Geopolymers based on the valorization of Municipal Solid Waste Incineration residues

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Abstract

The proper management of Municipal Solid Waste (MSW) has become one of the main environmental commitments for developed countries due to the uncontrolled growth of waste caused by the consumption patterns of modern societies. Nowadays, municipal solid waste incineration (MSWI) is one of the most feasible solutions and it is estimated to increase in Europe where the accessibility of landfill is restricted. Bottom ash (BA) is the most significant by-product from MSWI as it accounts for 85 - 95 % of the solid product resulting from combustion, which is classified as a non-hazardous residue that can be revalorized as a secondary aggregate in road sub-base, bulk lightweight filler in construction. In this way, revalorization of weathered BA (WBA) for the production of geopolymers may be a good alternative to common reuse as secondary aggregate material; however, the chemical process to obtain these materials involves several challenges that could disturb the stability of the material, mainly from the environmental point of view. Accordingly, it is necessary that neoformed geopolymers are able to stabilize heavy metals contained in the WBA in order to be classified as non-hazardous materials. In this regard, the SiO_2/Al_2O_3 ratio plays an important role for the encapsulation of heavy metals and other toxic elements. The aim of this research is to formulate geopolymers starting from the 0 - 2 mm particle size fraction of WBA, as a unique raw material used as aluminumsilicate precursor. Likewise, leaching tests of the geopolymers formulated were performed to assess their environmental impact. The findings show that it is possible to formulate geopolymers using 100 % WBA as precursor, although more investigations are needed to sustain that neoformed geopolymer obtained can be considered as non-hazardous materials.

Keywords: Municipal Solid Waste Incineration; Valorization; Geopolymer; Bottom Ash; Heavy metals; Silica content

1. Introduction

Nowadays, within a circular economy, waste is a pressing environmental, social, and economic subject, which is gradually accepted and revalorized as a secondary resource. From the last 20 years, waste incineration (waste-to-energy plants, WtE) has grown progressively; hence, the incineration of municipal solid waste (MSWI) residues has become an appreciated alternative in waste management. Thermal energy recovery is a key in the management model of MSW in Europe. This allows the energy recovery contained in the MSW, decreasing the managed fraction in landfills, according to the management hierarchy and guidelines of Directive 2008/98 / EC [1]. The MSWI has increased by 60 % since the last 20 years [2], allowing reducing the waste mass and volume, and it generates energy over combustion. Although a substantial waste reduction can be obtained, during the MSWI process, about 20 % of the non-combustible residual material or bottom ash (BA) is collected at the furnaces exit, which mainly containes glass, ceramics, concrete, brick, rubble, sand, metal, stone, and fused clinker, ash, and slag, among others. BA are classified as non- hazardous waste (CRC 190101), and can be revalorized into secondary aggregates in road subbase, bulk lightweight filler for construction, cement bound materials applications, or civil engineering, previous weathering stabilization (WBA) during 2-3 months [3,4]. Prior to ageing process, freshly BA has to be conditioned by sieving processes and crushing rejection, magnetic separation of ferrous metals and separation of non-ferrous metals by eddy currents, and eliminating light components by blowing. The resulting WBA is rich in calcium oxide and silica, with minimum amounts of stabilized heavy metals. Around 45 % of the WBA weight has a particle size between 4 and 30 mm, being this mainly composed of glass and synthetic ceramics. The size below 4 mm, also presents glass as major material (primary and secondary glass) as well as most of the toxic and dangerous elements, mainly consisting of heavy metal oxides, which can be mobilized in the aqueous phase of a possible leaching process [5–7].

Although cement industry is one of the most polluting (responsible for about 5 % of CO_2 worldwide), Portland Cement (OPC) has been a major improvement for the society since it had been discovered in the 19th century. By this, cement industry must consider the task of finding new ways to reduce environmental pollution, consume

less energy, and reduce emissions. This can be achieved by developing other sustainable alternative cements with less greenhouse gas emissions and capable of incorporating waste in their formulations, contributing to the efficient strategic policy of the EU for 2050 [8].

