

Heavy metal chemical fractionation and immobilization in lightweight aggregates produced from mining and industrial waste

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ABSTRACT: The fractionation of five heavy metals in a washing aggregate sludge, a sewage sludge, a clay-rich sediment, the mixtures of these materials and the lightweight aggregates manufactured with them has been determined by applying the optimized European Community Bureau of Reference sequential extraction procedure in order to evaluate the effects of the heating process on the extraction of these elements. Additionally, preparation of eluates by aggregate leaching has been performed in accordance with the UNE-EN-1744-3 standard. The availability of all the studied heavy metals has been reduced by the thermal treatment, since most of the heavy metals have become part of the undigested material in the lightweight aggregates. Nickel has been the heavy metal that has presented the highest concentration in the eluates obtained after completion of the single extraction procedure in the lightweight aggregates. The studied lightweight aggregates may be used in lightweight concrete manufacturing from the standpoint of heavy metal leaching.

Keywords: Availability; Leaching; Sequential extraction procedure; Single extraction procedure; Thermal treatment

INTRODUCTION

The occurrence and prevalence of Heavy metals (HMs) contamination in the Earth have been increased by anthropogenic activities such as mining and wastewater purification. The properties of metals in soils and other substrates (fly ash, sewage sludge, etc.) depend on the physic-chemical form in which they occur (Gleyzes *et al.*, 2002; Nouri *et al.*, 2009). In the element analysis, the total concentrations are usually evaluated (Kapoor and Viraraghavan, 1998; Dantas *et al.*, 2003; Chen *et al.*, 2010). However, they do not provide sufficient information about the bioavailability and toxicity of metals, since changes in the environmental conditions (temperature, pH, redox potential or organic ligand concentration) cause selective release of the total metal content from the solid to the liquid phase (Sahuquillo *et al.*, 2003). Thus, the interaction with sediments or soils and the bioavailability or toxic effects of the metals can only be studied by determining their chemical partitioning (Quevauviller, 1998). Sequential extraction procedures

(SEPs) are operationally-defined methods that are widely applied for assessing this chemical partitioning and the mobility of elements in soils and waste materials (Guevara-Riba *et al.*, 2004). These SEPs are based on the use of selective reagents chosen to successively dissolve the different mineralogical fractions thought to be responsible for retaining the larger part of the trace element (Gleyzes *et al.*, 2002). The optimized European Community Bureau of Reference (BCR) is a sequential extraction procedure (BCR-SEP), established by a group of expert laboratories, that is the only one which allows validation based on two certified reference materials (Rauret, 1998; Gómez-Ariza *et al.*, 2000; Davidson *et al.*, 2006). This method has been described in detail in another study (Sahuquillo *et al.*, 1999).

Among techniques that may be used to reduce or nullify the toxicity of wastes, ceramization and encapsulation represent the two main strategic options, but there is a basic difference between them: in the case of ceramization, the residue becomes an integral (chemical) part of the new product and its future

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behaviour will be good. On the contrary, when encapsulation occurs, the residue is physically retained, no chemistry changes occur and the future evolution will be more dependent on the environment and/or the conditions than on its stability and resistance (Castells, 2000).

According to Wunsch *et al.* (1996), the use of thermal treatment can produce the inertisation of HMs in residues of waste incineration against leaching. Lightweight aggregates (LWAs) from different types of residues with concentrations of HMs which exceeded regulations have been manufactured by Huang *et al.* (2007). After sintering, only trace amounts of HMs were detected in the products. However, Cheeseman *et al.* (2005) has shown that, under acid conditions and at relatively low sintering temperatures, there are very little differences in metal leaching between raw materials and sintered product. Therefore, the leaching evaluation of products such as LWAs manufactured with waste potentially contaminated by HMs should be performed under a wide range of conditions (low pH, changes in redox potential, etc). This work is a part of a more general research project. The first part has been previously reported (González-Corrochano *et al.*, 2009) and describes the manufacturing process of artificial lightweight aggregates by pelletizing and sintering of washing aggregate sludge (W), sewage sludge (SS) and a clay-rich sediment (C). It also shows the chemical and physical properties of the raw materials and products. The objectives of the research presented here are: 1) To determine the concentrations of the HMs associated with the different fractions of the raw materials used in the manufacturing of artificial LWAs in the previous work ; 2) To determine the chemical fractionation of these elements in the LWAs through the BCR-SEP; 3) To evaluate the effect of the ceramic process on the availability of the heavy HMs found in the raw materials, by means of the Extraction ratio (ER); 4) To determine whether the products comply with current regulations on construction materials and the leaching of toxic elements. This research has been carried out in the School of Environmental Sciences, University of Castilla-La Mancha (Toledo; Spain), from January to July 2010.

