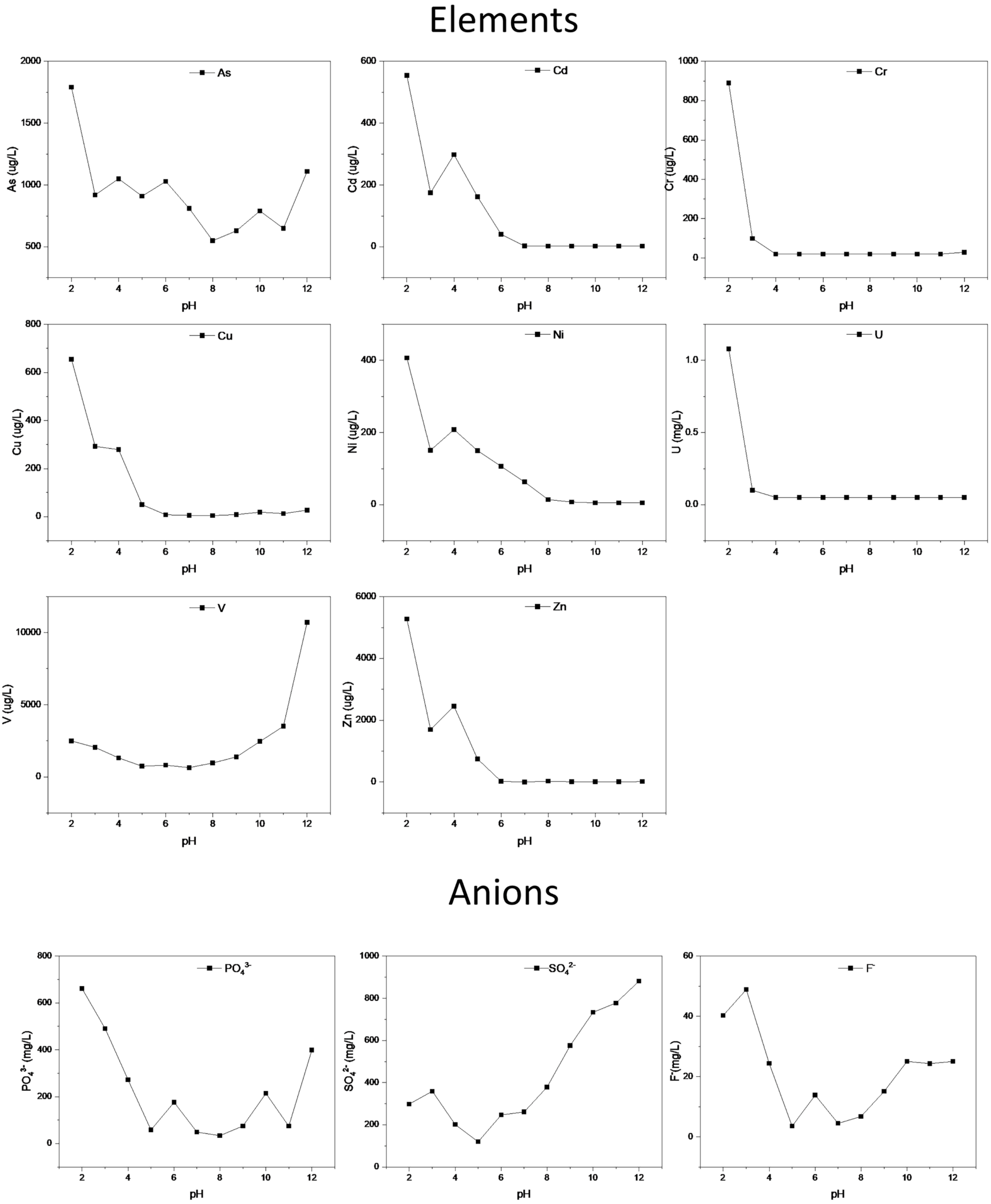

Fig. 5. Effect of pH on element concentration in LDW.

fractions of the test. The results also showed values < 40 mg m^{-2} for phosphates in all fractions.

Only five potential contaminants were detected in the leachates: Cr, V, Zn, F^- and SO_4^{2-}. Normalised concentrations of these compounds are shown in Fig. 6. Following the EN 16,637–2 standard (Annex B, assessment of release mechanisms), the different release mechanisms (dissolution, diffusion, wash-off and depletion) could be identified from the normalised concentration curve shapes. It has been reported that surface wash-off, matrix dissolution, and diffusion are the three main mechanisms of contaminant release from concrete monoliths [55,56]. In

