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Evaluation of Coal Bottom Ash for clay brick manufacturing: a preliminary study

Evaluación de cenizas de fondo de carbón para la fabricación de ladrillos de arcilla: estudio preliminar

Janneth Torres-Agredo [1a], Luisa Fernanda Mosquera-Idrobo [1b], Patricia Paz-Villegas [2a], Miguel Fernando Díaz-Huertas [2b]

[1] Grupo de Investigación Materiales y Medio Ambiente (GIMMA), Facultad de Ingeniería y Administración,
Universidad Nacional de Colombia, Colombia. Emails: [a] jtorresa@unal.edu.co, [b] lufmosqueraid@unal.edu.co.
Orcid: [a] 0000-0002-4094-8387.
[2] C.I. Lago Verde S.A.S., Colombia. Emails: [a] gerenciadeoperaciones@cilagoverde.com.co,
[b] gestionambiental@cilagoverde.com.co.

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Abstract

Currently, industrial, economic, and social growth has produced large amounts of solid waste, which harms the environment and human health. Coal bottom ash (CBA) is a waste produced by burning coal. A preliminary study on CBA, to be used as raw material for the clay bricks manufacture, is presented. CBA was characterized through the Laser Granulometry, X-ray Fluorescence (XRF), X-ray diffraction (XRD) techniques; besides, the real and apparent density and the content of organic matter. Furthermore, the environmental tests Toxicity Characteristic Leaching Procedure (TCLP) and Daphnia Pulex acute toxicity test, were applied. It was found that the CBA is an amorphous material, and is composed of oxides of silica, iron, aluminum, and others, while the environmental tests satisfactorily met the applicable standards. According to the results, it is concluded that the CBA has a great potential to be used in the manufacture of bricks.

Keywords: bottom ash; clay bricks; coal; reuse; waste management; solid waste; Laser Granulometry, X-ray Fluorescence; X-ray diffraction.

Resumen

El crecimiento industrial, económico y social ha generado grandes cantidades de residuos sólidos que causan impactos negativos al medioambiente y a la salud humana. Se presenta un estudio preliminar de cenizas de fondo de carbón (CBA), residuo de la combustión del carbón, para ser usado como materia prima en la fabricación de ladrillos de arcilla. Se aplicaron técnicas de granulometría láser, fluorescencia de rayos X y difracción de rayos X; además, se determinó la densidad real y aparente y el contenido de materia orgánica. Se aplicaron técnicas ambientales a través del ensayo de TCLP (*toxicity characteristic leaching procedure*) y ecotoxicidad por *Daphnia pulex*. Se encontró que el residuo es un material amorfo, compuesto por óxidos de silicio, hierro, aluminio y otros; además, el residuo cumple con la normatividad medioambiental. De acuerdo con los resultados, se concluye que este residuo tiene un gran potencial para ser usado en la fabricación de ladrillos de arcilla.

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Palabras clave: aprovechamiento; carbón; cenizas de fondo; gestión de residuos; residuo sólido; ladrillos de arcilla; granulometría láser; fluorescencia de rayos X; difracción de rayos X.

1. Introduction

In recent decades, quick industrial, economic and social growth has caused large amounts of solid waste, producing negative impacts on the environment and human health [1]. Currently, waste disposal is one of the main environmental problems since they do not only pollute the environment but also generate a burden on the land [2]. Some of these solid wastes are mining waste, chemical process waste, foundry waste, and combustion waste [1].

The World Coal Association [3], reports that coal accounts for 30% of global primary energy consumption and more than 40% of electric power generation. It is estimated that coal will continue to be the second-largest energy source in the world until 2030, and the third from 2030 to 2040 behind liquid fuels and natural gas [4]. The use of coal as an energy source generates large amounts of waste, such as coal combustion products (CCPs); these include flying ash (CFA - coal fly ash), bottom ash (CBA - coal bottom ash), boiler slag, and flue gas [5].