MATERIALS AND METHODS

Studied samples

Two types of artificial LWAs resulting from a previous study (González Corrochano *et al.*, 2009) have

been selected: 1) W75S25-1225-15, manufactured with a mixture (W75S25) of 75 % (wt) W and 25 % (wt) SS, fired at 1225 °C for 15 min in a rotary kiln (Nannetti®; TO-R 120-14 model); 2) W50C50-1200-15, produced from a mixture (W50C50) of 50 % (wt) W and 50 % (wt) C, heated at 1200 °C for 15 min. They present quartz and plagioclases (González Corrochano *et al.*, 2011). The chemical fractionation of W, SS, C and the mixtures has been also tested.

Optimized BCR sequential extraction procedure (BCR-SEP)

The operating conditions and the geochemical fractions obtained in each step of the optimized BCR-SEP are shown in the Fig. 1. The concentrations of five HMs in the extracts (F1, F2, F3 and F4) have been measured by inductively coupled plasma-mass spectroscopy (ICP-MS, Thermo Electron, XSeriesII).

In general, the availability of the elements associated with the different fractions decreases in the following order: F1 > F2 > F3 > F4 (Ma and Rao, 1997; Zemberyova *et al.*, 2006). The sum of F1, F2 and F3 ($\Sigma 1+2+3$), also called “non-residual fraction”, is considered to be the concentration that can be released under specific environmental conditions (changes in pH and potential redox). On the contrary, the elements associated with F4 are considered to be those that can not be released under the environmental conditions found in nature (Dang *et al.*, 2002). The sum of F1, F2, F3 and F4 ($\Sigma 1+2+3+4$) is also called “pseudo-total concentration”, since the elements in the undigested phases are not included.

In each step, the extraction ratio has been calculated following the equation:

$$ER = \frac{[mixture_{x,y}]}{[LWA_{x,y}] * \left(1 - \frac{LOI}{100}\right)} \quad (1)$$

Where $[mixture_{x,y}]$ and $[LWA_{x,y}]$ are the concentrations (mg/kg), of the element x, in the fraction y, in the mixture and in the LWAs, respectively; LOI is the loss on ignition (%) of the mixtures during the manufacturing of the LWAs in the rotary kiln. LOI values of W75S25 and W50S50 have been 24.26 % and 10.68 %, respectively (González Corrochano *et al.*, 2009).