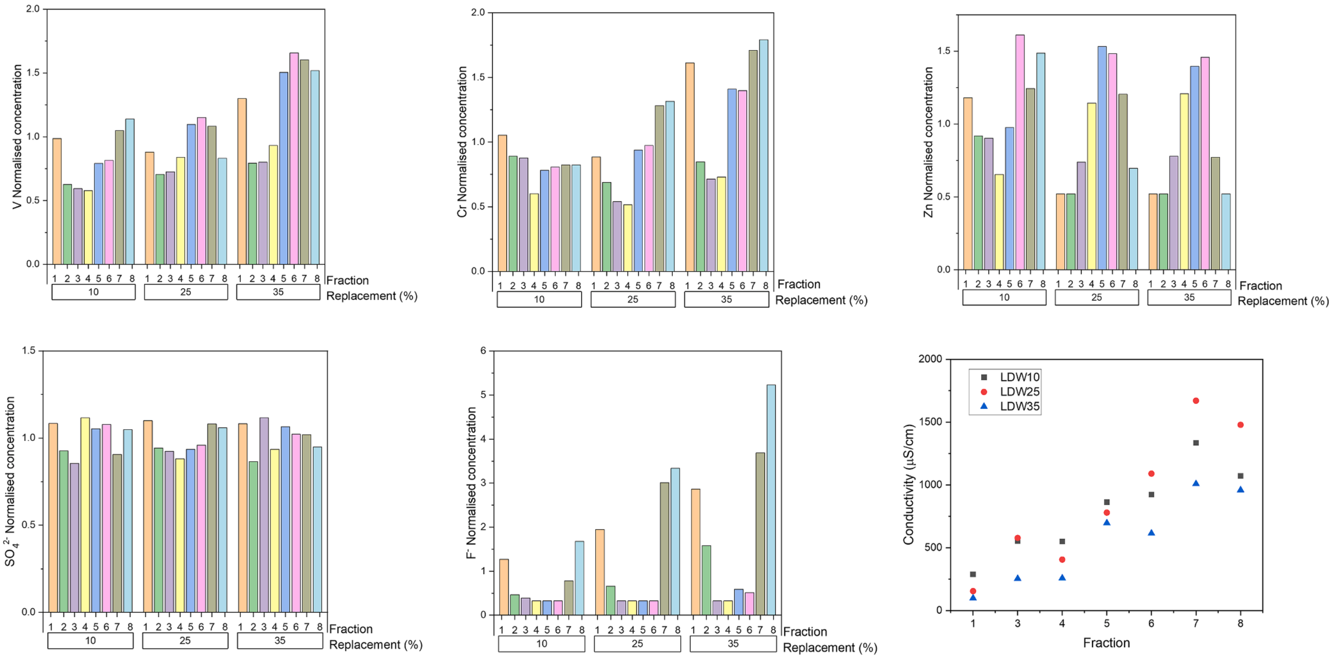

Fig. 6. Normalised concentration and conductivity (µS cm^{-1}) of the leaching test (EN 16,637–2) fractions (1–8) (concentration of V, Cr, Zn, F^- and SO_4^{2-} was < 0.04 mg m^{-2} in all fractions in the reference mortar).

this study, the following mechanisms of release were detected: V and Cr (surface wash-off, followed by diffusion); Zn and F^- (unidentified); SO_4^{2-} (dissolution).

Fig. 6 also shows the conductivity variations over time. An increasing curve was observed over a long time, which was related to the diffusion/dissolution of chemical species into the leachate.

It was expected that V, F^- and SO_4^{2-} could be leached, as suggested by the leaching study of the previously discussed residue. On the other hand, although Zn and Cr are not favoured for leaching under the given pH conditions, they were still detected in the leachate. Additionally, while As was leached in the pH-dependent leaching study, it remained below the detection limit in the leachates from LDW-cement, indicating its successful immobilisation within the matrix.

Regarding the immobilisation mechanism, physical encapsulation and chemical reactions are main mechanisms for heavy metal immobilization in cementitious materials [57]. Chemically, metal ions such as Pb^{2+}, CrO_4^{2-} and Zn^{2+} can substitute into C–S–H phases or replace ions in ettringite [58,59], and may also form low-solubility hydroxide precipitates under high pH conditions.

Specifically, Liu et al. [60] studied the vanadium immobilisation mechanism from spent SCR catalyst in cement with sulfurizing agent, concluding that it could be immobilised through formation of VO_4^{3-}-bearing compounds in hydrating cement matrix. Li et al. [59] investigated the immobilisation of arsenic within a cement matrix, observing that this process occurred through two distinct phases. Firstly, physical encapsulation took place, leading to the formation of calcium arsenate precipitates during the early hydration stage (0–3 days). As the hydration process progressed (3–28 days), arsenic retention was increasingly dominated by its incorporation into ettringite, which exhibited a significantly higher immobilisation efficiency compared to the earlier mechanisms. Other studies have also reported the effective immobilisation of As in cement-based matrices, highlighting this approach as a suitable management option for the treatment of As-containing waste [61]. Regarding F^-, various Ca-bearing hydrates such as portlandite, calcium silicate hydrate (CSH), and ettringite may to remove fluoride by precipitating CaF_2 and/or adsorbing F^- ions [62].