It should be noted that coal has a significant amount of trace elements that after combustion are concentrated in CCPs. For example, coal ashes have some elements such as Arsenic, Cadmium, Lead, Mercury, and Selenium, which represent 1% of the total ashes [6]. Since these elements are potentially dangerous, the removal of coal ashes is of great environmental concern, due to the leaching of heavy metals to sources of surface and groundwater [7].

From the coal ashes, 10-20% corresponds to the bottom ashes [8]. CBAs are thick particles, too large to be transported in the flue gases; therefore they collide with the furnace walls and fall into the bottom [9]. It is estimated that approximately 8.5 million tons of CBA are produced annually worldwide [10], and only 5.28% of the ashes are reused in different processes [11].

Regarding the environmental impact of the bottom ashes, Singh et al. [12] point out that the methods of open disposal of CBAs in the various industrial sectors and thermal power plants cause environmental pollution and risks to human health. For example, in Malaysia, CBAs are considered hazardous waste [10].

On the contrary, in the United States, coal combustion products are classified as non-hazardous waste in subtitle D of the Resource Conservation and Recovery Act (RCRA); however, parameters are established to ensure that landfills are located, built, and closed properly, performing groundwater monitoring [13].

On the other hand, in recent years studies have been reported on the use of CBAs for brick making. Andreola et al. [14], conducted a study on the performance of clay bricks made with the addition of CBA between 2.5 and 20% concerning the amount of clay, with a cooking temperature of 1010 °C. In the bricks produced, the appearance of efflorescence was found due to the content of soluble salts. The authors concluded that the CBA is not the one indicated for these applications, since a greater amount of mixing water was required, therefore causing an increase in water absorption and porosity.

These results contradict those found by Da Fonseca et al. [15], who conducted a study on the possibility of using CBA to produce clay bricks at an industrial level, using proportions between 2.5% and 20% and cooking temperatures between 900 °C and 1100 °C. Before the preparation of the mixtures, a milling process was done to the CBA obtaining an average particle size of 138µm. These authors found a reduction in water absorption and open porosity, due to a fluxing action that the residue shows; in addition, the compressive strength improved with the increase in the cooking temperature. Finally, they conclude that these CBAs can be used in the ceramic brick industry.

Refractory bricks have also been made with the addition of this residue; this is how Braganca et al. [16], studied the possibility of adding CBA as a partial replacement of the chamotte (calcined and ground clay). In this case, they used two chamotte replacement ratios (5% and 10%) and a cooking temperature of 1350 °C. The thermal conductivity properties, compression strength, and density were evaluated. The authors found that for all properties evaluated, the performance of bricks added with CBA was comparable to commercial bricks. The only alteration that occurred was in the color of the product; however, the authors mention that these bricks could be marketed as a green product.

These findings indicate that coal-bottom ashes have a great potential to produce bricks. Therefore, the present study aims at evaluating the characteristics of the CBA, so that they can be used as secondary raw material in the manufacture of clay bricks. It will therefore be possible to give an added value to the waste, reducing the amount of volume of the waste for final disposal.

2. Materials and methods

The CBAs were obtained from a Colombian company whose economic activity consists of the manufacture of clay bricks; CBAs are produced during the oven cooking operation at a temperature of 850 °C. The ashes are collected at the bottom of the oven, as shown in Figure 1.

Three (3) samples were taken during the study, which were called CBA1, CBA2, and CBA3. Likewise, a sample of the mineral coal used in the company was taken, to know its chemical composition. The coal bottom ashes were analyzed through chemical and mineralogical composition, density, particle size, and organic matter content due to fire loss (Loss on ignition - LOI). In addition, an environmental characterization was carried out.

The chemical composition was carried out using the X-ray fluorescence technique (XRF). The samples were reduced in particle size with an agate ball mill and then passed through a 100 µm mesh sieve. Then, they were dried at 105 °C for 12 hours. Finally, semi-quantitative analysis was carried out with the SemiQ5 software, to detect all the elements present in the sample, excluding Hydrogen, Carbon, Lithium, Beryllium, Boron, Nitrogen, Oxygen, and the transuranic elements.