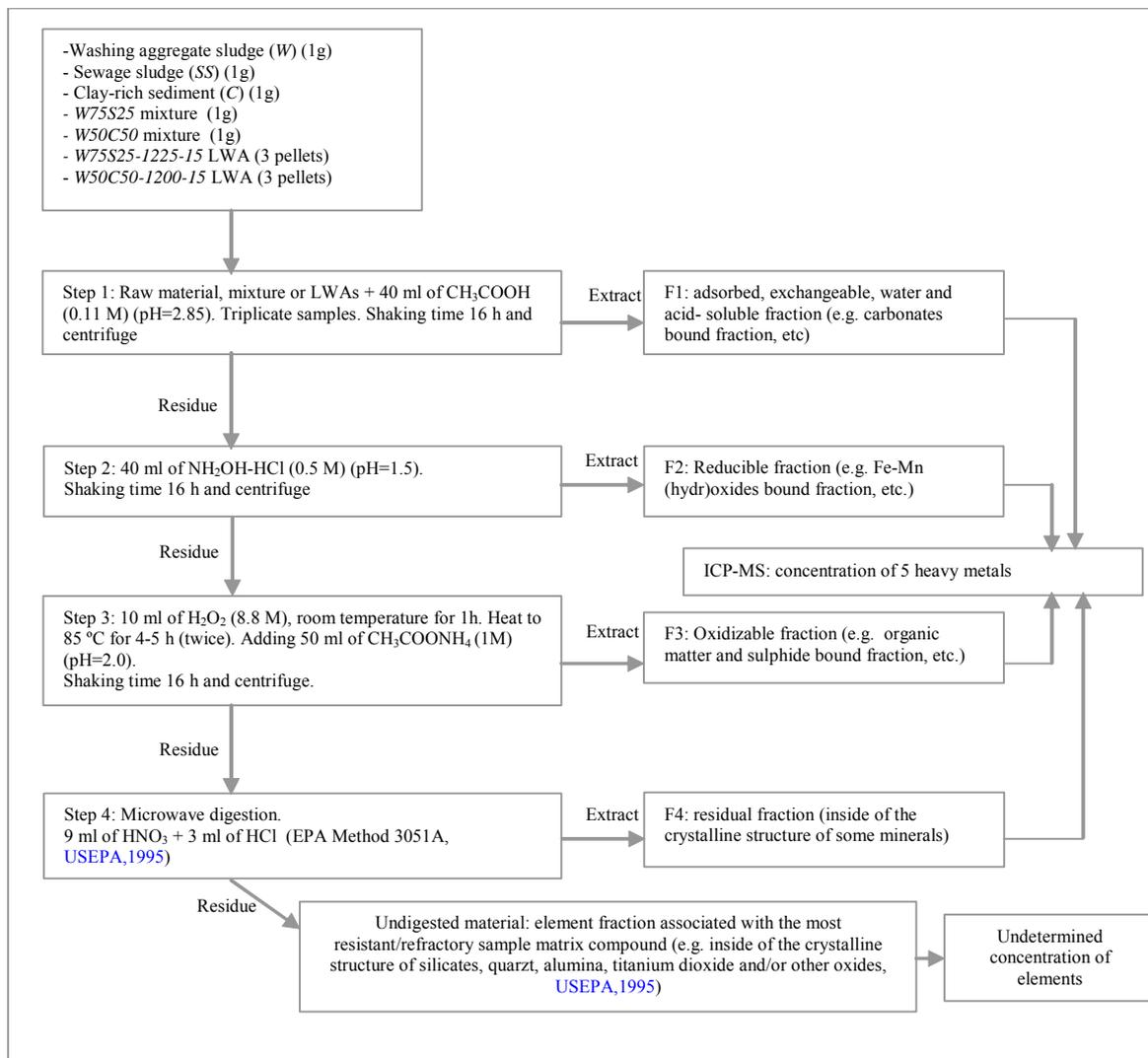


Fig. 1: Flow diagram of the operating conditions and the geochemical fractions obtained in each step of the optimized BCR sequential extraction procedure

Preparation of eluates by aggregate leaching

The test has been performed in accordance with the [UNE-EN-1744-3 standard \(2003\)](#). The liquid: solid ratio (wt) used has been 20:1. The concentrations of the same HMs have been measured by ICP-MS (Thermo Electron, *XSeriesII*).

RESULTS AND DISCUSSION

Optimized BCR sequential extraction of heavy HMs in the raw materials

Knowledge of the HM concentrations associated with the most labile fractions (F1, F2 and F3) of the

washing aggregate sludge, the sewage sludge and the clay-rich sediment is of great importance, since *W*, *SS* and *C* are exposed to different environmental agents that can change the conditions of the sites where the materials are located and produce variations on the element availability associated with each fraction.

In *W*, the HM with the highest concentration associated with the adsorbed, exchangeable, water-and-acid-soluble fraction (F1) has been Ni ([Table 1](#)). In *SS*, it has been Zn ([Table 2](#)). In *C*, Ni has also been the HM bound to F1 in the highest concentration ([Table](#)

3). Consequently, these two elements are the HMs which exhibit the highest available quantities when changes in pH occur (Table 4).

The HMs with the highest concentrations associated with the reducible and oxidizable fractions (F2 and F3, respectively) have been Cr, in *W* (Table 1), and Zn, in *SS* (Table 2) and in *C* (Table 3). Therefore, if changes in the redox conditions occur, Cr and Zn are the HMs which exhibit the highest available concentrations (Table 4). The major changes in the behaviour of redox-controlled trace elements typically occur at the interface between water and the solid phase, because this interface marks the change from an anoxic to an oxic medium (Schlieker *et al.*, 2001). Thus, when ponds of *W* or landfills are soaked by rainwater and then dried, these metals may be released to the environment.

Cr has also been the HM bound to the residual fraction (F4) in the highest concentration in *W* (Table 1), and Zn has been the one with the highest concentration in *SS* (Table 2) and in *C* (Table 3).