In this study, the horizontal dynamic surface leaching test (EN 16,637–2) results were compared with the threshold values set by the Soil Quality Decree (SQD) (Table 2), which is a regulatory framework that establishes specific criteria for the (re)use of mineral materials in construction applications, particularly for "shaped materials" [63]. The SQD sets the maximum allowed release of inorganics, expressed as mg m^{-2}, after a testing period of 64 days using a tank leaching test. The cumulative leaching values at 64 days (ε_{64}) obtained for the analysed contaminants were consistently lower than the maximum allowable limits defined by the SQD (Table 2), indicating compliance with the established environmental safety standards. The ε_{64} of the LDW mortars was also compared with that of a cement mortar containing biomass fly ash as a secondary cementitious material [64]. It was observed that the only element showing a significant difference was F, which was 2 orders of magnitude higher in the LDW-mortars than in the biomass-mortars. However, as previously mentioned, the ε_{64} of F^- is below the SQD limits.

Although leaching behaviour was addressed in the present study, future research should focus on evaluating the potential risks associated with more aggressive degradation scenarios, such as wet-dry cycling, carbonation, and acid exposure, which may affect the stability and mobility of contaminants over extended periods.

3.3.2. Radiological impact

Since LDW contains significant concentrations of radionuclides, it

Table 2
Cumulative concentration of potential contaminants leached from LDW-mortar in the horizontal dynamic surface leaching test (EN 16,637–2) compared to SQD limit levels and cement mortar with 40 % wt. replacement of OPC with fly ash (40FA1) [64].

Contaminant	LDW10 ε_{64} (mg m^{-2})	LDW25 ε_{64} (mg m^{-2})	LDW35 ε_{64} (mg m^{-2})	SQD limit ε_{64} (mg m^{-2})	40FA1 ε_{64} (mg m^{-2})
V	3.1 ± 0.3	3.5 ± 0.3	4.8 ± 0.4	320	0.837
Cr	0.56 ± 0.09	0.60 ± 0.12	0.86 ± 0.14	120	1.82
Zn	3.4 ± 0.3	3.0 ± 0.3	2.8 ± 0.3	800	16
F^-	690 ± 100	1280 ± 160	1840 ± 240	2500	39
SO_4^{2-}	70,500 ± 500	68,900 ± 500	70,400 ± 500	165,000	-

was necessary to carry out a radiological characterisation of the manufactured mortars. The results of the radioactive characterisation of the LDW mortars are shown in Table 3.

The LDW showed ^{210}Po (5120 ± 80 Bq kg^{-1}), ^{210}Pb (5000 ± 110 Bq kg^{-1}) and ^{238}U (7900 ± 600 Bq kg^{-1}) concentrations above the safety threshold (1000 Bq kg^{-1}) established by the 2013/59/EURATOM Directive. Similar activity concentrations have been obtained in a previous work [7].

The activity concentration index (*I*) functions as a regulatory parameter to verify compliance with radiation safety limits, ensuring that external gamma exposure from construction materials remains within the annual threshold of 1 mSv. This requirement, stipulated in Article 75(1) of the 2013/59/EURATOM Directive, provides a standardised approach to control potential radiological risks associated with building materials. The index is derived from the following calculation (Eq. (3)):

$$I = \frac{C_{226Ra}}{300} + \frac{C_{232Th}}{200} + \frac{C_{40K}}{3000} \tag{3}$$

where C_{226Ra}, C_{232Th} and C_{40K} are the activity concentrations for ^{226}Ra, ^{232}Th and ^{40}K, respectively, expressed in Bq kg^{-1}. This index should not exceed the value of unity ($I \leq 1$) for materials used in bulk amounts such as concrete, to ensure that the additional external dose received by occupants living in buildings constructed with these materials does not exceed the reference value of 1 mSv year^{-1} [65]. Table 3 shows that index *I* is lower than 1 for all the manufactured mortars and for LDW itself, indicating that LDW is a suitable material for use as cement replacement in mortars without radiological implications.

