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1. Production of 1.5 Mt of coal fly ash (FA)
2. Low retail value ~6\$/ton
3. Less than 50% of the FA is used (in 2016)

Scrubbing Process

Granitoid quarries

1. Extraction of 2Mt annually
2. Waste powder after sawing is mixed with underground water or underground water/metal grit/ $\text{Ca}(\text{OH})_2$ mix

Potential improvement of mechanical and chemical properties of concrete (as partial substitute to sand in concrete)

1. No storage cost
2. Lowering CO₂ emissions and reduce energy demand
3. Reducing the use of natural sources
4. Forming a potentially better concrete mixture with increased economic and technological benefits

1 Amounts of words: Manuscript (5,102) + Figure (223) + Tables (1,429) = 6,754
2 + References (1,944) + Supplementary information (2,664) = 11,362

3 Production of Environmentally Friendly Sand-like Products from 4 Granitoid Waste Sludge and Coal Fly Ash for Civil Engineering

5 Roy Nir Lieberman¹, Yaniv Knop², Natalia Moreno Palmerola¹, Carmen Muñoz¹, Haim Cohen³, Maria
6 Izquirdo¹, Jose Angel Lorenzo⁴, Belen Taboas⁴, Oriol Font¹, Xavier Querol¹

- 7 1. *Institute of Environmental Assessment and Water Research (IDAEA), Consejo Superior de*
8 *Investigaciones Cientificas (CSIC), Barcelona, 08034, Spain.*
9 2. *Department of Civil Engineering, Ariel University Samaria, Ariel, 40700 Israel.*
10 3. *Department of Chemical Sciences, Ariel University, Ariel, 40700 Israel.*
11 4. *Centro Tecnológico del Granito, 36410 O Porriño, Galicia, Spain.*

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25 Abstract

26 Processing granitoid rock for civil engineering, countertops, and paving generates
27 sub-economical (transportation costs exceed their retail value) granite powder sludge.
28 This paper evaluates the possibility of mixing granite waste (GW) produced at the
29 Spanish quarries of Galicia with a second sub-economical substance, Class F coal fly
30 ash (FA <20 wt.% CaO). The GW would reduce the leachability of toxic trace
31 elements enriched in Colombian fly ashes and form a sand-like product that could
32 serve as a substitute product and would reduce the costs in industrial concrete and
33 civil engineering projects.

34 To investigate both the capability of the GW to reduce FA leaching (by physical
35 adsorption) and the potential use of the aggregate product as a partial substitute in
36 industrial concrete, the physical (e.g. morphology, crystallographic patterns, and
37 surface area), mineralogical, and chemical (including leachability and chemical
38 composition) properties of the aggregate product were tested. Moreover, properties of
39 the concrete (compressive strength, ion penetration, workability, and electron
40 microscope scanning results (SEM)) were also analyzed.

41 The use of the end scrubbed product in the concrete mixture was found to enhance the
42 performance of the concrete mixture, reducing the leaching of the hazardous
43 elements, and improving the concrete produced.

44 *Keywords: granite powder waste, fly ash, aggregates, toxic trace elements, physical*
45 *adsorption.*

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1. Introduction

Granitoid rock is the most abundant component of the earth's continental crust. Granitoid rock, primarily used in civil engineering and sculpture, is durable against most weathering processes which explains the increased demand (6% a year in the last 10 years) for granitoids (Calmon et al., 1997; Hojamberdiev et al., 2011; Menezes et al., 2005; Vieira et al., 2004). The granitoid quarry industry began in Ancient Egypt and is currently considered the primary worldwide rock industry. Granite manufacturers are found around the globe, including China (the largest), Brazil, India, Saudi Arabia, Spain, Taiwan, Germany, Canada, Italy, Sweden, and the US (mainly MA, GA, NH, SD, and ID) with a total production of 309-349 million m²/year and annual international trading of ~6 billion US dollars (Calmon et al., 1997; Hojamberdiev et al., 2011; Menezes et al., 2005; Vieira et al., 2004). However, sawing granite generates much powder waste containing relatively high concentration SiO₂, Al₂O₃, Fe₂O₃, CaO with several potentially toxic trace elements (e.g. Mo, As). With no efficient treatment of the powder waste during processing, disposal especially in transport, the powder waste causes severe ecological damage (including groundwater contamination) to its surroundings. Yet, there are several uses for the sub-economical (transportation costs exceed their retail value (Karanja, 2017)) granite waste (GW) including its primarily use in the ceramic industry. Furthermore, powder GW was found to reduce production cost, energy needs (specifically by replacing cement which requires a very high energy demand), and storing procedure costs including underwater storage of the granitoid powder and storage in pits until the suspended particles settle (Hojamberdiev et al., 2011; Mármol et al., 2010; Torres et al., 2009; Vieira et al., 2004). GW powder can also be used as raw material for geopolymer synthesis (Tchadjié et al., 2016), fertilizers (Karimi et al., 2012), filler

73 material for roads and embankments, soil stabilization, and fillers in the production
74 of thermoset resin polymers (Singh et al., 2016). In the mortar industry, GW replaced
75 10% of the cement with no loss of compressive strength (after 28 days) (Abukersh
76 and Fairfield, 2011). Moreover, mixing 30% of recycled red granite or granite powder
77 as partial substitute for sand (although the water demand increases) improved
78 properties of the concrete produced (Abukersh and Fairfield, 2011; Felixkala and
79 Partheeban, 2010; Ho et al., 2002; Mármol et al., 2010).

80 Spain has approximately 185 granitoid quarries, most of them located in Galicia
81 (Northwestern Spain). This area suffered intense erosion during the Mesozoic and
82 Cainozoic periods that led to the leveling of sediments to their granite sources.
83 Furthermore, the tectonian cainozoic history of Galica contributed to the exposure of
84 its very active granite reliefs. Those granites were fixed during the middle of the
85 tertiary age when the formation of the major weathering surfaces by chemical erosion
86 or by accumulation (of magma crystals) occurred (Vidal Romaní, 1989). The grantoid
87 quarries in Spain extract ~2 million tonnes (Mt) of granite annually (in 2004). With
88 the increase of worldwide demand, production rose to ~3Mt in 2014 (Harold A.
89 Taylor, 2010; SNSS, 2004), with an annual turnover of more than 336 million Euro
90 (Granito, 2012).

91 Quarrying is not the only major polluting industry in Spain. Coal combustion energy
92 power plants whose total energy production is 274TWH (in 2014) and predicted to
93 increase to ~312.5TWh by 2020 also produce large quantities of potentially hazardous
94 byproducts (Global, 2016; OECD/IEA, 2015). Coal combustion, either, using
95 bituminous (mainly imported from Colombia or South Africa) or anthracite (local or
96 imported), provides ~9% of the electricity produced (12Mt in total). To date, equal
97 amounts of local (anthracite, located mainly in Northern Spain) and imported coals

are used. The inorganic content coals (~20%) lowers their calorific value and requires either treating the coals (by washing with water) or combusting the coal in a mixture of several coals (mostly anthracite coal from Russia or bituminous coals from Colombia or South Africa). This process produces coal fly ash (FA) primarily used as aggregate of clinker substitute or natural sand in the concrete or cement industry (Ahmaruzzaman, 2010; Berndt, 2009; Chris C Ramaseyer, 2009; Feuerborn, 2005; Guerrero et al., 2004; Joshi and Lohita, 1997; Kayali, 2005). Using electrostatic precipitators to separate the FA from the flue gas and yielding more than 99.9%, the annual yield of FA produced in the combustion process is ~1.5M (García-Fraile del Álamo, 2015). These FA are primarily composed of glassy materials forming the aluminosilicate matrix (composed primarily from quartz (SiO_2) and mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$), some hematite and magnetite (mostly in Fe rich coals High sulfate combusted coals also contain gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) or anhydrite (CaSO_4) (García-Fraile del Álamo, 2015; NCAB, 2017).

In recent years, industries have sought options to increase the value, reduce storing cost (landfill costs are increasing every year), and create a higher demand for coal production waste byproducts. Subsequently, FA has been used in agriculture (Arivazhagan et al., 2011; Basu et al., 2009; Li et al., 2014), road construction (Ishai, 2011; Kumar and Patil, 2006; Singh and Shah, 2017; Yao et al., 2015), fire-resistant panels (Li et al., 2014; Li et al., 2015; Li et al., 2017, 2018) and as chemical scrubber for acidic wastes, mine drainage, motor oil regeneration waste, and phosphate waste (Lieberman et al., 2015a; Lieberman et al., 2015b; Lieberman et al., 2017a; Lieberman et al., 2014b; Lieberman et al., 2016a; Orakwue et al., 2016; Qureshi et al., 2016; Rios et al., 2008).