Since Cd presents higher environmental mobility than most HMs, it is efficiently absorbed by plants and bioaccumulative by aquatic and terrestrial organisms (Moreno Grau, 2003); it is important to note

the low concentrations of this element are associated with F1, F2 and/or F3 (Tables 1-3).

In all the raw materials, the HMs studied were mainly associated with F4 (Tables 1-3); therefore, the concentrations that may return to the environment are lower than if these elements have been primarily bound to any of the other three fractions.

In order to evaluate the potential agricultural use, the pseudo-total concentrations of HMs in *W*, *SS* and *C* have been compared to the limit values specified in Spanish Royal Decree 1310/1990, regulating the use of sludge from sewage treatment plants in the agricultural sector. Table 5 (2nd, 3rd and 5th columns) shows that *W* has pseudo-total concentrations of HMs below the limit values specified for the use of this residue for cultivation. On the contrary, agricultural application of *SS* is not possible, since this residue shows pseudo-total concentrations of most of the studied HMs above the established limit values (Table 5; 2nd, 3rd and 6th columns). Agricultural use of *C* is possible only if the soil pH is greater than 7 because its pseudo-total concentration of Ni has been higher than the limit value for agricultural soils with pH less than 7 (Table 5; 2nd, 3rd and 7th columns).

Table 1: Concentration (mg/kg) of each element associated with the different geochemical fractions in the washing aggregate sludge

Raw material		Washing aggregate sludge (<i>W</i>)					
Geochemical fraction element (mg/kg)	1 (Exchangeable, soluble)	2 (Reducible)	3 (Oxidizable)	4 (Residual)	Σ 1+2+3 (Non-residual)	Σ 1+2+3+4 (Pseudo-total)	
(HMs)	Cr	0.09 (0.12)	5.13 (6.59)	4.65 (5.97)	68.02 (87.33)	9.86 (12.67)	77.88
	Ni	0.64 (3.37)	1.72 (9.09)	1.08 (5.70)	15.47 (81.84)	3.43 (18.16)	18.90
	Cu	0.08 (0.46)	2.15 (13.02)	0.13 (0.79)	14.17 (85.73)	2.36 (14.27)	16.53
	Zn	0.11 (0.17)	3.07 (4.86)	0.89 (1.40)	59.17 (93.56)	4.07 (6.44)	63.24
	Cd	0.04 (17.50)	0.02 (10.46)	- - -	0.17 (72.04)	0.06 (27.96)	0.23

In brackets = pseudo-total concentration percentage. In gray = Fraction to which the element is mainly associated. - - - = Below the detection limit

Table 2: Concentration (mg/kg) of each element associated with the different geochemical fractions in the sewage sludge

Raw material		Sewage sludge (<i>SS</i>)					
Geochemical fraction element (mg/kg)	1 (Exchangeable, soluble)	2 (Reducible)	3 (Oxidizable)	4 (Residual)	Σ 1+2+3 (Non-residual)	Σ 1+2+3+4 (Pseudo-total)	
(HMs)	Cr	6.66 (7.60)	3.40 (3.88)	35.23 (40.21)	42.32 (48.30)	45.29 (51.70)	87.61
	Ni	5.02 (11.29)	2.34 (5.27)	4.72 (10.60)	32.43 (72.85)	12.09 (27.15)	44.51
	Cu	19.78 (5.38)	15.15 (4.12)	81.60 (22.19)	251.19 (68.31)	116.54 (31.69)	367.72
	Zn	139.89 (18.81)	141.43 (19.02)	108.14 (14.54)	354.12 (47.72)	389.46 (52.38)	743.59
	Cd	0.24 (15.33)	0.23 (14.97)	0.21 (13.44)	0.86 (56.25)	0.67 (43.75)	1.54

In brackets = pseudo-total concentration percentage. In gray = Fraction to which the element is mainly associated

In Spain, there is no legislation on the use of *W*, *SS* and/or *C* in the manufacturing of LWAs; consequently, the results obtained have been compared to the maximum levels established in other European countries (Cappuyens and Swennen, 2009). Both raw materials have HM values below the established limits, as may be observed in Table 5 (4th, 5th, 6th and 7th columns); therefore, *W*, *SS* and *C* may be used for the production of LWAs.