Although LDW comes from a cleaning process of the leachates from a NORM industry waste (phosphogypsum), the *I* index is comparable to that of commercial Portland cement. This is due to the fact that ^{238}U and ^{210}Po are alpha emitters and do not affect the index. Meanwhile, ^{210}Pb is a low-energy gamma emitter, thus it has no influence on its calculation either. However, although ^{238}U, ^{210}Po and ^{210}Pb are not taken into account in the *I* index, they still may represent an environmental issue. These radionuclides could be mobilized when the material reaches the environment and then pass to waters and from here to the food chain, producing potential increases of the radiological effective doses. Then, to simulate this mobility, the activity concentrations of these elements in the liquid fractions obtained in the leaching test that was previously commented were measured by alpha and gamma spectrometry. The results were under 5 mBq l^{-1} for ^{238}U, ^{210}Po and ^{210}Pb in all samples and in all fractions. These results showed that radionuclides are immobilised in the cementation matrix. According to the results, the activity concentrations in the not only the leachates were quite low and were in the same order of magnitude as in seawater [7], but also it has been calculated the indicative dose from the Spanish regulation (Royal Decree 314/2016) [66]. The indicative dose is the estimated amount of radiation a person would receive from drinking water over a year, used as a benchmark to assess its radiological safety. Using these values (5 mBq l^{-1} for ^{238}U, ^{210}Po and ^{210}Pb), it can be concluded that the indicative dose for these leachates is < 0.1 mSv. Therefore, the environmental impact and the effective dose equivalent received by the population would be negligible.

However, while the current exposure is minimal, it would be important to consider the full life-cycle radiological impact of materials containing enhanced concentrations of naturally occurring radioactive materials (NORM). During end-of-life scenarios such as demolition, cutting, or crushing of the mortars, there would be a potential release of particles containing radionuclides. Inhalation of these particles could pose a radiological risk, particularly in enclosed or poorly ventilated environments. From a waste management perspective, Spanish regulations (Royal Decree 1217/2024) specify that materials with activity concentrations below 1000 Bq kg^{-1} of ^{238}U, ^{210}Po and ^{210}Pb may be declassified as a radioactive waste. The activity levels measured in this study (Table 3) fall below this threshold, indicating that disposal of these mortars would not require classification as radioactive waste under current national regulations. Nevertheless, these considerations highlight the need to assess both operational safety and end-of-life implications when evaluating the radiological suitability of construction materials containing NORM.

4. Conclusions

This study presents the potential use of phosphogypsum leachate cleaning waste (LDW) as partial cement replacement in mortars, assessing its pozzolanic activity, mechanical performance, leaching behaviour, and environmental impact.

LDW showed pozzolanic activity due to its high calcium content. The evaluation of the resistant activity index demonstrated an improvement in compressive strength when up to 10 % LDW was used as a cement substitute in mortars.

In the surface leaching test, only five potential contaminants (Cr, V, Zn, F$^-$, SO_4^{2-}) were detected, and the following mechanisms of release were identified: V and Cr (surface wash-off, followed by diffusion); Zn and F$^-$ (unidentified); SO_4^{2-} (dissolution). Cumulative leaching values remained below SQD regulatory limits, indicating environmental compliance. Additionally, the *I* index was < 1 in the LDW-mortars and in LDW itself. Lastly, low activity concentrations (< 5 mBq l^{-1}) of ^{238}U, ^{210}Po, and ^{210}Pb suggested minimal radiological impact.

Future studies should consider, internal microstructure study, and risk associated with more aggressive degradation and end-of-life scenarios, which may affect the stability and mobility of contaminants.

CRediT authorship contribution statement

F.J. Soto-Cruz: Writing – review & editing, Writing – original draft, Software, Methodology, Investigation, Data curation, Conceptualization. **J. Rosales:** Writing – review & editing, Writing – original draft, Supervision, Methodology, Investigation, Conceptualization. **J.P. Bolívar:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization. **I. Ramos-Lerate:** Supervision, Resources, Project administration, Funding acquisition. **Francisco Agrela:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization. **M.J. Gázquez:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial

Table 3
Activity concentration (Bq kg^{-1}) of LDW, ordinary Portland cement (OPC), and the LDW mortars. The gamma index (*I*) was calculated.

	^{238}U	^{226}Ra	^{210}Pb	^{228}Ra	^{228}Th	^{40}K	^{210}Po	^{232}Th	*I*
OPC	20 ± 1	22 ± 2		13 ± 1	13 ± 1	338 ± 20	18 ± 1	12 ± 1	0.25
LDW10	135 ± 11	9.0 ± 1.6	376 ± 11	10 ± 2	6.7 ± 0.6	370 ± 30	234 ± 4	5.9 ± 0.3	0.18
LDW25	400 ± 40	5 ± 2	410 ± 20	9 ± 3	2.1 ± 0.9	210 ± 30	330 ± 20	5.6 ± 0.3	0.11
LDW35	580 ± 30	10.9 ± 1.4	830 ± 20	8.4 ± 1.9	7.3 ± 0.6	340 ± 20	590 ± 30	7 ± 3	0.18
LDW	7900 ± 600	39 ± 3	5000 ± 110	< 20	< 5	60 ± 10	5120 ± 80	< 0.4	0.20

interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.rineng.2025.107913.

Data availability

Data will be made available on request.

References

[1] A. Maalouf, A. Mavropoulos, *Re*-assessing global municipal solid waste generation, Waste Manag. Res. 41 (2023) 936–947, https://doi.org/10.1177/0734242X221074116/ASSET/IMAGES/LARGE/10.1177_0734242X221074116-FIG5.JPEG.