The alkali activated materials (AAM) essentially comprise any binder system deriving from the reaction between an alkali hidroxide/silicate and an aluminumsilicate precursor [9]. It has to be emphasized that among AAM, there is a subclass named geopolymers, where the binder phase is a very coordinated aluminosilicate with pseudo-zeolitic amorphous structure [10–12]. Geopolymer consists of the reaction of a powder precursor, with high content in amorphous aluminosilicates, with strong alkaline activator solution, which general formula is $M_n[-Si-O_2]_z[-Al-O]_n·wH_2O$. These materials have an increasing interest in scientific and technological fields, and they are an excellent alternative to the extensive usage of OPC. Compared to OPC, they are more sustainable and have less environmental footprint, as well as, the required energy to be manufactured is less, generating between 60 and 80 % less CO_2 emissions [13].

Overall, the geopolymerization reaction is a polycondensation which starts when aluminosilicates are dissolved in the alkaline medium to form free silica tetrahedral and aluminum cations coordinated with oxygen. Tetrahedrons are joined by oxygen atoms to form a similar three-dimensional structure of a zeolite (-Si-O-Al-O-). The cation of the activator alkaline solution provides the positive charge that maintains the solution electroneutrality. Under proper temperature conditions, between 20 and 90ºC, and a certain curing time, a gel by ionic interaction solidifies into a solid tridimensional structure, which is the base of geopolymer. The mechanism can be summarized in the following steps [14]: dissolution of the amorphous phases in the alkaline solution, condensation of the monomers, and gelling. The precursors are oriented and gel viscosity increases to form the three dimensional network. Finally, monomers polymerize and crystallization takes place. Depending on the relation between alumina and silica, different monomers and polymers can be formed [15–18], as Poly(sialate) [Si-O-Al-O], Poly(sialate-siloxo) [-Si-O-Al-O-Si-O-], and Poly(sialate-disiloxo) [-Si-O-Al-O-Si-O-Si-O-], with SiO_2/Al_2O_3 proportions 2, 4, and 6 respectively. Accordingly, geopolymers properties are affected by the SiO_2/Al_2O_3, R_2O/Al_2O_3, and SiO_2/R_2O ratios, where R is sodium or potassium cation used to alkaline activation [15], among other parameters [19].

There are many characteristics of geopolymers, such as the ability to incorporate waste materials or by-products as raw materials, low emission of greenhouse gases due to their low synthesis temperature, low energy consumption, low cost by the use of waste materials as a raw material, good compressive strength, low fluid permeability, good chemical resistance [20], excellent fire resistance, stability at high temperatures [21], and durability. For these properties, geopolymers are an excellent alternative to OPC for construction materials, being already used in the formulation of structural elements and civil engineering with excellent results in some countries [22].

While any aluminosilicate with high silicon and aluminium content can be alkali-activated, the material also has to be soluble in a basic media; hence, the amorphous or vitreous phase of the material has to represent a substantial percentage of the total [23]. Geopolymers are a remarkable key for the valorization of by-products that have completed their life cycle, being these used as a secondary source in their second production cycle. By this, wastes which contain alumina and silica can be used as precursors to the formation of geopolymers, regardless of their alkali content. Accordingly, geopolymer can be formulated using a silicate or aluminosilicate as precursors, while alkaline sources can include any soluble substance supplying alkali metal cations, raise the pH of the mixture of reaction, and accelerate the dissolution of the solid precursor (such as alkali hydroxides, silicates, carbonates, sulfates or aluminates). There are several research studies on geopolymers which used a wide variety of waste materials for their formulation: metallurgical slag, fly ash from thermal power, red mud from aluminum industry, ash from rice husk combustion, vanadium mining waste, ashes from palm oil, waste from ceramic and glass industry, among others [8–14]. Their properties depend on the composition of these secondary sources and on the used alkaline ion, and especially, on the polycondensation conditions reaction. In this way, the amount of reactive fraction of WBA is primary significant to evaluate its possible use in alkaline activation process [24].