In the lightweight aggregates

The concentrations of Cr associated with the four

fractions and its pseudo-total concentration was decreased in both types of LWAs (ERs>1, Figs. 2a and 3a). Since the boiling point of Cr is approximately 2665 °C, higher than the heating temperatures, volatilization of this element can not take place. The thermal treatment produces changes in the LWAs, such as oxidation of organic matter, reaction of Fe₂O₃, decarbonation of calcium carbonate (González-Corrochano *et al.*, 2009) and formation of new crystalline phases (González-Corrochano *et al.*, 2011), which, in turn, cause changes in the redox potential

Table 3: Concentration (mg/kg) of each element associated with the different geochemical fractions in the clay-rich sediment

Raw material	Clay-rich sediment (C)					
Geochemical fraction element (mg/kg)	1 (Exchangeable, soluble)	2 (Reducible)	3 (Oxidizable)	4 (Residual)	Σ 1+2+3 (Non-residual)	Σ 1+2+3+4 (Pseudo-total)
Cr	0.07 (0.14)	0.96 (1.97)	1.35 (2.77)	46.25 (95.12)	2.37 (4.88)	48.63
Ni	0.38 (1.03)	2.25 (6.03)	1.61 (4.33)	33.00 (88.61)	4.24 (11.39)	37.24
(HMs) Cu	0.10 (0.26)	3.38 (9.02)	1.03 (2.75)	32.94 (87.97)	4.50 (12.03)	37.45
Zn	0.18 (0.14)	3.65 (2.83)	2.55 (1.97)	122.74 (95.06)	6.38 (4.94)	129.12
Cd	---	---	---	0.23 (100.00)	ND	0.23

In brackets = Pseudo-total concentration percentage; In gray = Fraction to which the element is mainly associated. - - - = Below the detection limit. ND = Not determined

Table 4: HMs that might present the highest available concentrations when changes in the environmental conditions occur

Raw materials and LWAs Potential environmental changes	Washing aggregate sludge (W)	Swage sludge (SS)	Clay-rich sediment (C)	W75S25-1225-15	W50C50-1200-15
pH (Highest F1)	Ni (0.64)	Zn (139.89)	Ni (0.38)	Ni (0.90)	Ni (0.91)
Redox conditions (Highest Σ F2+F3)	Cr (9.78)	Zn (249.57)	Zn (6.2)	Zn (1.25)	Cu ≈ Ni ≈ Zn (≈0.15)
pH + redox conditions (Highest Σ F1+F2+F3)	Cr (9.86)	Zn (389.46)	Zn (6.38)	Zn (1.82)	Ni (1.06)

In brackets = Maximum potential available concentration (mg/kg)

Table 5: Comparison between the concentrations of HMs in the washing aggregate sludge, sewage sludge and clay-rich sediment and the limit values for different reuses of these wastes

Element	Limit value for agricultural Soils pH soils <7 (mg/kg)	Limit value for agricultural soils pH soils > 7 (mg/kg)	Limit value for reuse in building materials (mg/kg)	Pseudo-total concentration in the W (Table 1) (mg/kg)	Pseudo-total concentration in the SS (Table 2) (mg/kg)	Pseudo-total concentration in the C (Table 3) (mg/kg)
Cr	100	150	1250	77.88	87.61	48.63
Ni	30	112	250	18.90	44.51	37.24
Cu	50	210	375	16.53	367.72	37.45
Zn	150	450	1250	63.24	743.59	129.12
Cd	1	3	10	0.23	1.54	0.23

and the pH. The Cr mobility is affected by these factors (Moreno Grau, 2003); therefore, the non-residual and residual Cr may have undergone geochemical changes and become a part of the undigested material.

The undigested material may be composed of plagioclases, amorphous / vitrified material (LWA matrix) and other minerals in quantities undetectable by X-ray diffraction (XRD). The chromium atom substitutions in the structure of plagioclases have not been reported (Deer et al., 1992); consequently, the Cr associated with the non-residual and residual fractions of the raw materials may have become a part of the matrix of the LWAs. This is supported by the fact that aggregates designed for construction have been satisfactorily manufactured from vitrified chromium-contaminated soils (Meegoda et al., 1996). As regards chromium, the heating treatment has produced a decrease in the Ni and Cu associated with all the fractions in W75S25-1225-15 (ERs>1,

Figs. 2b and 2c). This decrease has been also observed in the reducible, oxidizable and residual Ni and Cu concentrations in W50C50-1200-15 (ERs>1, Figs. 3b and 3c). Volatilization of these elements is also impossible (boiling points over 2500 °C); therefore, they must have undergone geochemical changes. In the first type of LWAs, Ni and Cu must have become a part of the undigested material. In the second type, transformation of the Ni and Cu to more exchangeable/soluble forms may have occurred, since the concentrations associated with F1 increase after the thermal treatment (ER=0.54, Fig. 3b; ER=0.00, Fig. 3c). This notwithstanding, a proportion of reducible, oxidizable and residual Ni and Cu must have become a part of the undigested material, since the pseudo-total concentration has been decreased after the heat treatment (ER=1.42, Fig. 3b; ER=4.72, Fig. 3c).