[2] Z. Quan, X. Xu, W. Wang, J. Jiang, S. Gao, Do industrial solid waste recycling and technological innovation promote low-carbon development in China? New insights from NARDL approach, Sci. Total Environ. 916 (2024) 170446, https://doi.org/10.1016/J.SCITOTENV.2024.170446.

[3] S.M. Pérez-Moreno, M.J. Gázquez, R. Pérez-López, I. Vioque, J.P. Bolívar, Assessment of natural radionuclides mobility in a phosphogypsum disposal area, Chemosphere 211 (2018) 775–783, https://doi.org/10.1016/J.CHEMOSPHERE.2018.07.193.

[4] J. Matsimbe, M. Dinka, D. Olukanni, I. Musonda, Performance evaluation and mix design of ambient-cured fly ash-phosphogypsum blended geopolymer paste and mortar, Results Eng. 24 (2024) 103280, https://doi.org/10.1016/j.rineng.2024.103280.

[5] A.A. Cuadri, S. Pérez-Moreno, C.L. Altamar, F.J. Navarro, J.P. Bolívar, Phosphogypsum as additive for foamed bitumen manufacturing used in asphalt paving, J. Clean. Prod. 283 (2021) 124661, https://doi.org/10.1016/J.JCLEPRO.2020.124661.

[6] L.F.O. Silva, M.L.S. Oliveira, T.J. Crissien, M. Santosh, J. Bolivar, L. Shao, G. L. Dotto, J. Gasparotto, M. Schindler, A review on the environmental impact of phosphogypsum and potential health impacts through the release of nanoparticles, Chemosphere 286 (2022) 131513, https://doi.org/10.1016/J.CHEMOSPHERE.2021.131513.

[7] F.J. Soto-Cruz, S.M. Pérez-Moreno, E. Ceccotti, A. Barba-Lobo, J.P. Bolívar, M. Casas-Ruiz, M.J. Gázquez, Valorisation diagnosis of waste from the decontamination of phosphogypsum leachates through a combined calcium carbonate/hydroxide process, Heliyon 10 (2024) e30610, https://doi.org/10.1016/J.HELIYON.2024.E30610.

[8] S.F. Lütke, M.L.S. Oliveira, S.R. Waechter, L.F.O. Silva, T.R.S. Cadaval, F.A. Duarte, G.L. Dotto, Leaching of rare earth elements from phosphogypsum, Chemosphere 301 (2022) 134661, https://doi.org/10.1016/j.chemosphere.2022.134661.

[9] R. Pérez-López, F. Macías, C.R. Cánovas, A.M. Sarmiento, S.M. Pérez-Moreno, Pollutant flows from a phosphogypsum disposal area to an estuarine environment: an insight from geochemical signatures, Sci. Total Environ. 553 (2016) 42–51, https://doi.org/10.1016/J.SCITOTENV.2016.02.070.

[10] F. Wu, The treatment of phosphogypsum leachate is more urgent than phosphogypsum, Environ. Res. 262 (2024) 119849, https://doi.org/10.1016/J.ENVRES.2024.119849.

[11] S.M. Pérez-Moreno, C. Romero, J.L. Guerrero, M.J. Gázquez, J.P. Bolívar, Development of a process for the removal of natural radionuclides and other stable pollutants from acid phosphogypsum stacks leachates, J. Environ. Chem. Eng. 11 (2023) 109032, https://doi.org/10.1016/J.JECE.2022.109032.

[12] S.M. Pérez Moreno, C. Romero, J.L. Guerrero, A. Barba-Lobo, M. Gázquez, J. P. Bolívar, Evolution of the waste generated along the cleaning process of phosphogypsum leachates, J. Environ. Chem. Eng. 11 (2023) 111485, https://doi.org/10.1016/J.JECE.2023.111485.

[13] J. Liu, F. Zha, L. Xu, B. Kang, X. Tan, Y. Deng, C. Yang, Mechanism of stabilized/solidified heavy metal contaminated soils with cement-fly ash based on electrical resistivity measurements, Measurement 141 (2019) 85–94, https://doi.org/10.1016/j.measurement.2019.03.070.

[14] S. Ahmad, A.A. Bahraq, H.R. Khalid, L.O. Alamutu, Stabilization/solidification of heavy metal-contaminated marl soil using a binary system of cement and fuel fly ash, Environ. Monit. Assess. 195 (2023) 1557, https://doi.org/10.1007/s10661-023-12176-8.

[15] X. Xiong, Y. Zhang, L. Wang, D.C.W. Tsang, Overview of hazardous waste treatment and stabilization/solidification technology. Low Carbon Stab. Solidif. Hazard. Wastes, Elsevier, 2022, pp. 1–14, https://doi.org/10.1016/B978-0-12-824004-5.00031-1.