The aim of this study is a scientific, technological, sustainable, and environmental challenge focused on the development and synthesis of new geopolimers using WBA as a unique raw material used as aluminumsilicate precursor, for its subsequent reuse as secondary granular materials. The formulation will be performed using as precursor the finest fraction (< 2 mm) of WBA, in which the geopolimeric matrix can act as a binder for encapsulation (solidification) and immobilization of the heavy metals. The synthesis and formulations from 100 % WBA were carried out evaluating the geopolymers leaching potential and their environmental requirements for their use as secondary granular materials. On the other hand, considering only the finer fraction of the WBA, it will allow adjusting the particle size distribution of this secondary granular material to the optimal curve of Fuller [25].

2. Experimental procedure

2.1. Materials

WBA generated during the incineration of municipal solid wastes in waste-to-energy plants is composed of both amorphous and vitreous phases, being Si the most abundant element due to lime-soda glass (primary glass) and amorfous phases formed during incineration process (secondary glass) [26]. Si amount is followed by Ca, mainly from crystalline ceramic materials [27]. Accordingly, WBA can be defined as a silica-based powder with the proper chemical and physical characteristics for its use as the only precursor for geopolymers formulation, for its subsequent reuse as secondary granular materials or other. The only aluminosilicate solid precursor used for the present study derives from the 0 - 2 mm fraction of WBA, which was previously dried, milled, and used in the form of powder with grain size of 80 μm to enhance their reactivity and homogeneity.

Sodium hydroxide as alkaline solution was used in all formulations and it was prepared by dissolving pellets (99.6 wt %, Carlo Erba, Italy) in desionized water to obtain NaOH 8M. Besides, sodium silicate solution was used in all formulations as activator. Sodium silicate was used as a viscous liquid produced for the cement industry (Ingessil, Verona, Italy) with SiO_2/Na_2O molar ratio of 3 (27.09 wt% of SiO_2 and 8.85 wt% of Na_2O) and density of 1.373 $g \cdot cm^{-3}$. Therby, the alkaline solution was formed by mixing NaOH 8M and sodium silicate ($Na_2O:SiO_2=1:3$) solutions and stirred during 5 minutes. After that, WBA sample was added to the solution, mixed in a plastic container, until the paste was homogeneous, and subsequently cast into Teflon® molds.

Specimens of 40 mm diameter and 20 mm thickness were prepared. The samples were sealed from atmosphere for 24h, and the characterization was performed progressively with curing under room conditions (23ºC temperature and around 50 % of relative humidity). Different formulations were carried out considering several parameters such as elemental analysis (XRF) results of the WBA sample as well as the Si/Al and Na/Al content of the formulation, which have to be in the range 1.8-2.5 and 0.9-1.2 [28,29], respectively. It is important to highlight that the liquid to solid ratio in the formulation has to be as less as possible in order to improve the consolidation, which was evaluated by the integrity test of each formulation by submerging the sample during 24 h in water.

2.2. Methods

Different characterization techniques were simultaneously used to evaluate the WBA contained in the geopolymer specimens from the best formulations.

2.2.1. X-Ray Fluorescence (XRF)

X-ray fluorescence semi-quantitative analysis was performed on the calcined (550ºC) residue of WBA sample to study the presence of the major and minor elements in the form of the most stable oxides. This analysis is mandatory to proper prepare the formulations regarding the Si/Al and Na/Al ratios. A spectrophotometer Thermo ARL ADVANT'XP+X-ray fluorescence (XRF) sequential Spectrometer (Thermo Scientific) was used.

2.2.2. X-Ray Diffraction (XRD)

The different crystalline phases, contained in WBA, were qualitatively identified by X-ray diffraction in an X-Ray diffractometer PANAnalytical PW 3710 mpd control from Philips.