An X-ray fluorescence spectrometer was used, MagixPro PW-2440 Philips equipped with a Rhodium tube, with a maximum power of 4 KW, which has a sensitivity of 100 ppm in the detection of heavy metal elements.

The mineralogical composition was made from X-ray diffraction (XRD). The measurement was performed on a PANalytical X-ray diffractometer, EMPYREAN model. The sample was measured in a Bragg-Brentano optical configuration with a high-speed solid-state detector for data acquisition, called PIXCEL 3D 1x1. A quantitative analysis of the crystalline phases was carried out using the Rietveld method and the amorphous content based on the "Internal standard method". This test was performed for samples CBA2 and CBA3.

The particle size distribution was determined from the Laser Granulometry technique, between a range of 0.02 to 2000 µm, with the MasterSizer 2000 equipment. On the other hand, the real and apparent density was determined; as well as the organic matter content due to fire loss according to ASTM D 7348-13, at 950 °C.

For the environmental characterization, the leaching test was applied, Toxicity Characteristic Leaching Procedure (TCLP) according to EPA Method 1311 [17]. The leachable metals analyzed were Cr, Hg, Ba, As, Ag, Cd, Se, Pb.

Figure 1. Sampling site of CBA. Source: own elaboration.

Table 1. Chemical composition of mineral coal

Compositions	(wt%)	Compositions	(wt%)
SiO_2	16.28	Ba	0.07
SO_3	8.47	Sr	0.04
Al_2O_3	6.75	V	0.02
Fe_2O_3	1.99	Zr	0.02
K_2O	0.79	Cr	80 ppm
TiO_2	0.52	Rb	77 ppm
CaO	0.43	Zn	74 ppm
MgO	0.36	Pb	71 ppm
Na_2O	0.18	Y	40 ppm
P_2O_5	0.17	Nb	19 ppm

Source: own elaboration.

In addition, the ecotoxicity of the residue was determined by employing the acute toxicity test for Daphnia pulex following the protocol established in the EPA (2002 EPA 821-R-02-012); corrosivity was also determined according to the 9040C "pH Electrometric Measurement" method. This was done to use the waste with social and environmental responsibility.

3. Results and discussion

3.1. Chemical composition of mineral coal

Table 1 shows the chemical composition of mineral coal, which is mainly composed of SiO_2, SO_3, and Al_2O_3. Choi et al. [18], reported the chemical composition of anthracite carbons, where its main components were SiO_2 (15.2% - 21.6%), Al_2O_3 (10% - 13.1%), and Fe_2O_3 (1.4% - 1.8%) In that sense, the coal studied shows an Al_2O_3 content below that reported, however, it does not move far from the range; therefore, it can be said that the composition of the mineral coal is similar to that of anthracite coal. On the other hand, the mineral coal studied has relatively low concentrations of toxic metals such as V, Cr, and Pb.

3.2. Chemical composition and physical properties of CBA

The chemical composition of coal-bottom ashes is presented in Table 2. It is observed that these ashes are mainly composed of SiO_2 and Al_2O_3, with small amounts of Fe_2O_3, K_2O, SO_3, CaO. This composition is consistent with the mineral coal presented in Table 1. The evaluated CBAs have a chemical composition similar to that reported by other authors [8], [11], [19], [20], [21], [22], [23].

It should be noted that the CBA from anthracite and bituminous coals are characterized by low amounts of calcium, and the sum of the compounds of SiO_2, Al_2O_3, and Fe_2O_3 is close to 90% [23]. The above coincides with the chemical composition obtained for CBA1, CBA2, and CBA3. In addition, the evaluated CBAs can be classified as Class F ashes (pozzolanic compounds; $SiO_2+Al_2O_3+Fe_2O_3>70\%$) [8], [12], [15].

It is worth mentioning that the background ashes studied contain toxic metals such as V, Cr, and Pb. The concentrations of these metals in the ashes do not show a significant variation with mineral coal. It should be noted that metals are present in ashes in a relatively small fraction, however, their possibility of leaching to the environment can affect their potential use.