[16] B. Biakhmetov, S. You, A. Dostiyarov, Sustainable waste management and circular economy. Low Carbon Stab. Solidif. Hazard. Wastes, Elsevier, 2022, pp. 545–554, https://doi.org/10.1016/B978-0-12-824004-5.00032-3.

[17] B.R.S. Calderón-Morales, A. García-Martínez, P. Pineda, R. García-Tenório, Valorization of phosphogypsum in cement-based materials: limits and potential in eco-efficient construction, J. Build. Eng. 44 (2021) 102506, https://doi.org/10.1016/j.jobe.2021.102506.

[18] H. Chen, C.L. Chow, D. Lau, Developing green and sustainable concrete in integrating with different urban wastes, J. Clean. Prod. 368 (2022) 133057, https://doi.org/10.1016/j.jclepro.2022.133057.

[19] H. Chen, R. Qin, C.L. Chow, D. Lau, Recycling thermoset plastic waste for manufacturing green cement mortar, Cem. Concr. Compos. 137 (2023) 104922, https://doi.org/10.1016/j.cemconcomp.2022.104922.

[20] H. Chen, R. Qin, D. Lau, Recycling used engine oil in concrete design mix: an ecofriendly and feasible solution, J. Clean. Prod. 329 (2021) 129555, https://doi.org/10.1016/j.jclepro.2021.129555.

[21] R. Rajamma, R.J. Ball, L.A.C. Tarelho, G.C. Allen, J.A. Labrincha, V.M. Ferreira, Characterisation and use of biomass fly ash in cement-based materials, J. Hazard. Mater. 172 (2009) 1049–1060, https://doi.org/10.1016/J.JHAZMAT.2009.07.109.

[22] F. Ma, L. Chen, Z. Lin, Z. Liu, W. Zhang, R. Guo, Microstructure and key properties of phosphogypsum-red mud-slag composite cementitious materials, Materials (Basel) 15 (2022) 6096, https://doi.org/10.3390/ma15176096.

[23] T. He, Y. Da, C. Shi, M. Wang, J. Wang, Y. Feng, Z. Li, Recovery of thermally treated fluorine-containing sludge as the substitutions of Portland cement, J. Clean. Prod. 260 (2020) 121030, https://doi.org/10.1016/j.jclepro.2020.121030.

[24] A. Bouregba, A. Diouri, B. Elghattas, A. Boukhari, T. Guedira, Influence of Fluorine on Clinker burnability and mechanical properties of CPA Moroccan cement, MATEC Web Conf. 149 (2018) 01075, https://doi.org/10.1051/matecconf/201814901075.

[25] O. Dominguez, A. Torres-Castillo, L.M. Flores-Velez, R. Torres, Characterization using thermomechanical and differential thermal analysis of the sinterization of Portland clinker doped with CaF2, Mater. Charact. 61 (2010) 459–466, https://doi.org/10.1016/J.MATCHAR.2010.02.002.

[26] L. Xie, M. Deng, J. Tang, K. Liu, The effect of fluorapatite in limestones on the mineral compositions of Portland cement clinkers, Constr. Build. Mater. 273 (2021) 122042, https://doi.org/10.1016/j.conbuildmat.2020.122042.

[27] Y. Huang, J. Qian, X. Kang, J. Yu, Y. Fan, Y. Dang, W. Zhang, S. Wang, Belite-calcium sulfoaluminate cement prepared with phosphogypsum: influence of P2O5 and F on the clinker formation and cement performances, Constr. Build. Mater. 203 (2019) 432–442, https://doi.org/10.1016/j.conbuildmat.2019.01.112.

[28] R.L. Rudnick, S. Gao, Composition of the continental crust, in: Treatise Geochemistry, 2nd Ed, 4, 2014, pp. 1–51, https://doi.org/10.1016/B978-0-08-095975-7.00301-6.

[29] M.J. Gázquez, M. Contreras, S.M. Pérez-Moreno, J.L. Guerrero, M. Casas-Ruiz, J. P. Bolívar, A review of the commercial uses of sulphate minerals from the titanium dioxide pigment industry: the case of Huelva (Spain), Minerals 11 (2021) 575, https://doi.org/10.3390/min11060575.

[30] Instruction IS-33, From the nuclear safety council on radiological criteria for protection against the natural radiation (In Spanish), Spain, 2011.

[31] CEN, E.N. 196-5:2011 Methods of testing cement - Part 5: pozzolanicity test for pozzolanic cement, (2011).

[32] J. Rosales, M. Rosales, J.L. Díaz-López, F. Agrela, M. Cabrera, Effect of processed volcanic ash as active mineral addition for cement manufacture, Materials (Basel) 15 (2022) 6305, https://doi.org/10.3390/ma15186305.