2.2.3. Thermogravimetrical Analysis (TGA)

Thermogravimetrical analysis (TGA) of WBA was performed by means of two different instruments: a NETZSCH 409 simultaneous TG/DTA and a Simultaneous SDTQ600 from TA Instruments, both analyses under static air atmosphere. The scanning rate was 10ºC·min^{-1} in a temperature range between 20ºC and 1200 - 1400ºC in both cases. The amount was kept in a mass range of 100 ± 1 mg using platinium 90 μL crucibles. The differences observed in the TGA studies were evaluated by assessing the weight loss (%) and Derivative Thermogravimetry (DTG) versus temperature.

2.2.4. Scanning Electron Microscopy (SEM)

The morphologies of the geopolymer formulations were characterized using an environmental scanning electron microscope (ESEM - Quanta200 - FEI, 20KV. Sputtering Au/Pd). The samples were stuck on the sample holder using a double-sided tape. The working conditions were high vacuum and high voltage (20 kV), and the image obtained by back scattered electrons. A chemical analysis was needed to evaluate the Si/Al parameter, accomplished with energy dispersion spectroscopy (EDS) coupled with SEM equipment.

2.2.5. Fourier transform infrared spectroscopy (FT-IR)

FT-IR spectroscopy is a powerful technique to evaluate and identify the chemical characterization of the main functional groups in the structure of the two final geopolymeric formulations under study (I and J). A Spectrum Two™ from Perkin Elmer supported by Dynascan™ interferometer and OpticsGuard™ Technology was used to perform the analyses. Liquid and solid state substances can be measured by this equipment in wavelength range between 4000 cm^{-1} and 350 cm^{-1}, by standard spectral resolution of 0.5 cm^{-1}.

2.2.6. Leaching test (EN 12457-4)

The heavy metals and metalloids release (As, Ba, Cu, Hg, Mo, Ni, Pb, total Cr, and Zn) values ($mg \cdot kg^{-1}$) of WBA as raw material and I and J formulations were determined using the batch leaching test in stirring distilled water for 24h with a solid/liquid ratio of 10 (EN-12457-4).

3. Results and discussion

3.1. Characterization of weathered bottom ash (WBA)

The chemical composition for the WBA sample obtained by XRF analysis is shown in Table 1, where it is shown that Si and Ca are the main elements followed by Fe, Na, Al, Mg and K with traces of chlorides and sulphates. Finally, the Loss of ignition (LOI) was calculated as the mass loss difference after calcining at 1100ºC, obtaining a value of 25.4 %.

Table 1. Average chemical composition of WBA.

Oxides	SiO_2	CaO	Fe_2O_3	Na_2O	Al_2O_3	MgO	K_2O	Cl	CuO	SO_3	TiO_2	P_2O_5	ZnO	LOI
BA (%)	43.3	16.9	14.1	7.58	5.80	2.22	1.11	0.14	0.23	0.65	0.35	1.97	0.18	25.4

Mineralogical analysis performed by means XRD for the 0 - 2 mm fraction of WBA showed the presence of muscovite, calcite, quartz, hydroxylapatite, hydrotalcite, calcium aluminium iron oxide carbonate hydroxide hydrate, and finally calcium sodium aluminum oxide as major cristallyne phases. In this sense, it is noticeable the heterogeneity of the WBA sample. Besides, in Figure 1 is shown the result of the TG analysis for the 80 μm (0 - 2 mm fraction) WBA as raw material. As it can be observed, the sample has four main steps with a total decomposition of 29 %, which denote the presence of organic matter, carbonates, and other hydroxides and hydrated salts, decomposing with temperature. Data obtained by TGA are in agreement with those measurements such as humidity performed from 25 to 110ºC, being 3.84 %, the organic material from 110 to 550ºC, 11.58 %, and carbonates from 550 to 860ºC, 10.43 %. Also the value of loss of ignition (LOI) is in line with the previous measurement.
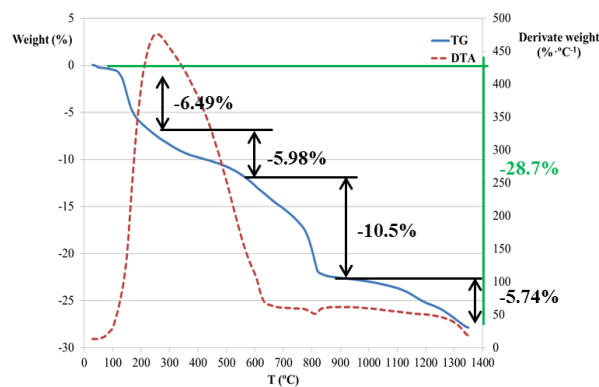