The present study evaluates the potential of reducing leaching of toxic trace elements from the FA and GW by adding sand-like fine aggregates (AP). The incorporation of the end AP formed in concrete would raise economic retail value of both FA and GW value (e.g. FA costs \$6/ton in Europe (Argiz et al., 2015), and \$9-15/ton of granite waste (Karanja, 2015), and could be used as a supplement in concrete production. Thus, using the scrubbed product as partial substitute in concrete after full fixation would reduce the environmental impact of these by-products. Moreover, the current procedure cost ~\$50/ton while the suggested treatment including the raw materials is estimated in ~\$20/ton.

2. Materials & Methods

2.1 Experimental samples

Three Class F (<20% CaO) FAs and four granitoid quarry byproduct wastes (GW) were selected for this study.

The FAs selected were produced as a byproduct of three coal power plant facilities:

- (1) Los Barrios power plant in Los Barrios, Southern Spain which uses Colombian bituminous coal. The Los Barrios FA (LBFA) was produced from the combustion of two pulverized bituminous coals – 45% of B-446 MV XIN Jin Hai and 55% of B-447 MV Pleiades Dream.
- (2) Orot Rabin power plant in Hadera, Israel which uses Colombian bituminous coal. The Colombian FA (COFA) from this facility was produced from the combustion of 100% Drummond Colombian coal.
- (3) Rutenberg power plant in Ashkelon, Israel which uses South African bituminous coal. The South African coal was imported from the Goedehoop mine which produces South African FA (SAFA).

The coals combusted in Israel underwent beneficiation process (washing with water) to lower their S, Cl, and F levels thus increasing the inorganic content of the coals and

producing FA enriched with alkaline and alkaline earth elements (Lieberman et al., 2015a; Lieberman et al., 2017a). As Na oxides are found in lower content and set in a lower temperature compared to Ca oxides, the interactions with the aluminates and silicates on the FA surface were lower than that of the Ca.

A $\approx$ 35 kg of each sample of FA was collected from the electrostatic precipitators and then transferred to sealed polyethylene containers for storage until further analysis.

The physical and chemical properties of the FA were described in previous studies (Lieberman et al., 2015b; Lieberman et al., 2017a; Lieberman et al., 2016b; Lieberman et al., 2014c). All FA bulk XRD analysis in this study indicated that mullite ($3\text{Al}_2\text{O}_3\cdot 2\text{SiO}_2$) and quartz (SiO_2) were the primary crystalline species.

Moreover, traces of hematite (Fe_2O_3) and gypsum ($\text{CaSO}_4\cdot 2\text{H}_2\text{O}$) were detected in LBFA, lime (CaO) in SAFA, and hematite in COFA. XRD patterns also indicated that the main component of the 3 FAs was a glassy amorphous matrix. These results are consistent with the chemical composition of the FAs indicated high aluminosilicate (>70 wt.%), CaO (3.5-10 wt.%), Fe (3-8.9 wt.%). Scanning electron microscope (SEM) analysis showed that all FAs were primarily composed of glassy particles, with magnetite, hematite, and unburned carbon particles. The average particle size analysis showed that the COFA has the finest average particle size with a median of $13\text{ }\mu\text{m}$, the LBFA has similar particle size with a median $13.5\text{ }\mu\text{m}$, and the SAFA particles are slightly coarser with a median of $20.2\text{ }\mu\text{m}$ (Table 1). Also, the surface area of the FA was analyzed indicating a relatively large surface area for non-porous materials ($1.42\text{-}2.42\text{ m}^2/\text{g}$).

The GW selected were quartz rich GW from four sawing processing facilities of O Porriño in northwestern Spain:

1. GW1 collected from Station 1 (Telar 1) with a moisture content of 27.4%, a loss on ignition (L.O.I.) 3.1 wt.%.
2. GW2 collected from Station 2 (Telar 2) with moisture content of 27.2%, L.O.I 1.4 wt.%
3. GW3 collected from Station 3 (Telar 3) with moisture content of 27.6%, L.O.I 1.0 wt.%
4. GW4 collected from Station 4 (Disco 1) with moisture content of 29.5%, L.O.I 2.6 wt.%

Samples received from Telar stations (1-3) were mixed with calcium hydroxide (Ca(OH)_2), metal grit, and recirculated underground well waters. GW4 was mixed only with water.

2.2 Sand-like mixtures preparation

Prior to the fixation process, both the FA and the GW were homogenized for particle size and uniformity (Dan Ravina and Omri Lulav, 2006). Then, 40 AP mixtures were prepared (FA/GW) with 5 ratios of 1/1-1/14 for LBFA/GW producing 20 samples, 3 ratios of 1/6-1/14 for COFA/GW producing 12 samples and 2 ratios of 1/1-1/2 for SAFA/GW producing 8 samples using a temperature-controlled batch reactor with mechanical stirrer (Model CH-450, Hsingtai Machinery Industry Co. Ltd.). Full homogenization of the AP was obtained by mixing the AP for 35 mins at 250rpm, while adding 15-30% Milli-Q water to prevent formation of a non-homogenous solids. The process was followed by drying of the AP for 12h (in oven) at 50 °C. To evaluate the best candidates for use in concrete, chemistry characterization and leaching, mineralogy, particle grain size, and surface area (using BET) analyses were performed on all APs. Then, the chosen sand-like products were used as a partial substitute for natural sand in concrete mixtures with a replacement content of 100 kg m^{-3} (Table 2), and afterwards underwent leaching tests according to the European

Decision protocol (2003/33/EC) using 1/10 kg/L solid/liquid ratio with agitation period of 24h±30min in an end-over-end shaker (Model. Reax 2, Heidolph).

2.3 Concrete mixtures

Concrete mixtures were prepared and tested to evaluate the effect of four sources of GW with FA. The mixture design is described in Table 2.

Mixing all aggregates were mixed with 80% of water for 3 minutes, followed by addition of the cement (EN 197-1:2011 - CEM II/A-M (S, L, V) 42.5 N, chemical composition shown in Table S1) to the mixture and mixed for another 2 minutes. Fly ash, slag, limestone, and gypsum were added to the cement. The final stage consisted of adding the admixture (combination of lignosulfonate and polycarboxylate ether based superplasticizers) with the additional 20% water and mixing for another 5 minutes to reduce the amount of water and improve its flow. A consistent amount of water ($w/c=0.65$) was added maintaining a water-cement ratio of 0.65, and the performance of the fresh concrete (slump) was measured 10 min after mixing according to Israeli Standard IS 26/2.1 (written in compliance with the EU standard test CEN/TC 104) (SIS, 2012).

The casted 20x20x20cm cubical specimens were cured at 21°C and 95% humidity.

The fresh and harden properties of the concrete mixtures were tested; strengths development, slump, air content, specific weight and resist chloride penetration.

For each ratio 4 cubed were produced and tested. The compressive strength of the casted specimens was measured by a press (controls) according to Israeli standard IS 26/4.1 (the Israeli national standard that is in compliance with CEN/TC 104), after 28 days curing (SIS, 2012). Four cubes from each mixture were tested. Furthermore, specific gravity and air content analysis according to IS 26/2.5 and IS 26/2.6

respectfully (in compliance with the CEN standard test EN12350), were performed on each concrete sample (SIS, 2012).

Furthermore, chloride penetration test was performed on each concrete sample according to ASTM C1202 (ASTM, 2004).

The properties of the concrete mixtures containing the partial substitute were compared to those prepared with no FA or AP. Moreover, to evaluate the environmental risk of the concrete, the European Decision protocol (EN12457-2) was applied to all samples (European-Council-Decision, 2003).

2.4 Characterization methods

2.4.1 Chemical analysis

Using an acid digestion ($\text{HNO}_3\text{:HF:HClO}_4$) method, the characterization of 57 major and trace elements was determined for all materials (Querol et al., 1995). Major elements were analyzed by Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES, Model no. iCAP 6500 Radial, Thermo Fisher Scientific), and ICP Mass Spectrometry (ICP-MS, Model X-Series II, Thermo Fisher Corp.) was used for trace elements. To determine the accuracy of the analysis, international coal FA reference material NIST SRM-1633b and reagent blanks underwent an acid-digestion procedure.