[33] V.D. Pinheiro, J. Alexandre, G. de C. Xavier, M.T. Marvila, S.N. Monteiro, A.R.G. de Azevedo, Methods for evaluating pozzolanic reactivity in calcined clays: a review, Materials (Basel) 16 (2023) 4778, https://doi.org/10.3390/ma16134778.

[34] F. Massazza, Pozzolanic cements, Cem. Concr. Compos. 15 (1993) 185–214, https://doi.org/10.1016/0958-9465(93)90023-3.

[35] R. Snellings, G. Mertens, J. Elsen, Supplementary cementitious materials, Rev. Mineral. Geochem. 74 (2012) 211–278, https://doi.org/10.2138/rmg.2012.74.6.

[36] M. Pyzalski, K. Durczak, A. Sujak, M. Juszczyk, T. Brylewski, M. Stasiak, Synthesis and investigation of the hydration degree of CA2 phase modified with boron and fluorine compounds, Materials (Basel) 17 (2024) 2030, https://doi.org/10.3390/ma17092030.

[37] M. Pang, Z. Sun, M. Chen, J. Lang, J. Dong, X. Tian, J. Sun, Influence of phosphorus slag on physical and mechanical properties of cement mortars, Materials (Basel) 13 (2020) 2390, https://doi.org/10.3390/ma13102390.

[38] W. Guo, M. Zhou, Y. Liu, L. Chen, X. Chen, Effect of SO3 content in circulating fluidized bed fly ash on expansion performance of circulating fluidized bed ash-slag unfired brick, Constr. Build. Mater. 489 (2025) 142239, https://doi.org/10.1016/j.conbuildmat.2025.142239.

[39] Y. Zhao, J. Gao, C. Liu, X. Chen, Z. Xu, The particle-size effect of waste clay brick powder on its pozzolanic activity and properties of blended cement, J. Clean. Prod. 242 (2020) 118521, https://doi.org/10.1016/j.jclepro.2019.118521.

[40] M.K. Ardoğa, S.T. Erdoğan, M. Tokyay, Effect of particle size on early heat evolution of interground natural pozzolan blended cements, Constr. Build. Mater. 206 (2019) 210–218, https://doi.org/10.1016/j.conbuildmat.2019.02.055.

[41] AENOR, UNE 83-811-92 Mortars. Test Methods. Fresh Mortars. Determination of Consistence. Flow Table (Reference Method), Spanish Association for Standardisation (AENOR), 1992.

[42] AENOR, EN 196-1 Methods of Testing Cement. Part 1: Determination of Strength, Spanish Association for Standardisation and Certification (AENOR), Spain, 2016.

[43] J. Rosales, M. Cabrera, F. Agrela, Effect of stainless steel slag waste as a replacement for cement in mortars. Mechanical and statistical study, Constr. Build. Mater. 142 (2017) 444–458, https://doi.org/10.1016/J.CONBUILDMAT.2017.03.082.

[44] CEN, EN 16637-2:2023, Construction products: assessment of release of dangerous substances - Part 2: horizontal dynamic surface leaching test, (2023).

[45] UNE-EN 14429:2015. Characterization of waste - leaching behaviour test - influence of pH on leaching with initial acid/base addition, (2015).

[46] P. Martin, G.(Gary) Hancock, Australia. Supervising Scientist., Routine analysis of naturally occurring radionuclides in environmental samples by alpha-particle spectrometry, Supervis. Sci. (2004).

[47] R.L. Lozano, J.P. Bolívar, E.G. San Miguel, R. García-Tenorio, M.J. Gázquez, An accurate method to measure alpha-emitting natural radionuclides in atmospheric filters: application in two NORM industries, Nucl. Instrum. Methods Phys. Res. Sect. A Accel. Spectrom. Detect. Assoc. Equip. 659 (2011) 557–568, https://doi.org/10.1016/j.nima.2011.08.006.

[48] M.W. Yang, W.H. Wu, Y.J. Ruan, L.M. Huang, Z. Wu, Y. Cai, F.F. Fu, Ultra-sensitive quantification of lysozyme based on element chelate labeling and capillary electrophoresis-inductively coupled plasma mass spectrometry, Anal. Chim. Acta 812 (2014) 12–17, https://doi.org/10.1016/j.aca.2014.01.003.

[49] X. Zhang, X. Xue, D. Ding, P. Sun, J. Li, Y. He, A study of the mechanical properties, environmental effect, and microscopic mechanism of phosphorus slag-based uranium tailings backfilling materials, J. Clean. Prod. 446 (2024) 141306, https://doi.org/10.1016/j.jclepro.2024.141306.

[50] H. Yang, Y. Che, F. Leng, High volume fly ash mortar containing nano-calcium carbonate as a sustainable cementitious material: microstructure and strength development, Sci. Rep. 8 (2018) 16439, https://doi.org/10.1038/s41598-018-34851-4.