Figure 1. TGA result for the WBA raw material.

The FT-IR spectrum of WBA raw material is shown in Figure 2. It can be observed a peak around 1400 cm^{-1} which is related to carbonates. Besides, in the WBA spectrum before the alkali activation, it can be denoted an asymmetrical stretching peak around 1000 cm^{-1} because of the Si–O–Si and Si-O-Al bonds.
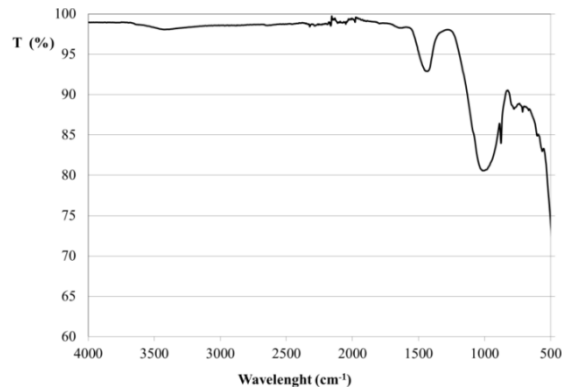
Figure 2. WBA FT-IR spectrum.

3.2. Characterization of geopolymers

From all the specimens formulated, only two formulation were selected, as shown in Table 2, with the optimum Si/Al and Na/Al ratios and proper results regarding the integrity tests.

Table 2. Optimal geopolymer formulations, according the Si/Al and Na/Al ratios and integrity test results.

	WBA (g)	NaOH 8 M (ml)	NaSiO$_3$ (ml)	H$_2$O (ml)	Si/Al	Na/Al	integrity test after 24 h
I	18	1	14	5	2.56	1.51	Consistent
J	18	2	12	5	2.33	1.51	Consistent

Figure 3 presents the micrographies for the I and J specimens, showing not dense microstructure, but geopolymerization occurs because wide areas of geopolymeric gel are evident. Besides, in I and J samples cured during 15 days more unreacted particles are evident with respect to the other samples corresponding to longer time of curing.
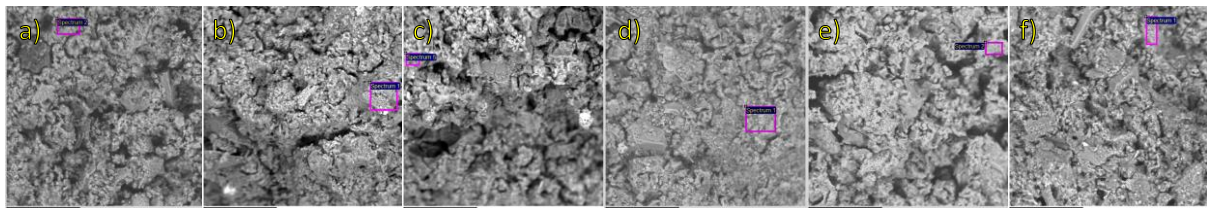

Figure 3. Micrographies for different geopolymeric samples a) I and b) J after 15 days of curing; c) I and d) J after 2 months of curing; e) I and f) J after 3 months of curing.