Important decreases in the concentrations of Zn associated with F1, F2 and F3 in W75S25-1225-15

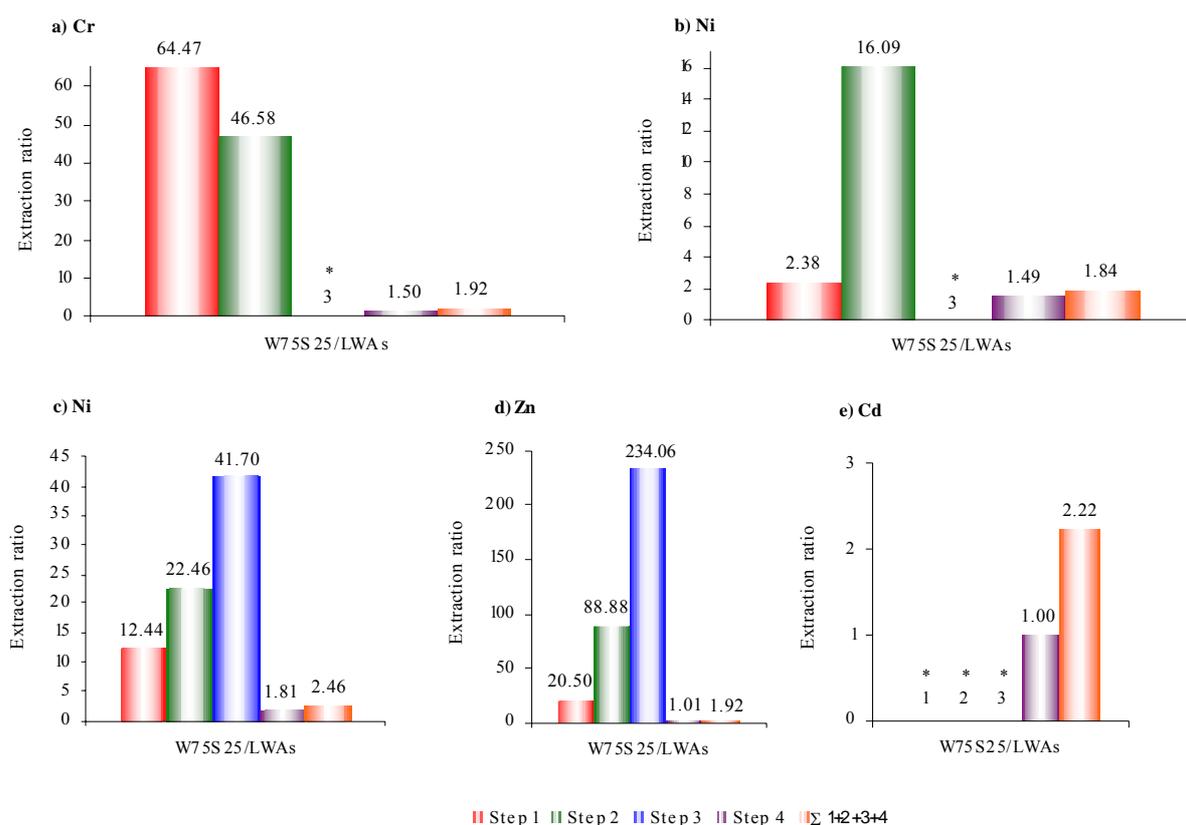


Fig. 2: Extraction ratios between W75S25/LWAs. a) Cr; b) Ni ; c) Cu; d) Zn; e) Cd. (*): Concentration in the LWAs is below the detection limit in this fraction (number)

have been produced by the thermal treatment (Fig. 2d). In W50C50-1200-15, the heat treatment has caused the largest decrease in the concentration of Zn associated with F3 (Fig. 3d). Unlike Cr, Ni and Cu, part of the Zn associated with the above-mentioned fractions may have volatilized, since its boiling point is approximately 900 °C. In the LWAs produced with W and C, the reducible, oxidizable and residual Ni may have been converted into a more soluble form, since the concentration associated with F1 has been increased (ER=0.75, Fig. 3d). The pseudo-total concentrations in W75S25 and W50C50 decreased after the heating process, but, as Zn may have volatilized, the increase in this element in the undigested material can not be verified, unlike in the case of Cr, Ni and Cu.