On the other hand, a lump of clay suitable for brick making must have a SiO_2 content that varies between 50 and 60%; as well as between 10 and 30% of Al_2O_3 [23], [24], [25]. As for the Fe_2O_3 content, clays with iron contents of less than 10% are used, since the presence of this compound can cause efflorescence problems in ceramic products [23].

However, the iron present in the ashes can play an important role in the color of ceramics, since raw materials with iron percentages between 5 to 7 produce red ceramics [15]. Finally, the CaO content varies up to 10% [23]; it is important to highlight that raw materials with low CaO contents will have less tendency to efflorescence problems in ceramic products [25].

In this case, the evaluated CBAs have the amount of SiO_2, Al_2O_3, Fe_2O_3, and CaO required for the production of ceramics. However, the presence of sulfur in CBAs can influence sulfate efflorescence formation [28].

According to the aforementioned, the background ashes under study have a chemical composition similar to the clays used for the manufacture of bricks, for this reason, it can be said that the ashes have great potential to be used as a clay substitute.

On the other hand, loss on ignition (LOI) of CB1, CB2 and CB3 was 0.4, 0.3 and 0.3, respectively. In the literature, LOI values of 0.02 to 8% have been reported for CBA; which coincides with the present study [8], [10], [16], [23], [28], [29], [30].

According to the average particle size, it is observed that CBA1 has finer particles compared to CBA2 and CBA3; this difference may be due to the efficiency of the combustion process. Also, the physical properties of coal bottom ashes are influenced by the type and degree of pulverization of coal and cooking temperature [23]. Likewise, Hashemi et al. [10], Singh [23], and Sutcu et al. [30] reported a particle size for CBA between 63 microns to 10mm.

Finally, Table 2 shows the real and apparent density of coal bottom ashes. It is observed that CBA2, presented a lower real density compared to CBA1 and CBA3.

Argiz et al. [8], Aydin [22], Singh [23] and Rafieizonooz et al. [27], reported densities ranging from 1.2 to 2.65 g/cm^3 for CBA. On the other hand, Yao et al. [31], found that coal ashes have apparent densities ranging from 0.54 to 0.86 g/cm^3; densities similar to those obtained in this work.

3.3. Mineralogical composition of CBA

In Figures 2 and 3, the X-ray diffractograms for the CBA2 and CBA3 ashes are shown. It is observed that the main crystalline phases of the ashes are Quartz and Hematite, minerals that contain the elements such as Si and Fe.

In Figure 2, it is observed that CBA2 presents Mullita, responsible for the high content of Aluminum [15].

In addition, the ashes have traces of other minerals such as Muscovite, Andradite, Ilmenite, and Anatase. X-ray diffractograms show that the ashes evaluated have an amorphous structure. X-ray diffractograms show that the ashes evaluated have an amorphous structure.

Table 3 shows the quantification of the mineralogical phases, where the amorphous phase is the main constituent of the ashes, with amounts of 56.6% and 62.5% for CBA2 and CBA3, respectively; followed by quartz and hematite. The mineralogical composition of the ashes is similar to that reported by other authors [10], [15], [29], [32].

On the other hand, for the manufacture of ceramic materials, the crystalline, and amorphous phases play an important role during sintering, since the addition of non-crystalline materials can be considered as flow agents that induce vitrification in a clay matrix, favoring the formation of ceramics with higher density, less water absorption and greater mechanical resistance [15].

Table 2. Chemical composition and physical properties of CBA

Characteristics	CBA1	CBA2	CBA3
SiO_2	59.77	60.52	59.04
Al_2O_3	27.89	27.24	24.41
Fe_2O_3	4.97	4.09	5.7
K_2O	1.61	1.44	1.67
SO_3	1.57	1.07	4.41
CaO	1.23	1.17	0.84
TiO_2	1.13	1.23	1.21
MgO	0.63	0.54	0.66
Ba	0.11	0.14	0.13
V	0.02	0.04	0.03
Cr	0.02	0.02	0.03
Pb	77 ppm	97 ppm	84.3 ppm
LOI	0.4	0.3	0.3
Particle size (µm)	127	207	236
Density (g/cm^3)	2	1.23	2.1
Apparent density (g/cm^3)	0.5	0.61	0.61

Source: own elaboration.