The chemical analysis performed by means EDS showed that all the Si/Al values are higher with respect to the above mentioned in Table 2 as well as the theoretical ones, but with the same order of magnitude. This fact can be associated because silicon is more rapid in the dissolution while Al is slow; it means the difficulty in dissolving crystalline phases containing aluminum.

The TG analysis shows very similar curves for I and J formulations, as it is shown in Figure 4. The curves show water loss for T < 200ºC, dehydration from thermal decomposition of hydroxides in the range 200 - 500ºC, corresponding to the not completed geopolimerization process and unreacted hydroxides contained in WBA and loss of carbonates in the range 600 - 900ºC, also present in the mineral phases of the WBA.
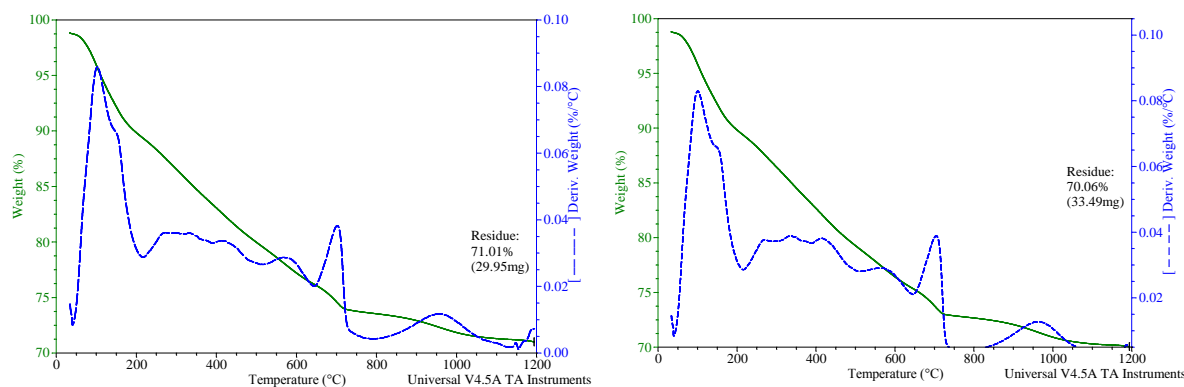
Figure 4. TGA results for the I and J formulations at 3 months of curing.

A comparison between the spectra of specimen I after one week and after three months of curing is shown in Figure 5a. In the same manner, the comparison between one week and three months for the J specimen is presented in Figure 5b. As it can be observed in both figures, the spectra show a band around 1400 cm^{-1}, which is typical from carbonates, resulting mainly from the carbonation of Ca^{2+} with atmospheric CO$_2$ during the weathering process. The presence of this phase in geopolymers derives from the composition of the used precursor. Besides, it can also be observed the asymmetrical stretching peak at 947.58 cm^{-1} due to the Si–O–Si and Si-O-Al formations. This alteration to inferior values (from 1000 to 947.58 cm^{-1}) has been reported elsewhere [30,31], suggesting a matrix chemical change in the geopolymer related to the degree of geopolimerization associated to the presence of tetrahedrally-coordinated Al in the Si-O-Si skeletal structure. Signal around 900 cm^{-1} can be identified as Al(VI)-OH stretching vibrations, although it can be considered as low geopolymerization degree in a sample. Finally, the peak around 875 cm^{-1} is also related to the presence of CaCO$_3$ in the samples analyzed.
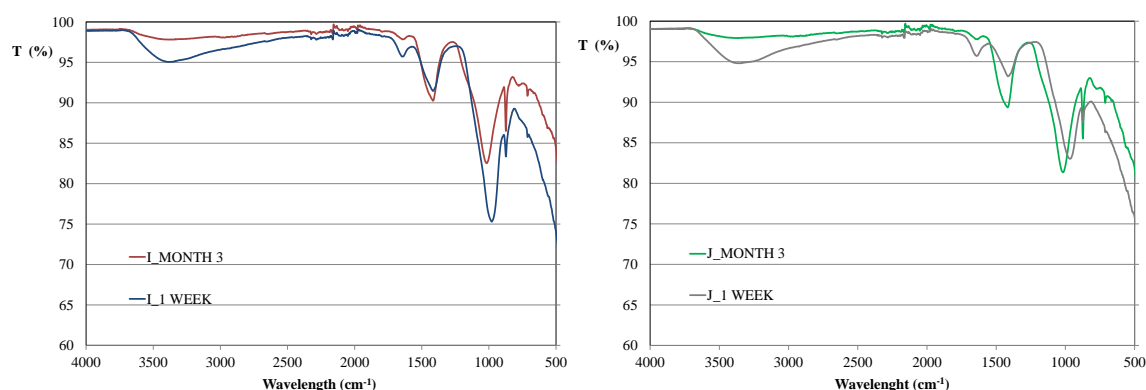