It is worth noting that the concentrations of Cd associated with F1 and F2 decreased after the thermal treatment in the LWAs manufactured with W and SS

(Fig. 2e). This decrease has not been proven in the LWAs produced with W and C, since the concentration of Cd in W50C50 has been undetectable. As Zn, it may be due to Cd volatilization during the heating process, since its boiling point is approximately 765 °C. The $\Sigma 1+2+3+4$ concentrations of both mixtures decreased after the ceramic process (ER>1, Figs. 2e and 3e), however, the increase in the concentration of Cd in the undigested material has not been verified, as in the case of Zn. An overview of the concentration trends in the non-residual fraction (the potential availability) and in the residual fraction may be observed in Table 6. It has been possible to establish that all the studied HMs have been elements whose non-residual concentrations decreased after the thermal treatment. They have been less available in the LWAs than in the raw materials; consequently, the heating process has been beneficial to the environment.

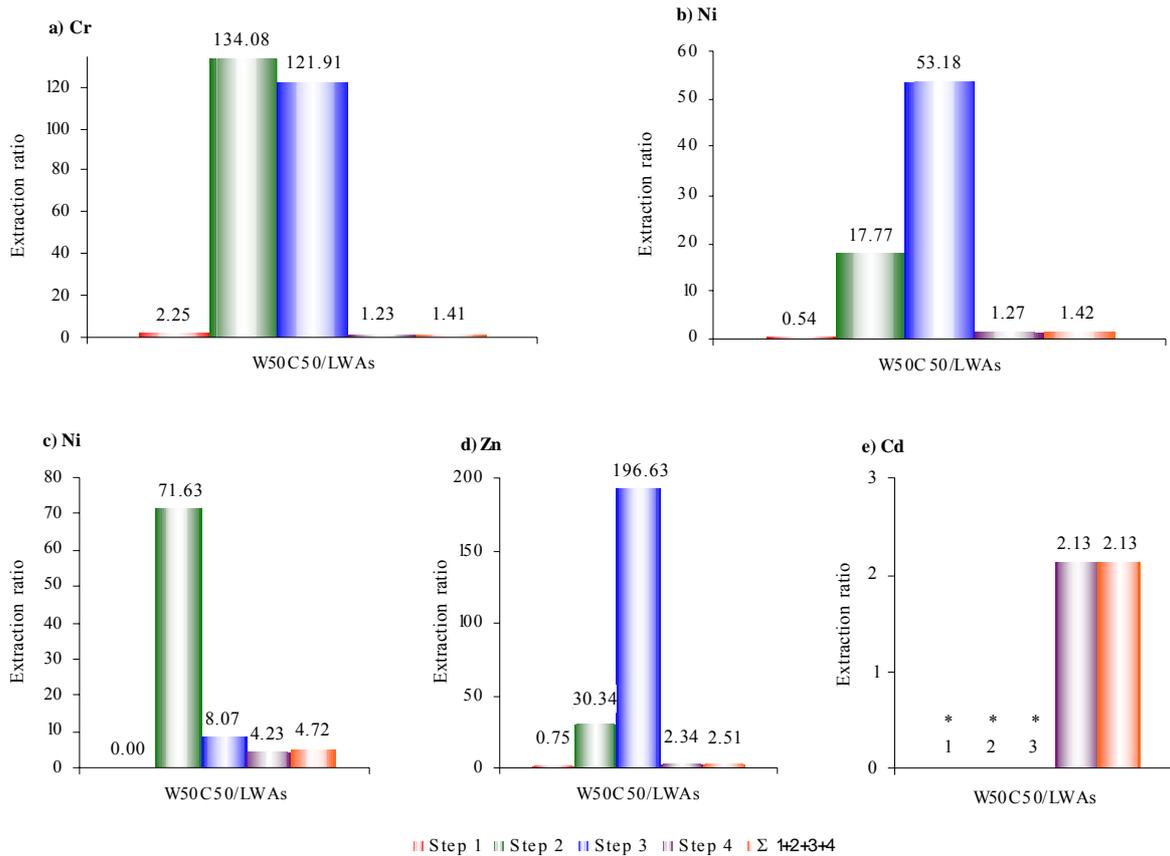


Fig. 3: Extraction ratios between W50C50/LWAs. a) Cr; b) Ni ; c) Cu; d) Zn; e) Cd. (*): Concentration in the LWAs is below the detection limit in this fraction (number)