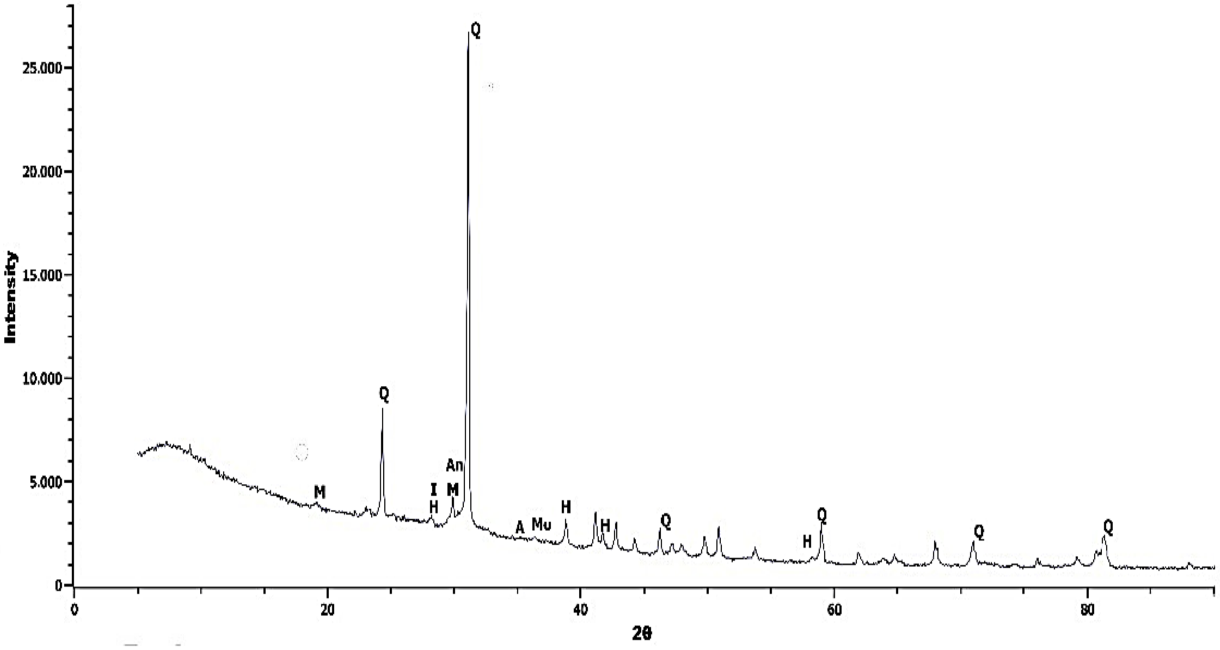

Figure 2. Diffractogram of CBA2. Q-Quartz; Mu-Muscovite; H-Hematite; An-Anatase; A-Andradite; I-Ilmenite; M-Mullite. Source: own elaboration.