XRF (Model Perl'X3, PANalytical) analysis were also performed to determine the major oxides in the raw materials (FA, GW).

The European Standard Leaching Test EN-12457-2 (European Council Decision 2003/33/EC) was applied on all samples to study the environmental effect of the leachates. All leachates were analyzed by ICP-AES, ICP-MS and Waters High Performance Liquid Chromatography (Model 717, Waters Corp.), with an auto-sampler (Model 1525, Waters Corp.), Binary HPLC pump, and conductivity detector

(Model 432, Waters Corp.). Mercury (Hg) concentration in the solid and the leachates was determined using Gold-Amalgam-Atomic-Absorption Spectrometer (Model. AMA 254, Leco). Moreover, a Loss On Ignition (L.O.I.) test was performed by weighing 1.000 g of each sample and inserting them into a Muffle furnace (Model. CWF 1300, Carbolite) at 1050 °C for 4h (Dean, 1974).

Ionic conductivity and pH were measured by conventional methods.

2.4.2 Mineralogy and surface transformation characterization

XRD patterns of powder samples were collected using a Bruker D8 Advance diffractometer with monochromatic Cu K α _{1,2} radiation ($\lambda=1,5405$) operated at 40kV and 40mA. The primary parallel X-ray beam was generated by a Göbbel mirror and the scattered beam was analyzed by a Sol-X detector with the following scanning parameters: from 4 at 60° of 2 θ , a step size of 0.05°.

Surface morphology, transformation (in sand-like fine aggregate (AP)), and composition were determined using a Scanning Electron Microscope with energy dispersive X-ray analyzer ((Model. Quanta 200, FEI).

The grain size distribution of the <63 μ m fraction of each material was determined by a laser light-scattering-based particle size, with a lower detection size of 0.1 μ m (Mastersizer 2000, Model. Hydro 2000MU, Malvern Instruments) (Sperazza et al., 2004). Ultra-sonic dispersion in ethanol was used for 30s on the particle suspension to reduce clusters.

All samples' surface area and pore size properties were analyzed by a Brunauer-Emmett-Teller (BET, Model Micromeritics Gemini V). The analysis was performed in 3 stages: (1) heating the sample to 50⁰ C, (2) 2h of N₂ and He bubbling to eliminate O₂, and (3) transferring the sample to liquid N₂ and perform the analysis.

(Micromeritics, 2014)

3 Results & Discussion

3.1 Byproducts characterization

3.1.1 Granitoid powder waste

The major minerals identified in the GWs were quartz, microcline (KAlSi_3O_8), albite ($\text{NaAlSi}_3\text{O}_8$), and muscovite ($\text{KAl}_2(\text{Si}_3\text{Al})\text{O}_{10}(\text{OH})_2$) as the dominant minerals (Figure S2). Traces of calcite (CaCO_3), maghemite-C (Fe_2O_3), and Fe-rich clinocllore ($\text{Mg, Fe}_6(\text{Si,Al})_4\text{O}_{10}(\text{OH})_8$) were also detected. Furthermore, the chemical composition of GW (Table 3) indicates an observable high concentration (compared to worldwide average) of soluble compounds (lime (CaO), Na_2O , and K_2O) leading to the high alkaline pH measured for GW samples; >11.1 for GW1, >11.2 for GW2, >10.5 for GW3, and >9.7 for GW4 (Blatt et al., 2006; Rudnick and Gao, 2003). The higher pH for GW1-3 compared to GW4 was the result of the added $\text{Ca}(\text{OH})_2$ used when mixing with the underground well water. Moreover, the composition analysis indicated a high content of Al and Fe (10.5-13.4 wt.%, as Al_2O_3 , and 3.3-17.1 wt.% as Fe_2O_3). However, the Al content was relatively lower than the worldwide average composition of granitoids (WA) and the upper continental crust composition. Also, trace elements, minor elements, and rare earth elements (REEs) concentrations were analyzed. Manganese (337-1140 mg/kg), Ni (17-190 mg/kg), Cu (43-188 mg/kg), As (4-10 mg/kg), Cr (9-320 mg/kg), Li (43-49 mg/kg), B (39-68 mg/kg) were detected in significant concentrations. Elements which were found in high concentrations were: (1) Sn (9-18 mg/kg), Ta (8-13 mg/kg) and Nb (42-61 mg/kg). These form usually minerals such as columbite-tantalite (Chicharro et al., 2015; Llorens-González et al., 2017), (2) W-Mo (20-52 mg/kg, and 1.5-59 mg/kg, respectively) that are considered typical in rock samples from Northern Spain (Chicharro et al., 2015; Llorens-González et al., 2017), and (3) Rb (166-196 mg/kg) was also detected in very high

concentrations. In contrast, Ba (200-277 mg/kg), Zr (60-86 mg/kg), Sr (75-107 mg/kg), and V (24-52 mg/kg) were in lower concentrations when compared to the WA and upper continental crust. REEs such as Nd (23-41mg/kg), Yb (1.4-2.8 mg/kg), Th (12-17 mg/kg), Dy (3.2-5.7 mg/kg), Gd (3.7-6.2 mg/kg), Sm (4.5-7.6 mg/kg), La (22-40 mg/kg), Y 14-27 mg/kg, correlated with the worldwide granitoid average composition and the upper continental crust composition (Blatt et al., 2006; Rudnick and Gao, 2003).

The SEM-EDX observations of the GW (Figure 1) particle components show that the particles are composed of Si, Al, Ca, Fe, and K, with a considerable amount of Na, P, Ti, S, and Mg, with a large portion of individual particles made by pure quartz (SiO_2). These observations support the wet-chemistry analysis showing high content of Fe, Ca, Al, Na, and K.

The particle size analysis indicates a similar distribution pattern in all GW, with medians of 11.87-14.46 μm (GW4 had the finest particle average, Figure S1 and Table 1), and a high percentage (~20%) of very fine particles below 2.5 μm . Therefore, these byproducts were defined as hazardous and required storage in water according to European and WHO regulations (EC, 2013; WHO, 2005). Surface area measurements were also performed and indicated relatively high surface areas (8-11 m^2/g).

The European standard leaching test (EN 12457-2, Table 5) was performed on the GW. The leachable levels of pollutants were well below the limits established by the European Council Decision (2003/33/EC) as non-hazardous, therefore the GW will not add environmental risk when used as a partial additive to concrete.

3.1.2 Fly ashes

FA compositions were determined by 3-step acid digestion and compared with previous studies (Córdoba et al., 2012; Guerrero et al., 2004; Lieberman et al., 2015a; Lieberman et al., 2015b; Lieberman et al., 2014b; Lieberman et al., 2016b; Lieberman et al., 2014c). Previous studies (Lieberman et al., 2015a; Lieberman et al., 2016a; Lieberman et al., 2016b) showed that COFA had the finest particle size (median of 13.1 μm) with a three-mode distribution (coarse - 50 μm , fine - 7 μm and finest - 0.7 μm). LBFA had particles similar to those of COFA with a median of 13.5 μm and a three modes distribution (coarse - ~60 μm , fine - ~11 μm , and finest – 0.8 μm). SAFA presented a coarse particle size compared to the LBFA and COFA (with a median of 20.2 μm), but with only two modes of distribution (coarser - 28 μm , and finer – 0.7 μm). Moreover, these studies also indicated a large surface area (for non-porous materials) for all 3 FA samples (1.4-2.4 m^2/g , Table 1). The results of leaching tests performed on the FA samples (Lieberman et al., 2015a; Lieberman et al., 2016a; Lieberman et al., 2016b; Lieberman et al., 2014c) indicated that the high leachable levels of Se, Me, and Sb, both the LBFA and the COFA were hazardous with levels exceeding the non-hazardous limit. However, leaching tests of SAFA indicated that all toxic trace elements were below the limits for non-hazardous waste as established by the European Directive.

3.2 Sand-like fine aggregates (AP) characterization and toxic trace elements immobilization

To determine the chemical characterization of the AP end products produced, all fixation products (LBFA-GW, COFA-GW, and SAFA-GW) were analyzed after an acid digestion procedure (Table S2).

As expected when compared to the starting GW materials, forming an AP using FA and GW increases the content of elements enriched in the FA such as Al, Ti, S, V, Ba, Se, Sb, Sr, As, and Zn content. Furthermore, when compared to the starting GW, a decrease of Ca, and several toxic trace elements (e.g. Cr, Cu, and Mn) was shown. In the original GW, Ca and these toxic elements were found in much higher concentration.