[51] L. Raki, J. Beaudoin, R. Alizadeh, J. Makar, T. Sato, Cement and concrete nanoscience and nanotechnology, Materials (Basel) 3 (2010) 918–942, https://doi.org/10.3390/ma3020918.

[52] W.-T. Lin, Characterization and permeability of cement-based materials containing calcium fluoride sludge, Constr. Build. Mater. 196 (2019) 564–573, https://doi.org/10.1016/j.conbuildmat.2018.11.126.

[53] H. Peng, A literature review on leaching and recovery of vanadium, J. Environ. Chem. Eng. 7 (2019) 103313, https://doi.org/10.1016/J.JECE.2019.103313.

[54] F.J. Soto-Cruz, S.M. Pérez-Moreno, A. Barba-Lobo, J.P. Bolívar, M. Casas-Ruiz, V. M. García, M.J. Gázquez, Acid neutralisation capacity (ANC) of biomass ashes and its potential use for phosphogypsum leachate cleaning, J. Water Process Eng. 70 (2025) 106968, https://doi.org/10.1016/j.jwpe.2025.106968.

[55] W. Gwenzi, N.M. Mupatsi, Evaluation of heavy metal leaching from coal ash-versus conventional concrete monoliths and debris, Waste Manag. 49 (2016) 114–123, https://doi.org/10.1016/j.wasman.2015.12.029.

[56] S. Overmann, X. Lin, A. Vollpracht, Investigations on the leaching behavior of fresh concrete – a review, Constr. Build. Mater. 272 (2021) 121390, https://doi.org/10.1016/j.conbuildmat.2020.121390.

[57] B. Guo, B. Liu, J. Yang, S. Zhang, The mechanisms of heavy metal immobilization by cementitious material treatments and thermal treatments: a review, J. Environ. Manage. 193 (2017) 410–422, https://doi.org/10.1016/j.jenvman.2017.02.026.

[58] C.E. Tommaseo, M. Kersten, Aqueous solubility diagrams for cementitious waste stabilization systems. 3. Mechanism of zinc immobilizaton by calcium silicate hydrate, Environ. Sci. Technol. 36 (2002) 2919–2925, https://doi.org/10.1021/es0102484.

[59] Y. Li, X. Min, Y. Ke, J. Fei, D. Liu, C. Tang, Immobilization potential and immobilization mechanism of arsenic in cemented paste backfill, Miner. Eng. 138 (2019) 101–107, https://doi.org/10.1016/j.mineng.2019.04.041.

[60] L. Liu, L. Wang, S. Su, T. Yang, Z. Dai, M. Qing, K. Xu, S. Hu, Y. Wang, J. Xiang, Leaching behavior of vanadium from spent SCR catalyst and its immobilization in cement-based solidification/stabilization with sulfurizing agent, Fuel 243 (2019) 406–412, https://doi.org/10.1016/j.fuel.2019.01.160.

[61] S. Kundu, A.K. Gupta, Immobilization and leaching characteristics of arsenic from cement and/or lime solidified/stabilized spent adsorbent containing arsenic, J. Hazard. Mater. 153 (2008) 434–443, https://doi.org/10.1016/j.jhazmat.2007.08.073.

[62] W.-H. Kang, E.-I. Kim, J.-Y. Park, Fluoride removal capacity of cement paste, Desalination 202 (2007) 38–44, https://doi.org/10.1016/j.desal.2005.12.036.

[63] A.P. Galvín, S. Sabrina, B. Auxi, A. Peña, A. López-Uceda, Leaching performance of concrete eco-blocks: towards zero-waste in precast concrete plants, J. Environ. Manage. 344 (2023) 118409, https://doi.org/10.1016/j.jenvman.2023.118409.

[64] L. Tosti, A. van Zomeren, J.R. Pels, R.N.J. Comans, Technical and environmental performance of lower carbon footprint cement mortars containing biomass fly ash as a secondary cementitious material, Resour. Conserv. Recycl. 134 (2018) 25–33, https://doi.org/10.1016/j.resconrec.2018.03.004.

[65] M.C. Llanes, M.J.G. González, S.M.P. Moreno, J.P.B. Raya, Recovery of ilmenite mud as an additive in commercial Portland cements, Environ. Sci. Pollut. Res. 25 (2018) 24695–24703, https://doi.org/10.1007/S11356-018-2498-9/TABLES/7.

[66] Real Decreto 314/2016, De 29 De Julio, Por El Que Se Modifican El Real Decreto 140/2003, De 7 De Febrero, Por El Que Se Establecen Los Criterios Sanitarios De La Calidad Del Agua De Consumo Humano, el Real Decreto 1798/2010, de 30 de diciembre, por el que, Spain, 2016. https://www.boe.es/eli/es/rd/2016/07/29/314.