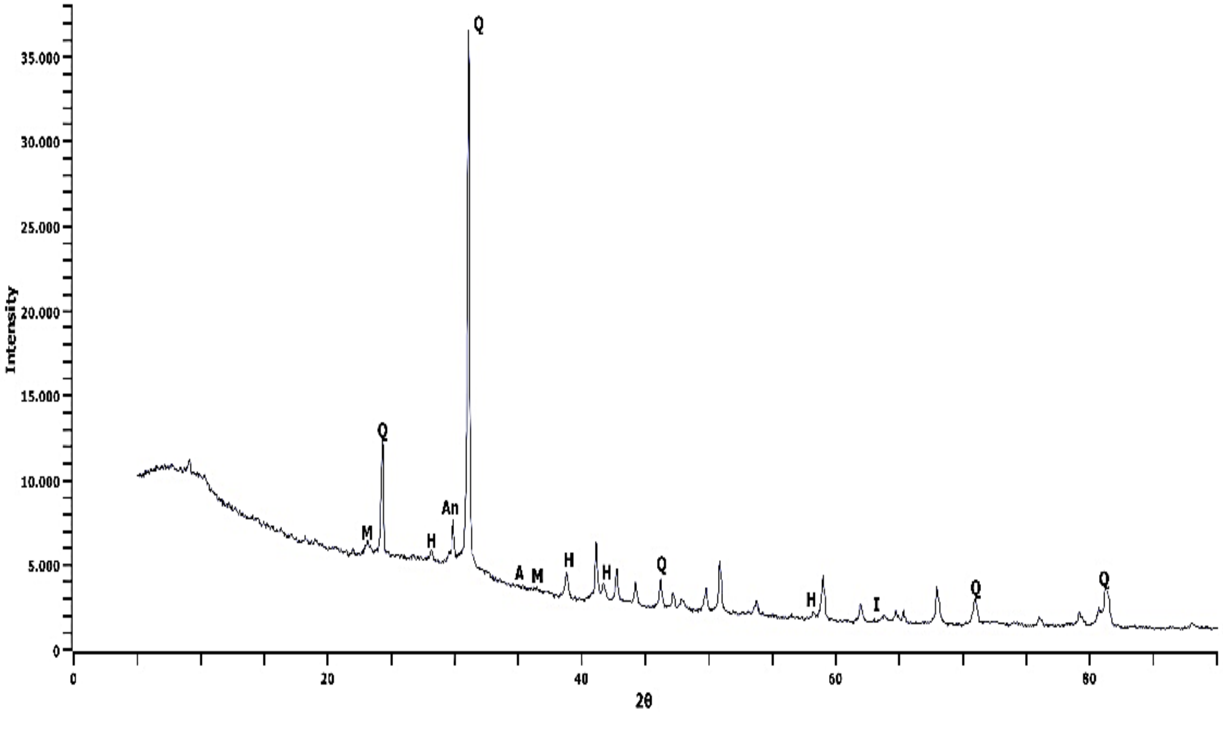

Figure 3. Diffractogram of CBA3. Q-Quartz; M-Muscovite; H-Hematite; An-Anatase; A-Andradite; I-Ilmenite. Source: own elaboration.

Table 3. Quantitative analysis of the crystalline and amorphous phases of CBA

Phase		Chemical formula	CBA2 (%)	CBA3 (%)
Crystalline	Quartz	SiO_2	35.4	29.7
	Hematite	Fe_2O_3	3.7	5.5
	Mullite	$Al_{5.65}O_{9.175}Si_{0.35}$	2.1	----
	Muscovite	$H_2Al_3KO_{12}Si_3$	1.1	1.6
	Andradite	$Ca_3Fe_2O_{12}Si_3$	0.5	0.3
	Ilmenite	$FeTiO_3$	0.5	0.1
	Anatase	TiO_2	0.2	0.3
Amorphous fraction			56.5	62.5

Source: own elaboration.

Likewise, the presence of minerals such as hematite and anatase can confer the red color to ceramic pastes [25].

Additionally, the studied CBAs have high quantities of quartz and amorphous material; therefore they could also be used as a degreasing material for the manufacture of construction ceramics [25], [26], [27]. According to their mineralogical composition, the ashes studied have great potential to be used for the manufacture of ceramic materials.

3.4. Environmental characterization of CBA

Heavy metal leaching of any material, when used in civil engineering applications, is an environmental concern [23]. Coal ashes have been considered hazardous waste, due to the presence of heavy metals that can leach and contaminate soils and water [10]. For this reason and, considering that the ashes evaluated have heavy metals, it is important to carry out an environmental analysis to assess their environmental impact before they are used as secondary raw material, and thus use the waste with social and environmental responsibility.