The primary minerals identified for FA+GW blends were quartz, muscovite or phlogopite ($\text{KMg}_3(\text{Si}_3\text{Al})\text{O}_{10}(\text{F},\text{OH})_2$), microcline, and albite were the primary mineral species present (Figure S3). Traces of amesite ($\text{Mg}_2\text{Al}(\text{AlSiO}_5)(\text{OH})_4$), calcite were identified in all samples; mullite was detected in SAFA APs (as a result of the higher FA concentration). Particle size analysis performed on all AP indicated an increase of size compared to the raw materials (FA or GW). The AP produced with either LBFA or COFA attained an average particle size of 15.6-17.7 μm , while that of SAFA attained relatively larger size of 22 μm (Figure S4). The increase of size is due to adhesion of finer particles of each of the raw materials (FA or GW) covered the coarser particles as verified by SEM images indicating the formation of the sand-like fine aggregates (Figure 2). This adhesion probably stems from the very good electrostatic interactions between the alumino-silicate matrix surface of the FA (considerable concentration of free silicates, SiO_3^{2-} , and aluminate, AlO_2^- , anions), positive cations (mainly Ca^{2+}) that serve as mediators between the two anion types and the negative polar oxide groups (mainly silicates) at the surface of GW.

Furthermore, EDX analysis performed on AP samples indicated an increase of Al, S, Ti content and a decrease of Si, Ca, Fe, P as a result of the addition of FA to the GW.

pH analyses performed on the leachates produced end-products was alkaline. Both LBFA and COFA (similar coal origin) indicated similar pH of 9.3-10.1 (Tables S3-S4, Figure 3), while that of the SAFA-GW AP was higher (pH10.8-11.1). Figures 3-4 show that the strongest controls on the mobility of several oxyanions across the AP produced are the FA types, and the ratio between the FA/GW. Most of the oxyanions (e.g. As, Ba, Mo, Se) follow the same pattern; high mobility when LBFA or COFA are used compared to SAFA probably due to higher pH (~11), and formation of new minerals (e.g. ettringite ($\text{Ca}_6\text{Al}_2(\text{OH})_{12}(\text{SO}_4)_3 \cdot 26\text{H}_2\text{O}$), hydrocalumite ($\text{Ca}_4\text{Al}_2(\text{OH})_{12}(\text{OH})_2 \cdot 6\text{H}_2\text{O}$, and thaumasite ($\text{Ca}_3\text{Si}(\text{CO}_3)(\text{SO}_4)(\text{OH})_6 \cdot 12(\text{H}_2\text{O})$) uptaking the oxyanions (Lieberman et al., 2015b; Lieberman et al., 2016b; Zhang and Reardon, 2003).

Leaching behavior of primary and trace elements (according to EN 12457-2) from the APs is presented in Figure 3 and Tables S3-S4. To determine the best mixture for the scrubbing of the GW and the use of the AP in concrete as partial substitute to cement or aggregates, several GWs and FAs. FA/GW ratios from 1/1 to 1/14 kg/kg were tested to fixate the potentially toxic trace elements. High leachability of Se and Mo from APs, produced by either the COFA or LBFA, were obtained (Table S3) as a result of their high concentration of FA and as most of the elements are sensitive to alkaline pH. However, the higher the GW content in the AP, the lower the Se and Mo. These results are in accordance with previous studies suggesting that higher Ca-source material contributes to the adsorption of more Ca compounds on the surface of FA particles, and lowers the yield of toxic trace elements (e.g. Se, and Mo) from the FA (Lieberman et al., 2015a; Lieberman et al., 2017a). With higher GW proportions in the AP (Figure 3 and Table S4), full fixation of all toxic trace elements was achieved, forming a safer and non-hazardous product. Indeed, in order to produce the

best APs to be used in concrete, the mixture composition seems to be dependent on the GW/FA ratio. Thus, with SAFA the best ratio was 1/1; whereas with LBFA and COFA the optimal ratios found were COFA/GW 1/10 – 1/14, and LBFA/GW 1/8 – 1/14.

3.3 Potential use of AP in industrial concrete production

Leaching tests using the concrete end products obtained by substituting up to 100 kg m⁻³ of sand by the mixed product of FA/GW were also performed (Table 3). The results indicated that in all samples the primary leachable elements and species were Ca, SO₄²⁻, Ba, K, Na and Sr. Moreover, after 28 days (final casting day by local standards), the potentially leachable toxic elements were well below the inert limits established by the European Council Decision, 2003/33/EC (with exception of Ba and Cr).

The performances of the fresh and hardened concrete mixtures were tested. The GW-FA blend in a 1-4 ratio was used as a 100kg m⁻³ partial replacement for natural sand. This ratio was chosen based on previous studies using a waste/FA sand-like product (Lieberman et al., 2014a). The slump, specific gravity, and air content of the fresh concrete were measured after 10 min of mixing (Figure 5A-C). No significant difference in slump was measured, thus, the workability and the amount of water necessary to achieve a constant slump was similar when using the APs as partial substitutes. However, concrete mixtures containing the AP indicated smaller specific gravity (Figure 5B), probably as a result of the higher sand density (2.6 g/cm³) when compared to that of the AP (2.2 g/cm³). Moreover, no change of air content was shown. All samples contained air percentage of 1.0-1.5%.

Compressive strength tests performed after 1 day and 28 days of casting indicated that addition of the AP resulted in larger compressive strength (~10-15%) when compared to the concrete mixture without AP (Figure 6A). No significant differences were obtained after 1 day of casting. The greater strength after 28 days of casting is the result of the reaction between the FA and the calcium hydroxide ($\text{Ca}(\text{OH})_2$) to C-S-H (Figure 7) (Celik et al., 2015; Schöler et al., 2015; Yao et al., 2015), and also due to increase of packing density of the harden paste, as explained by Knop (Knop and Peled, 2016; Knop et al., 2014). Furthermore, a resistance to chloride ion penetration test (Figure 6B) was performed determining that using the AP as partial substitute appreciably reduces the ion penetration.

Concrete leaching tests (Table 5) for all concretes produced were performed. Results indicate that the final concrete product achieved full neutralization and fixation according to the limits established by the European Directive (2003/33/EC). Similar results were reported by previous studies when using a sand-like product produced mixing FA with Ca enriched material (e.g. dolomite quarries wastes) (Lieberman et al., 2018; Lieberman et al., 2017b). Thus, a reasonable assumption from this study is when comparing the compressive strengths, workability, resistance to chloride ion penetration, leaching using the FA and GW as AP, in industrial concrete produces enhanced industrial concrete.

Based on the tests of the concrete blends, we conclude that by using FA-GW blend as partial replacement for natural sand in concrete improved properties of the harden concrete. Hence, by using the AP there is a potential to improve the fresh and harden properties. Furthermore, by using the AP formed as a partial substitute in the concrete blends, the procedure costs are reduced by ~60% as a result of the low economic retail cost of the FA and GW.

4 Conclusions

Preliminary tests performed on the casted concrete suggest that by partially replacing natural sand with a Class F FA/GW mixture (up to 10%) can improve its performance. The FA/GW added to the concrete mixture could potentially increase both the ecological (as neither disposal nor storage cost are needed, and natural sand substitution is a formerly undesirable byproduct), economical, and technological benefits of the fortified concrete. Moreover, because GW formed during the cutting of the stone and include a high proportion of breathable particles ($<PM_{2.5}$), it may produce negative health effects (such as cancer), pollution, and environmental damage. However, when mixed properly with the FA, an environmentally safe AP is formed.

Furthermore, since GW contains a high Ca content, adsorption of its' fine particles on the FA can form a physical binder which reduces the leaching potential of toxic trace elements, especially Mo and Se.

The resulted AP is characterized by low leaching yields of trace elements and potentially harmful oxyanionic elements enriched in the FA (e.g. Se, Mo, As). The lower leaching yield is achieved as a result of electrostatic interactions between the aluminosilicate matrix of the FA and the positive cations (mainly Ca^{2+}) serve as mediators between the two anion types (GW silicates and FA aluminosilicate). Also, formation of ettringite, hydrocalumite, and thaumasite can uptake harmful oxyanions.

