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To cite this article: J Bautista-Ruiz et al 2020 J. Phys.: Conf. Ser. 1708 012014

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1708 (2020) 012014 doi:10.1088/1742-6596/1708/1/012014

Fly ash as to solidify liquid radioactive waste

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Abstract. The "Termopaipa" power plant located in Boyacá, Colombia, generates electricity based on coal and whose residue is fly ash. In this study we consider it important to understand its physical, chemical and morphological characteristics to be used for purposes beneficial to the environment and to use them as a solidifier of liquid radioactive waste. The treatment of radioactive waste involves the set of physical and chemical processes that entail the change of some characteristic of the initially generated waste in order to optimize the safety and / or economy of its management. This treatment requires the transformation of the radioactive waste into an acceptable final product for its transport and / or temporary storage and / or definitive storage. This transformation can be carried out through the incorporation of residues in phases such as Mullite and quartz identified by X-ray diffraction, or encapsulation within porous structures present in the ashes observed by scanning electron microscopy and fixation based on an electrostatic interaction mechanism. originated on its surface due to the presence of aluminate and silicate groups identified by X-ray photoelectron spectroscopy.

1. Introduction

Fly ash (FA) is a material resulting from the combustion of pulverized coal in thermoelectric plants. Its composition and both physical and chemical properties depend on the type of coal, its origin, as well as the technological processes used in these plants [1-3].

Currently, applications for these carbon residues are being investigated due to their physical and chemical characteristics [4-6]. Several applications have been developed for this type of waste, such as: cement manufacturing, metal alloy manufacturing, zeolite production and environmental remediation [7-9]. Within the latter, the applications offered by fly ash are varied, highlighting the fixation, adsorption and absorption of industrial waste such as mercury, phosphates, organic components of water, motor oils [9-11]. At present, materials are being sought that allow solidifying liquid radioactive waste with other advantages compared to the materials that are currently used for this purpose, some of the advantages that are sought are: chemical and physical stability, high resistance to acids, elimination of moisture to low temperatures, high porosity and that it is a very economical material among others [12,13].

In the development of this work it was proposed to find the physical and / or chemical characteristics of the fly ash produced in a thermoelectric station in Boyacá Colombia so that it can be used as a solidifier for liquid radioactive waste; For this purpose, they were analyzed at the volume and surface

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level using various characterization techniques in order to know the associated physical and chemical phenomena that allow the use of ashes as a reagent for the solidification of liquid radioactive waste. The results allow to conclude that some properties present in these ashes would allow its use as a solidifier for radioactive liquid waste.

2. Materials and methods

The FA used in the current study is the combustion waste product from Colombian coal used in "Compañía Eléctrica de Sochagota" SA, "Termopaipa" IV located at km 5 highway Paipa to Tunja, in Boyacá, Colombia. The characterization techniques used in the work are fundamental to establish physical and chemical properties of the material under study both in volume: X-ray fluorescence (XRF) and X-ray diffraction (XRD) and in surface: scanning electron microscopy (SEM) and, X-ray photoelectric spectroscopy (XPS).

2.1. Compositional and mineral characterization

For the determination of the composition and concentration of the oxides present in the FA samples, a PANalytical energy dispersed X-ray fluorescence spectrometer, Axios Petro model of 4 kV was used with a xenon (Xe) detector of flow scintillation and rhodium (Rh) anode. Additionally, and parallel to the XRF analysis, a loss on ignition (LOI) analysis was performed at 105 °C.

The mineralogical analysis of the FAs was developed using a PANalytical diffractometer, Empyream model in θ -2 θ mode. The measurement was taken with K α 1 radiation (λ =1.7890 Å) using the Bragg-Brentano geometric configuration, with a scanning speed of $0.013^{\circ}/160$ s and tube voltage of 45 kV, in a range between $2\theta = 10^{\circ}$ and 90° .

2.2. Surface characterization of fly ash

The morphological characterization and chemical composition were developed in a field emission scanning electron microscopy (FE-SEM) model Nova Nano SEM 230. The voltage applied in the range 50 V to 30 kV.

The semi-quantitative determination of the composition and electronic structure of the sample was performed using X-ray photoelectron spectroscopy in an ultra-high vacuum (UHV) chamber with a base pressure of 10^{-10} mbar. The angle between the hemispherical analyzer (SPECS-PHOIBOS100) and the plane of the surface was maintained at 60° and the X-ray radiation was the K α line of magnesium (Mg) (1253.6 eV). The bond energy scale (BE) was calibrated for carbon (C) 1s for 285 eV. The reference spectrum was recorded with an energy step size of 40 eV; and an energy step size of 0.25 eV for high-resolution spectra. The binding energy scale (BE) was calibrated for C 1s for 285 eV. The analysis of the XPS results was carried out with the Casa XPS software. XPS data were performed with magnesium anode (Mg) K α ; the background signal from the spectrum was subtracted using a Shirley routine.