Table 4 shows the results obtained from the corrosivity tests, Daphnia Pulex ecotoxicity, and the TCLP leaching test, for CBA1 and CBA2. According to the pH presented by CBA1 and CBA2 (6 and 5.89 respectively), ashes are considered as a non-corrosive residue.

Regarding the ecotoxicity test in Daphnia Pulex, the ashes presented a percentage of immobilization of Daphnia lower than 10, which is below the maximum permissible established in EPA 821-R-02-012, which indicates a low toxic effect in test organisms. Therefore, ashes are considered a non-ecotoxic waste.

The leachable metals evaluated are below the maximum permissible levels established by the EPA, therefore, the ashes studied can be considered as non-hazardous waste. On the other hand, the concentrations of the leachable metals of CBA1 and CBA2 comply with the limits established by Malaysian environmental regulations for metals such as Cr (0.2 m/L), Cd (0.01 mg/L), and Pb (0.1 mg/L) [10].

Likewise, the concentrations of leachable metals from the evaluated ashes are below those reported in the literature for flying ashes [33].

It is worth noticing that low concentrations of leached metals from bottom ash could be due to the encapsulation of most of the hazardous elements within the amorphous material [34]. Also, it can be mentioned in general, that the bottom ashes usually have larger particles compared to the fly ash; therefore, there is a lower probability that heavy metal leaching will exceed the permissible limits [23], [35].

On the other hand, these results are according to those reported by Kierczak and Chudy [21], where they concluded that CBAs are an inert material, because the ashes do not present significant concentrations of inorganic pollutants and, the potential mobility of trace elements is relatively low. Jones et al. [9], evaluated the leaching capacity of coal bottom ash using the TCLP leaching test, finding that leachable metals such as Cd, Pb, As, Cr, Se, did not exceed the permissible limits established by the EPA.

According to the results obtained in the TCLP leaching, corrosivity, flammability, and ecotoxicity test, the carbon bottom ashes studied are a non-hazardous and non-ecotoxic waste, which makes it a residue with great potential to be used as secondary raw material.

Table 4. Environmental analysis of CBA

Test	CBA1	CBA2	Standard*
TCLP Cr (mg/L)	<0.1	<0.01	5
TCLP Hg (mg/L)	<0.001	<0.001	0.02
TCLP Ba (mg/L)	1.6	0.39	100
TCLP As (mg/L)	<0.022	<0.005	5
TCLP Ag (mg/L)	<0.01	<0.01	5
TCLP Cd (mg/L)	<0.006	<0.006	1
TCLP Se (mg/L)	0.001	0.05	1
TCLP Pb (mg/L)	0.06	0.06	5
Corrosiveness (pH)	6	5.89	2 - 12.5
Total Inmmobilization Daphnia pulex (%)	≤ 10.0	10	≤ 50 %

*According to EPA 1311 for TCLP, EPA 9040C for Corrosiveness and EPA 821-R-02-012 for Daphnia Pulex.

Source: own elaboration.

4. Conclusion

The chemical and mineralogical composition reported in the coal bottom ashes of the study shows that this residue has great potential to be used, as secondary raw material in the manufacture of clay products. Mainly because the mineral phases found in the ash correspond to the main components for the manufacture of clay products. Likewise, the DRX analysis showed a high content of amorphous material in the ashes, this characteristic can enhance the ceramic products with higher density, less water absorption, and greater mechanical resistance.

The coal bottom ashes studied could also be used as a degreasing material in ceramic bricks, due to its high content of quartz and amorphous material.

Regarding their environmental characteristics, the CBA studied can be considered as a non-hazardous and non-toxic waste, since they meet the maximum permissible levels established by the EPA.

The great potential of coal bottom ashes, to be exploited in the manufacture of ceramic products, will reduce the consumption of raw materials for the manufacture of bricks. In addition, it will allow producing an improved and ecological building material, bringing therefore, economic, and environmental benefits.

Considering that the present study consisted of an initial characterization of the ashes, it will be important to define in future investigations the optimal percentages of ash addition for the manufacture of clay bricks. In this case, the authors going to prepare since laboratory analysis until industrial scale, to use this waste in the future.

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