Figure 5. Comparison of FT-IR spectra for 3 months and 1 week of a) I specimen b) J specimen.

Furthermore, chemical analysis were performed to the I and J specimens. Table 3 shows some results regarding pH, conductivity, and amount of chlorides and sulphates released from the final two selected formulations (I and J) in the EN-12457-4 leached. As it was expected, pH was higher than WBA (controlled by calcium carbonate) because of the alkali behavior of the geopolymeric samples. Because of the amount of cations and anions in the solution, conductivity value was high. Finally, chlorides and sulphates concentrations were higher than their limits, which are 15000 and 20000, respectively.

Table 3. Results from leaching test (EN-12457-4) to I and J geopolymers.

	pH	k (mS·cm^{-1})	Cl- (mg·kg^{-1})	SO$_4^{2-}$ (mg·kg^{-1})
I	11.57	11.06	2707	3922
J	12.14	15.54	973	48816

The leaching test results of WBA regarding the heavy metals release [32] are presented in Table 4. The limits and the criteria of non-hazardous wastes acceptance in landfills [33] are also presented. Besides, it is also shown the leaching test results for the WBA 0 - 2 mm (without milling), which denoting values below the permitted limits for its reuse as secondary aggregate [34]. Likewise, for the I and J specimens, it can also be observed that all the results remain below the threshold established by landfill classification and revalorization legislations, except for the As. Accordingly, regarding the content of heavy metals and metalloids, it is important to highlight

that the encapsulation of heavy metals succeed as it was expected, despite the worst fraction of the WBA (0 - 2 mm) was used as unique aluminosilicate precursor.

Table 4. Analysis of the heavy metals and metalloids' content in mg·kg^{-1} by EN 12457-4 leaching test.

	As	Ba	Cr	Cu	Hg	Mo	Ni	Pb	Zn
Non hazardous limits	2.00	100.00	10.00	50.00	0.20	10.00	10.00	10.00	50.00
WBA	0.21	0.39	0.33	3.46	<0.01	0.77	0.06	0.01	<0.20
I	2.25	11.45	1.78	17.60	<0.01	1.64	10	0.75	16.4
J	2.41	3.62	1.28	23.5	<0.01	1.87	10	0.40	18.7

4. Conclusions

It has been achieved the formulation of geopolymers using 100 % WBA, with the size frection 0 - 2 mm particle size, as unique aluminisilicate precursor. An exhaustive characterization was performed into the WBA as raw material, as well as to the two specimens under study (I and J). The results from the calcination of the WBA are in agreement with those regarding the TG analysis. The XRF analysis denoted that Si and Ca were the two major elements. In this sense, leaching tests of the geopolymers were performed to assess their environmental impact. All the results, except As, remain below the threshold established by landfill classification and revalorization legislations. Then, it is obtained a succesfull encapsulation of heavy metals, although it was used the WBA fraction with higher content of heavy metals and metalloids (0 - 2 mm).

In conclusion, the findings show that it is possible to formulate 100 % WBA geopolymers, while more studies are required to validate that neoformed geopolymer obtained can be considered as non-hazardous material.

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