Leaching potential tests of the concrete mixtures defined that no additional environmental risk occurs during producing mixtures containing the AP. Moreover, most samples were considered as inert as all toxic trace elements were below the limits established in the 2003/33/EC European Decision (with exception to Cr and Ba). Furthermore, the improved performance obtained, would increase the use of

GW-FA mixture as partial substitute, lower operational costs, and benefit many fields such as (1) industry (producing non-hazardous byproducts), (2) environmental by reducing the environmental pollution - reducing the use of natural sources and the waste storing, and reduce energy demand, and (3) forming a potentially better concrete mixture that improve the product both from the economic and technological aspects.

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Supporting Information Available

Supporting information for this article includes additional figures and table: Size Chemical composition of the original cement (Table S1), Chemical composition of the fly ash and granitoid aggregate end products (Table S2), Leaching test according to EN12457-2 for AP produced using GW and FA (lower ratios) with the non-hazardous limit established (Directive 2003/33/EC) (Table S3), AP (using GW and FA) leaching test results according to the EN12457-2 (Table S4), Particle size distribution and cumulative percentage of granitoid byproducts (Figure S1), XRD

spectra of the GW produced in f Ponte Vendra quarries facilities, NW Spain. (Figure S2), XRD spectra of the AP produced by mixing Los Barrios fly ash with granitoid waste, NW Spain (Figure S3), Particle size distribution and cumulative percentage of produced Sand like products (Figure S4).

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731 Supplementary DATA

732 Table S1: Chemical composition of the original cement

Component	Wt. %
CaO	58.2
SiO ₂	20.6
Al ₂ O ₃	6.3
Fe ₂ O ₃	3.6
MgO	1.1
TiO ₂	0.41
K ₂ O	0.45
Na ₂ O	0.12
P ₂ O ₅	0.36
Mn ₂ O ₃	0.05
SO ₃	3.07
LOI	2.8
RI	N.A.

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742 Table S2: Chemical composition of the fly ash and granitoid aggregate end products

	SA-GW1	SA-GW2	SA-GW3	SA-GW4	CO-GW1	CO-GW2	CO-GW3	CO-GW4	LB-GW1	LB-GW2	LB-GW3	LB-GW4
.wt%	1/1	1/1	1/1	1/1	1/14	1/6	1/6	1/14	1/12	1/12	1/8	1/14
Al ₂ O ₃	27	20	24	20	22	12	11.8	12	14	12	12	11.7
B ₂ O ₃	0.17	0.11	0.10	0.04	0.14	0.03	0.02	0.02	0.02	0.06	0.02	0.09
BaO	0.076	0.15	0.17	0.14	0.14	0.06	0.06	0.06	0.05	0.05	0.05	0.05
CaO	2.1	7.3	8.2	7.1	6.3	4.4	3.8	4.4	2.7	4.2	3.6	4.0
Fe ₂ O ₃	10.7	5.4	8.1	7.7	3.1	7.8	10.0	10.0	3.4	7.8	9.7	11.4
K ₂ O	2.2	2.4	2.9	2.0	2.4	4.0	4.0	3.3	3.8	3.9	4.1	3.2
MgO	0.75	1.41	1.51	1.95	1.64	0.70	0.46	2.0	1.24	0.72	0.48	1.83
Na ₂ O	0.31	1.60	1.97	1.39	1.85	2.9	3.0	2.7	3.1	2.8	3.0	2.5
P ₂ O ₅	0.53	1.33	1.57	1.33	1.29	0.10	0.10	0.14	0.10	0.09	0.09	0.12
SO ₃	0.49	0.24	0.28	0.24	0.23	0.10	0.09	0.12	0.10	0.09	0.07	0.11
SrO	0.12	0.22	0.26	0.21	0.21	0.02	0.02	0.02	0.02	0.02	0.02	0.02
TiO ₂	1.24	0.95	1.11	0.93	0.94	0.28	0.29	0.34	0.35	0.28	0.29	0.31
mg/kg												
Li	175	211	173	172	42	48	54	48	41	50	50	50
Be	6.2	8.2	6.2	5.9	3.9	4.1	4.4	3.4	3.0	4.1	3.8	3.8
Sc	22	26	22	22	7.6	5.9	10.2	6.9	6.1	4.2	7.2	6.4
V	90	106	99	88	39	42	68	46	43	43	67	46
Cr	128	182	182	94	102	153	187	16	101	141	198	17
Mn	554	781	745	409	649	751	871	363	621	724	926	391
Co	19	23	24	65	10.6	10.8	19	95	9.2	9.2	17	100
Ni	68	104	108	38	78	109	133	26	83	99	141	24
Cu	93	139	132	75	90	129	140	58	101	124	152	60
Zn	91	105	88	89	54	55	60	56	57	56	63	62
Ga	32	38	30	31	17	18	18	18	16	17	16	18
Ge	6.4	7.4	6.4	6.2	3.1	3.2	4.2	3.0	2.6	2.7	3.2	2.4
As	13	18	16	13	11	14	16	10.5	14	16	18	12
Se	2.3	3.8	3.1	2.6	2.7	2.1	3.4	1.58	2.4	1.6	3.0	2.1
Rb	111	146	105	120	186	202	181	193	179	207	173	205
Y	56	70	53	57	27	29	21	27	25	27	19	28
Zr	266	326	256	265	91	113	94	104	83	105	78	107
Nb	24	30	23	24	10.0	13	10.2	11.9	8.7	11.2	8.8	14
Mo	16	23	21	7.7	22	28	24	3.9	22	26	30	4.4
Cd	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Sn	10.0	15	13	10.6	8.9	11.9	12	10.2	10.4	12	13	10.2
Sb	1.22	1.80	1.52	0.92	1.61	2.1	2.6	1.11	1.58	1.88	2.5	0.92
Cs	6.3	8.6	6.5	7.1	8.3	9.8	8.7	9.0	8.5	10.3	8.7	9.8
La	92	110	84	91	51	46	35	44	49	43	32	45
Ce	187	231	173	187	105	93	72	93	102	89	67	94
Pr	19	23	17	19	11.9	10.8	8.2	10.6	11.5	10.3	7.7	10.6
Nd	73	86	66	71	46	41	32	41	45	40	29	41
Sm	14	17	13	14	9.1	8.4	6.4	8.1	8.6	8.0	6.0	8.4
Eu	2.0	2.4	1.9	2.0	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.8
Gd	13	15	12	13	7.7	7.4	5.6	7.1	7.6	7.4	5.1	7.2
Tb	1.9	2.3	1.8	1.9	1.02	1.07	<0.01	1.00	0.99	1.01	<0.01	1.05
Dy	11.4	14	10.7	11.6	6.0	6.2	4.5	6.0	5.8	6.1	4.1	6.1
Ho	2.2	2.7	2.0	2.2	1.06	1.19	0.84	1.08	1.02	1.13	<0.01	1.13
Er	6.3	7.8	5.9	6.4	3.0	3.4	2.3	3.1	2.8	3.1	2.1	3.2
Tm	0.91	1.11	0.85	0.92	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Yb	5.8	7.3	5.6	6.1	2.6	3.1	2.1	2.8	2.6	2.9	2.0	3.0
Lu	0.83	1.04	<0.01	0.84	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Hf	7.8	9.6	7.5	7.8	3.1	3.7	3.1	3.5	2.9	3.7	2.6	3.8
Ta	2.0	2.6	2.0	2.0	<0.01	0.83	<0.01	0.91	<0.01	<0.01	<0.01	1.45
W	4.9	7.0	7.5	8.7	2.8	3.7	6.8	7.7	3.3	3.6	6.8	10.5
Tl	0.90	1.16	0.90	0.99	1.04	1.12	1.06	1.10	1.11	1.23	1.16	1.22
Pb	49	60	48	49	25	27	24	25	24	27	22	25
Bi	1.68	2.2	1.86	1.65	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Th	37	48	36	40	22	23	16	20	21	23	15	22
U	10.2	14	11.1	10.6	6.7	9.5	5.9	7.4	5.5	7.8	6.2	7.2

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745

746 Table S3: Leaching test according to EN12457-2 for AP produced using GW and FA

747 (lower ratios) with the non-hazardous limit established (Directive 2003/33/EC)