Chemical fractionation and immobilization in lightweight aggregates

Table 6: Extraction ratio values (ER), for each element, in the non-residual and in the residual fraction ($\Sigma 1+2+3$ and F4, respectively)

LWAs ER fraction	W75S25-1225-15		W50C50-1200-15	
	1.00 kg mixture / 0.76 kg LWA ^(a) $\Sigma 1+2+3$	F4	1.00 kg mixture / 0.89 kg LWA ^(b) $\Sigma 1+2+3$	F4
Cr	159.79 (-19.31)	1.50 (-22.27)	60.52 (-7.95)	1.23 (-9.97)
Ni	7.70 (-5.70)	1.49 (-7.05)	3.85 (-2.67)	1.27 (-4.32)
HM's Cu	27.80 (-24.44)	1.81 (-28.45)	15.01 (-3.12)	4.23 (-14.82)
Zn	74.65 (-101.97)	1.01 (-1.16)	14.54 (-6.27)	2.34 (-44.86)
Cd	>1 (-0.27)	1.00 (0.00)	ND	2.13 (-0.10)

In brackets: Concentration difference (mg/kg) between mixtures and LWAs (taking in account the LOI of mixtures). ND: Not determined because concentrations in the mixtures and in the LWAs are below the detection limit ^(a) LOI = 24.26 % (González-Corrochano *et al.*, 2009); ^(b) LOI = 10.68 % (González-Corrochano *et al.*, 2009)

Table 7: Obtained concentrations in the eluates and comparison between them and the limit values for inert wastes (according to Decision 2003/33/CE, European Council, 2003)

LWAs element (mg/kg) ^(a)	W75S25-1225-15	W50C50-1200-15	Limit values for inert wastes (mg/kg) ^(b)
Cr	0.23	0.04	0.5
Ni	2.08	0.57	0.4
HM's Cu	0.21	0.05	2
Zn	---	0.02	4
Cd	---	---	0.04

--- = Below the detection limit; ^(a) Liquid:solid ratio (wt) = 20:1, ^(b) If the liquid:solid ratio is 10:1

Leaching behaviour of HM's in the lightweight aggregates

After completion of the single extraction, Cr, Ni and Cu have been detected in the eluates from W75S25-1225-15 (Table 7). Cr, Ni, Cu and Zn have been found in the eluates from W50C50-1200-15 (Table 7).

The Spanish code for structural concrete (EHE-08, 2009) does not specify maximum concentration levels in the leachate from LWAs. Consequently, the studied LWAs may also be used in lightweight concrete manufacturing, from the standpoint of HM leaching. Nevertheless, a comparison between the obtained results and the limit values for inert wastes (European Council, 2003) has been carried out in Table 7. With the exception of Ni in both types of LWAs, all the studied HM's have presented concentrations below the established limits for a 10:1 liquid: solid ratio. The fact that the Ni concentration values have been higher than the limits does not necessarily mean that the LWAs can not be considered *inert materials*, since the ratios specified in Decision 2003/33/EC (2:1 and 10:1) are lower than the tested liquid: solid ratio (20:1); therefore, the limit values would be greater for the ratio used. It must be also taken into consideration that the test methods have been different (Decision 2003/33/EC instead of UNE-EN-1744-3, 2003).

CONCLUSION

The availabilities of Cr, Ni, Cu, Zn and Cd have been relatively low in the washing aggregate sludge, sewage sludge and clay-rich sediment. When changes occur in the pH, the HM's that have shown the highest available concentrations have been Ni, in W and C, and Zn, in SS. Also when changes occur in the redox conditions, the HM's with the highest available concentrations have been Cr, in W, and Zn, in SS and C. In the studied gravel pit, the sediment ponds may be used for cultivation when they are silted up. On the contrary, agricultural application of SS is not possible. W, SS and C may be used for the manufacturing of LWAs, since neither of them have presented HM values above the specified limits for re-use in building materials. The availability of all the studied HM's has been reduced by the thermal treatment. In general, the studied LWAs may be used in lightweight concrete manufacturing from the standpoint of HM leaching.

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