748

	LB/GW1	LB/GW1	LB/GW1	LB/GW2	LB/GW2	LB/GW2	LB/GW3	LB/GW3	LB/GW3	LB/GW4	LB/GW4	LB/GW4	Non-hazardous limit
mg/kg	1/2	1/4	1/6	1/2	1/4	1/6	1/2	1/4	1/6	1/2	1/4	1/6	
pH	9.7	9.7	9.8	9.8	9.8	9.9	9.9	10.0	10.0	9.7	9.7	9.4	>6
Al	14	9.8	11.4	11.5	13	10.8	9.5	8.1	7.1	9.5	5.8	3.6	
Ca	474	184	151	284	187	119	487	222	147	685	364	314	
Fe	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	
K	331	312	316	301	289	253	221	223	215	164	136	162	
Mg	15	6.6	3.8	10.0	5.8	3.2	46	37	25	35	26	22	
Na	684	491	476	613	538	447	615	442	380	416	230	255	
P	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.2	<0.4	<0.5	<0.6	
SO ₄ ²⁻	2712	1514	1381	1998	1522	1127	2419	1300	931	2478	1295	1253	20000
Si	15	23	24	16	19	26	15	21	22	21	31	32	
Cl ⁻	6.2	6.9	7.7	5.5	6.2	6.4	18	19	20	5.8	4.2	6.3	15000
NO ₃ ⁻	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
F ⁻	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	150
Br ⁻	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Li	0.82	0.48	0.40	0.72	0.59	0.45	0.85	0.54	0.42	0.67	0.33	0.37	
Be	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
B	27	16	12	22	20	12	27	17	12	29	16	14	
Sc	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Ti	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
V	1.82	1.65	1.45	0.24	0.61	0.50	0.18	0.20	0.09	3.4	2.5	1.87	
Cr	0.09	0.03	0.03	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.77	0.30	0.27	10
Mn	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Co	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Ni	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	10
Cu	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.01	50
Zn	0.03	0.05	0.03	0.02	0.03	0.03	0.06	0.04	0.03	0.02	0.03	0.03	50
Ga	0.18	0.13	0.13	0.07	0.11	0.07	0.04	0.03	0.02	0.10	0.06	0.03	
Ge	0.05	0.04	0.03	0.01	0.02	0.02	0.01	0.01	<0.01	0.14	0.10	0.06	
As	0.36	0.33	0.26	0.11	0.25	0.18	0.07	0.09	0.04	1.15	1.19	0.95	2
Se	2.4	1.63	1.21	1.62	1.35	1.09	1.83	0.91	0.53	3.0	2.21	1.62	0.5
Rb	0.73	0.67	0.68	0.64	0.62	0.52	0.47	0.45	0.43	0.34	0.27	0.31	
Sr	5.1	1.32	0.89	2.6	1.64	0.66	6.6	2.2	1.03	10.4	4.76	3.59	
Y	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Zr	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Nb	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Mo	10.2	5.1	4.5	3.9	4.2	1.85	2.4	0.88	0.23	8.1	3.8	3.7	10
Cd	0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	1
Sn	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Sb	0.18	0.13	0.12	0.06	0.08	0.05	0.05	0.04	0.02	0.17	0.10	0.08	0.7
Cs	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.01	<0.01	<0.01	
Ba	0.58	0.15	0.09	0.30	0.19	0.06	0.92	0.33	0.13	1.76	0.70	0.52	100
La	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Ce	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Pr	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Nd	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Sm	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Eu	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Gd	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Tb	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Dy	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Ho	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Yb	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Lu	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Hf	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Ta	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
W	1.62	0.90	0.82	0.32	0.62	0.26	0.21	0.15	0.06	2.41	1.49	1.91	
Tl	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Pb	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	10
Bi	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
Th	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	
U	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	

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752 Table S4: AP (using GW and FA) leaching test results according to the EN12457-2

mg/kg	CO:GW1 1:14	SA:GW1 1:1	LB:GW1 1:12	CO:GW2 1:6	SA:GW2 1:1	LB:GW2 1:12	CO:GW3 1:6	SA:GW3 1:1	LB:GW3 1:8	CO:GW4 1:14	SA:GW4 1:1	LB:GW4 1:14
pH	9.8	11.0	9.71	9.7	11.0	10	9.9	11.0	9.94	9.3	11.1	9.5
Al	4.4	57	4.5	6.5	49	5.1	4.4	46	3.3	3.6	40	2.1
B	3.8	9.1	3.4	8.2	9.6	3.6	8.3	9.9	4.7	6.4	7.5	5.4
Ba	<0.01	0.20	<0.01	<0.01	0.20	<0.01	0.24	0.28	<0.01	0.21	0.23	<0.01
Ca	67	577	72	166	581	42	217	594	99	171	609	122
Fe	<0.025	<0.025	<0.025	<0.025	<0.025	<0.025	<0.025	<0.025	<0.025	<0.025	<0.025	<0.025
K	206	186	171	172	166	144	163	147	156	137	105	134
Mg	3.5	2.9	2.7	8.2	3.0	1.78	54	3.5	30	36	3.1	27
Na	359	181	294	359	183	278	328	162	280	150	83	130
P	< 0,050	< 0,050	< 0,050	< 0,050	< 0,050	< 0,050	< 0,050	< 0,050	< 0,050	3.5	0.2	2.8
SO ₄ ²⁻	756	1442	639	1081	1418	391	1089	1279	522	786	1071	523
F-	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Si	27	28	26	14	33	36	17	32	18	25	42	38
Sr	0.2	7.9	0.20	0.82	8.3	0.10	1.60	9.0	0.35	1.77	7.3	0.76
Li	0.39	5.4	0.29	0.82	5.8	0.32	0.78	6.6	0.40	0.30	3.9	0.20
Be	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
B	3.0	6.5	2.8	6.0	7.1	3.0	6.2	7.3	3.9	5.1	6.0	4.3
Sc	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Ti	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.0	<0.01	0.0
V	0.22	0.85	0.30	0.05	0.86	0.11	0.08	0.79	0.03	1.22	0.66	0.94
Cr	<0.01	2.0	<0.01	<0.01	1.94	<0.01	<0.01	1.69	<0.01	0.10	1.25	0.10
Mn	<0.01	<0.01	<0.01	0.02	<0.01	0.01	0.02	<0.01	0.01	<0.01	<0.01	0.01
Co	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Ni	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Cu	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.01	<0.01	0.01
Zn	0.03	0.04	0.03	0.03	0.04	0.03	0.04	0.03	0.02	0.06	0.03	0.04
Ga	0.04	0.59	0.04	0.02	0.56	0.02	0.02	0.59	<0.01	0.04	0.35	0.01
Ge	0.01	<0.01	0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.07	<0.01	0.04
As	0.05	0.04	0.10	0.01	0.03	0.08	0.02	0.03	0.03	0.03	0.05	0.75
Se	0.35	0.09	0.43	0.35	0.18	0.17	0.46	0.27	0.24	0.83	0.31	0.81
Rb	0.40	0.46	0.34	0.36	0.41	0.28	0.32	0.34	0.30	0.29	0.27	0.27
Sr	0.22	9.1	0.19	0.92	9.8	0.09	1.83	10	0.40	2.0	8.6	0.83
Y	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Zr	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Nb	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Mo	0.13	1.21	0.34	0.15	1.15	0.15	0.20	1.05	0.13	0.94	0.72	1.14
Cd	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Sn	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Sb	0.02	0.04	0.03	0.01	0.04	<0.01	0.02	0.04	<0.01	0.07	0.04	0.04
Cs	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Ba	0.02	0.22	0.01	0.10	0.23	<0.01	0.28	0.32	0.04	0.24	0.26	0.07
La	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Ce	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Pr	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Nd	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Sm	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Eu	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Tb	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Dy	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Ho	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Er	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Tm	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Yb	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Lu	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Hf	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Ta	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
W	0.01	0.52	0.02	<0.01	0.50	<0.01	0.01	0.57	0.01	0.31	0.43	0.35
Tl	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Pb	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Bi	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Th	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
U	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.01	<0.01	<0.01

Figure S1: Particle size distribution and cumulative percentage of granitoid byproducts. (A) GW1, (B) GW2, (C) GW3, and (D) GW4

757 Figure S2: XRD spectra of the GW produced in f Ponte Vedra quarries facilities, NW
 758 Spain. (A) GW1, (B), GW2, (C) GW3, and (D) GW4. Q: quartz, M: microcline, C:
 759 Calcite, Ms: Muscovite, Cl:clinochlore, Mg:Maghemite, A: albite

773 Figure S3: XRD spectra of the AP produced by mixing Los Barrios fly ash with
 774 granitoid waste, NW Spain. (A) GW1, (B), GW2, (C) GW3, and (D) GW4. Q: quartz,
 775 Ms: Muscovite, Cl: Clinocllore, Mu: mullite, M: microcline, A: Albite, C: calcite,
 776 Mn: magnetite
 777

Figure S4: Particle size distribution and cumulative percentage of produced Sand like products. (A) LB-GW1 1:12, (B) LB-GW2 1:12, (C) LB-GW3 1:8, (D) LB-GW4 1:14, (E) CO-GW1 1:6, and (F) SA-GW1 1:1

Tables

Table 1: Particle size and surface area analyses performed for starting samples

Sample	Average particle size (μm)	Surface area ($\text{m}^2 \text{g}^{-1}$)
GW1	14.5	8.4
GW2	12.9	11.7
GW3	13.4	N.A.
GW4	11.9	N.A.
LBFA	13.5	1.4
COFA	13.1	1.6
SAFA	20.2	2.4

Table 2: Mix design of the concrete (m³)

Material	Concrete reference kg m⁻³	Concrete mixture with FA/GW mixture, kg m⁻³ * 100 kg m⁻³ partial replacement
CEM II 42.5 N/AM-SLV	260	260
Coarse aggregate (14-25 mm)	850	850
Fine aggregate (0-5 mm)	750	750
Sand (0-1.2 mm)	400	300
Water	180	180
Chemical admixture (PCE & LS)*	2.6	2.6

*PCE - Polycarboxylate ether superplasticizer, LS - Low Shrinkage

Table 3: Chemical composition of the fly ash (FA) and granitoid byproduct waste (GW)(analysis of dry samples). Average worldwide composition (Blatt et al. 2006), Upper Continental crust (Rudnick and Gao, 2003)

<i>Compound –wt. %</i>	<i>LBFA</i>	<i>SAFA</i>	<i>COFA</i>	<i>GW1</i>	<i>GW2</i>	<i>GW3</i>	<i>GW4</i>	Average worldwide composition	Upper continental crust
<i>SiO₂</i>	53	41	56	63.4	63.5	58.5	68.0	72	66.6
<i>Al₂O₃</i>	20.5	31	23	12.0	11.6	10.5	13.4	14.4	15.4
<i>Fe₂O₃</i>	8.9	2.7	6.3	4.4	3.8	4.0	2.7	1.2 1.7 as FeO	5.04
<i>CaO</i>	3.51	9.5	3.4	7.7	10.6	17.1	3.3	4.1	3.59
<i>K₂O</i>	1.91	0.8	1.6	4.3	4.3	3.3	4.1	3.7	2.80
<i>MgO</i>	1.91	2.1	1.7	0.7	0.3	1.7	1.3	0.71	2.48
<i>Na₂O</i>	4.5	0.4	2.3	3.0	3.1	2.6	3.2	3.7	3.27
<i>P₂O₅</i>	0.17	2.3	0.3	0.1	0.1	0.1	0.1	0.12	0.15
<i>SO₃</i>	0.72	0.7	0.9	0.1	0.1	0.1	0.1	N.A.	N.A.
<i>TiO₂</i>	0.79	1.6	1	0.2	0.2	0.2	0.3	0.3	0.64
<i>LOI</i>	2.03	5	2.3	3.06	1.43	1.01	2.56	N.A.	N.A.
<i>Element (mg/kg)</i>									
<i>Li</i>	76	387	102	43	48	49	48		24
<i>Be</i>	5.6	9.2	10	2.7	3.6	2.8	2.9		2.1
<i>B</i>	419	161	322	64	39	65	68		17
<i>Sc</i>	34	26	35	4.4	3.0	4.7	4.1		14
<i>V</i>	260	143	272	24	26	52	25		97
<i>Cr</i>	90	153	119	92	171	320	9		92
<i>Hg</i>	0.07	0.4	0.22	<0.001	<0.001	<0.001	<0.001		N.A.
<i>Mn</i>	331	436	331	590	715	1143	337	500	774
<i>Co</i>	23	30	38	10	6.9	17	84		17.3
<i>Ni</i>	9.2	48	99	64	100	190	17		47
<i>Cu</i>	81	88	82	78	115	188	43		28
<i>Zn</i>	144	148	175	57	56	62	58		67
<i>Ga</i>	32	57	43	17	17	15	17		17.5
<i>As</i>	63	16	27	7.1	9	10	4.0		4.8
<i>Se</i>	41	10	27	<0.01	<0.01	<0.01	<0.01		0.09
<i>Rb</i>	90	34	63	182	196	166	186		84
<i>Sr</i>	1180	2530	844	75	84	82	107		320
<i>Y</i>	47	85	54	23	27	14	24		21
<i>Zr</i>	222	279	235	76	86	60	78		193
<i>Nb</i>	18	76	50	42	61	48	45		12
<i>Mo</i>	45	10	25	25	36	59	1.48		1.1
<i>Cd</i>	3.2	0.01	2.1	<0.01	<0.01	<0.01	<0.01		0.09
<i>Sn</i>	3.1	<0.001	4	9	13	18	10		2.1
<i>Sb</i>	15	0.9	15	<0.01	1.08	1.66	<0.01		0.4
<i>Cs</i>	7.6	4.7	5.7	6.7	8.1	6.9	7.1		4.9
<i>Ba</i>	3770	1790	3580	277	285	200	241		628
<i>La</i>	46	121	52	40	36	22	35		31
<i>Ce</i>	84	225	98	90	80	49	77		63
<i>Pr</i>	10	27	12	11	9	6	9		7.1
<i>Nd</i>	37	89	43	41	37	23	36		27
<i>Sm</i>	7.6	18	9.5	7.6	7.3	4.5	7.0		4.7
<i>Eu</i>	2.5	2.7	2.0	<0.01	<0.01	<0.01	<0.01		1.0
<i>Gd</i>	7.5	17	9.4	6.2	6.1	3.7	5.8		4.0
<i>Dy</i>	9	16	9.4	5.1	5.7	3.2	5.3		3.9
<i>Er</i>	5.5	8.6	5.3	2.6	3.0	1.55	2.8		2.3
<i>Yb</i>	4.1	8.4	5.2	2.3	2.8	1.42	2.7		1.96
<i>Ta</i>	2.2	10	5	5.5	13	10	8.2		0.9
<i>W</i>	6.2	18	15	20	25	52	44		1.9
<i>Pb</i>	31	80	44	20	23	18	21		17
<i>Th</i>	14	47	19	16	17	12	17		10.5
<i>U</i>	6.8	17	8	5.9	7.1	5.4	6.7		2.7

Table 4: GW leaching test results according to the EN12457-2 compared with the non-hazardous limit (Directive 2003/33/EC)

mg/kg	GW1	GW2	GW3	GW4	Non-hazardous limit
pH	11.1	11.2	10.5	9.7	6
Al	56	61	5.8	5.9	
Ca	64	116	66	50	
Fe	<0.1	<0.1	<0.1	0.33	
K	342	239	216	151	
Mg	<0.1	<0.1	5.8	26	
Na	340	227	269	124	
P	<0.1	<0.1	<0.1	<0.1	
Si	67	50	86	70	
Sr	0.05	0.08	0.05	0.12	
SO ₄ ²⁻	305	139	196	237	20000
Cl ⁻	11.3	11.7	26.0	11.3	15000
NO ₃ ⁻	0.56	<0.1	0.72	<0.1	
F ⁻	<0.1	<0.1	<0.1	<0.1	150
Li	0.11	0.14	0.16	0.12	
Be	<0.01	<0.01	<0.01	<0.01	
B	0.19	0.25	0.19	0.11	
Sc	<0.01	<0.01	<0.01	<0.01	
Ti	<0.01	<0.01	<0.01	0.02	
V	0.10	0.11	0.17	0.02	
Cr	0.18	0.21	0.05	<0.01	10
Hg	<0.01	<0.01	<0.01	<0.01	0.2
Mn	<0.01	<0.01	<0.01	<0.01	
Co	<0.01	<0.01	<0.01	0.01	
Ni	<0.01	<0.01	<0.01	<0.01	10
Cu	<0.01	0.02	0.02	0.03	
Zn	<0.01	0.02	0.02	0.02	50
Ga	0.14	0.15	0.03	<0.01	
Ge	<0.01	<0.01	<0.01	<0.01	
As	<0.01	<0.01	0.01	0.06	2
Se	<0.01	<0.01	<0.01	<0.01	0.5
Rb	0.72	0.53	0.40	0.28	
Sr	0.05	0.07	0.05	0.12	
Y	<0.01	<0.01	<0.01	<0.01	
Zr	<0.01	<0.01	<0.01	<0.01	
Nb	<0.01	<0.01	<0.01	<0.01	
Mo	0.44	0.26	0.14	0.05	10
Cd	<0.01	<0.01	<0.01	<0.01	1
Sn	<0.01	<0.01	<0.01	<0.01	
Sb	<0.01	<0.01	<0.01	<0.01	0.7
Cs	<0.01	<0.01	<0.01	<0.01	
Ba	<0.01	<0.01	<0.01	0.016	100
La	<0.01	<0.01	<0.01	<0.01	
Ce	<0.01	<0.01	<0.01	<0.01	
Pr	<0.01	<0.01	<0.01	<0.01	
Nd	<0.01	<0.01	<0.01	<0.01	
Sm	<0.01	<0.01	<0.01	<0.01	
Eu	<0.01	<0.01	<0.01	<0.01	
Gd	<0.01	<0.01	<0.01	<0.01	
Tb	<0.01	<0.01	<0.01	<0.01	
Dy	<0.01	<0.01	<0.01	<0.01	
Ho	<0.01	<0.01	<0.01	<0.01	
Er	<0.01	<0.01	<0.01	<0.01	
Yb	<0.01	<0.01	<0.01	<0.01	
Lu	<0.01	<0.01	<0.01	<0.01	
Hf	<0.01	<0.01	<0.01	<0.01	
Ta	<0.01	<0.01	<0.01	<0.01	
W	0.61	0.51	0.95	1.30	
Tl	<0.01	<0.01	<0.01	<0.01	
Pb	<0.01	<0.01	<0.01	<0.01	10
Bi	<0.01	<0.01	<0.01	<0.01	
Th	<0.01	<0.01	<0.01	<0.01	
U	<0.01	<0.01	<0.01	0.01	

Table 5: Concrete leaching test results according to the EN12457-2 compared with the inert, non-hazardous limit, and hazardous limits (Directive 2003/33/EC)

mg/kg	Standard	GW1/FA + cement	GW2/FA + cement	GW3/FA + cement	DISCO1/FA + cement	Inert limit	Non- hazardous limit	Hazardous limit
pH	12.5	12.4	12.4	12.4	12.4		>6	
Conductivity (mS)	7.1	7.1	6.9	7.2	7.3			
Al	<0.1	2.46	<0.1	1.58	2.51			
Ca	7030	6930	7010	6680	6700			
Fe	<0.1	<0.1	<0.1	<0.1	<0.1			
K	537	736	728	620	661			
Mg	<0.1	<0.1	<0.1	<0.1	<0.1			
Na	329	400	408	342	363			
P	<0.1	<0.1	<0.1	<0.1	<0.1			
SO4 ²⁻	57	38	40	37	36	1000	20000	50000
Si	2	<0.1	<0.1	<0.1	<0.1			
Li	0.78	2.74	2.80	2.15	2.34			
Be	<0.01	<0.01	<0.01	<0.01	<0.01			
B	0.11	0.07	0.05	0.06	0.05			
Sc	0.06	0.01	<0.01	<0.01	<0.01			
Ti	0.02	0.01	<0.01	<0.01	<0.01			
V	0.03	0.01	0.01	0.01	<0.01			
Cr	0.81	0.70	0.70	0.70	0.65	0.50	10	70.00
Mn	<0.01	<0.01	<0.01	<0.01	<0.01			
Co	0.03	0.01	<0.01	0.01	<0.01			
Ni	0.06	0.06	0.06	0.06	0.05	0.40	10	40.00
Cu	0.02	0.01	0.01	0.01	0.01	2.00	50	100.00
Zn	0.05	0.03	0.03	0.03	0.03	4.00	50	200.00
Ga	0.01	0.05	0.05	0.04	0.05			
Ge	<0.01	<0.01	<0.01	<0.01	<0.01			
As	0.01	<0.01	<0.01	<0.01	<0.01	0.50	2	25.00
Se	0.02	0.01	<0.01	0.01	<0.01	0.10	0.5	7.00
Rb	1.79	3.3	3.2	2.5	2.9			
Sr	31	41	44	38	33			
Y	<0.01	<0.01	<0.01	<0.01	<0.01			
Zr	<0.01	<0.01	<0.01	<0.01	<0.01			
Nb	<0.01	<0.01	<0.01	<0.01	<0.01			
Mo	0.13	0.08	0.07	0.08	0.07	0.50	10	30.00
Cd	<0.01	<0.01	<0.01	<0.01	<0.01	0.04	1	5
Sn	<0.01	<0.01	<0.01	<0.01	<0.01			
Sb	<0.01	<0.01	<0.01	<0.01	<0.01	0.06	0.7	5
Cs	<0.01	0.07	0.05	0.03	0.05			
Ba	25	33	34	30	26	20.00	100	300.00
La	<0.01	<0.01	<0.01	<0.01	<0.01			
Ce	<0.01	<0.01	<0.01	<0.01	<0.01			
Pr	<0.01	<0.01	<0.01	<0.01	<0.01			
Nd	<0.01	<0.01	<0.01	<0.01	<0.01			
Sm	<0.01	<0.01	<0.01	<0.01	<0.01			
Eu	<0.01	<0.01	<0.01	<0.01	<0.01			
Gd	<0.01	<0.01	<0.01	<0.01	<0.01			
Tb	<0.01	<0.01	<0.01	<0.01	<0.01			
Ho	<0.01	<0.01	<0.01	<0.01	<0.01			
Er	<0.01	<0.01	<0.01	<0.01	<0.01			
Tm	<0.01	<0.01	<0.01	<0.01	<0.01			
Yb	<0.01	<0.01	<0.01	<0.01	<0.01			
Lu	<0.01	<0.01	<0.01	<0.01	<0.01			
Hf	<0.01	<0.01	<0.01	<0.01	<0.01			
Ta	<0.01	<0.01	<0.01	<0.01	<0.01			
W	<0.01	<0.01	<0.01	<0.01	<0.01			
Tl	<0.01	<0.01	<0.01	<0.01	<0.01			
Pb	<0.01	0.01	0.01	0.01	0.01	0.50	10	50.00
Bi	<0.01	<0.01	<0.01	<0.01	<0.01			
Th	<0.01	<0.01	<0.01	<0.01	<0.01			
U	<0.01	<0.01	<0.01	<0.01	<0.01			

Figures

Figure 1: SEM-EDX images showing the granitoid starting materials: (A) GW1, (B) GW2, (C) GW3, (D) GW4. Yellow marker indicates the EDX analysis for each sample.

Figure 2: SEM-EDX images showing the AP materials: (A) general area of AP product, (B) AP fixated with FA covering GW, (C) AP fixated with GW particles covering FA. Yellow marker indicates the EDX analysis for each sample, and green marker shows AP particles formed

Figure 3: Leaching yield arising from applying the EN12457-2 test to aggregate products obtained using different FA/GW ratios and compared with the non-hazardous limit. (A) COFA/GW, (B) LBFA/GW, and (C) SAFA/GW

Figure 4: Leaching yield of oxyanionic species arising from applying the EN12457-2

test to aggregate products obtained using different FA/GW ratios

Figure 5: Properties of the fresh concrete mixtures with GW/FA blend, compared to concrete mixture without partial replacement (A) Slump; (B) Specific weight; and (C) Air content.

Figure 6: Compressive strength and chloride ion penetration tests performed on the concrete mixtures. (A) compressive strengths after 1 and 28 days of casting; (B) ion penetration in the concrete containing the AP as partial substitute compared to standard concrete. *Standard deviation of 0.5MPa

Figure 7: X-ray diffraction of the $\text{Ca}(\text{OH})_2$ peak for two concrete mixture, one containing FA/GW3 as partial substitute to sand and another with no GW or FA added

Highlights

- The study evaluates the potential of using coal fly ashes and granitoid waste powder byproducts as partial substitute to natural sand (10%) in concrete.
- Fixation occurs via the aluminate and silicate anions at the surface of the fly ash – Ca^{2+} as mediators – silicate anions of the granitoid waste powder
- End products (aggregates and concrete) are considered to have better performances and were found as non-hazardous
- New and original method could reduce the environmental pollution by reducing the use of natural sources, waste storing and energy demand.