3. Results and discussion

The results obtained through the characterization techniques are presented and it is discussed which characteristics serve of this type of material to be a candidate as a solidifier for liquid radioactive waste.

3.1. Chemical and mineralogical composition

Table 1 shows the main compounds (expressed as oxides) and the composition expressed as a percentage by weight (% Wt). As shown in Table 1, the major components of FA are oxides of silicon, aluminum, and iron. According to the X-Ray diffraction (Figure 1), fly ash consists of Mullite ($Al_{4.80}O_{9.60}Si_{1.20}$), Quartz (SiO_2), Hematite (Fe_2O_3), and magnetite (Fe_3O_4), It is possible to classify these ashes as type F by the sum of $SiO_2 + Al_2O_3 + Fe_2O_3$ oxides > 70%, the classification of type F ash was confirmed by XRF. When comparing the percentages by weight obtained in this investigation with those reported by [9,10], which are works in applications for the immobilization of radioactive waste.

The presence of mineral phases of Mullite and Quartz, which are mainly composed of aluminum and silicon, are of special interest because they attribute fixing properties to it as mentioned Vasilieva *et al.*

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2005 [12], because these crystalline phases interact chemically so that the components of radioactive waste remain fixed, and by the action of an external heat source they are incorporated into the crystalline lattice allowing the stability of radioactive waste

Table 1. Compositional analysis of fly ash

in decreasing order of concentration.	
Compound chemical	(% Wt)
SiO	64.87

Compound chemical	(% Wt)
SiO_2	64.87
Al_2O_3	24.60
Fe_2O_3	4.50
K_2O	1.37
CaO	1.32
TiO_2	1.20
P_2O_5	0.65
Na ₂ O	0.51
MgO	0.41
SO_3	0.12
SrO	0.11
BaO	0.10

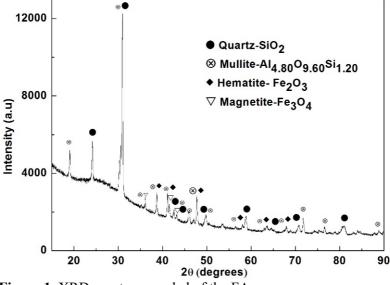


Figure 1. XRD spectra recorded of the FA.

3.2. Morphology and composition of surface

The morphology of the FAs was characterized by SEM technique was used to evaluate the samples in detail Figure 2 shows particles with high spherical symmetry. The morphological analysis showed a particle with spherical symmetry with an average diameter of 53 µm, with smaller spheres inside this type of particle is known as the plerosphere, whit spherical particles with a wide distribution of particle size with diameters of 13 µm, 14 µm, 16 µm, 17 µm, which can often be hollow or solid and are known as cenospheres, Santaella 2001 [3]. The two types of hollow particles are of special interest Vasilieva et al. 2005 [12], he has suggested using these to encapsulate and solidify radioactive waste and then are attached to them by the action of an external heat source.

The analysis showed that the main elements of the region near the surface are oxygen, carbon, silicon, aluminum and sulfur, the presence of aluminum and silicon play an important role Van der Merwe et al. 2014 and Chujiang et al. 2003 [14,15], suggested that the high concentration of these elements allows the formation of aluminate and silicate groups responsible for causing a negative electrical charge on the surface; this would allow the surface to electrically fix radioactive waste having a positive electrical

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nature. The presence of aluminum and silicon gives rise to crystalline phases such as mullite and quartz and is responsible for the negative electrical charge on the surface.

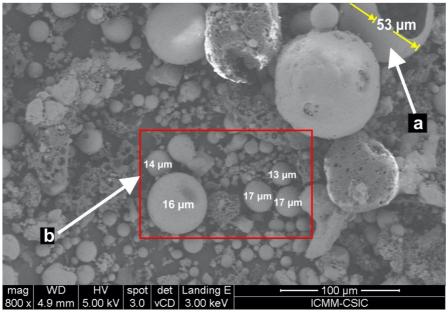


Figure 2. General morphology of a sample and details of the surface of FA.

4. Conclusions

Particles with hollow spherical symmetry on the surface of fly ash can be used as encapsulates of liquid radioactive waste. The negative electrical charge on the surface is useful in generating an electrical mechanism to attract liquid radioactive waste.

Acknowledgement

The authors thank "Dirección de Investigaciones" (DIN) of the "Universidad Pedagógica y Tecnológica de Colombia", to "Compañía Eléctrica Sochagota S.A.E.P." – "Termopaipa IV", Colombia, especially to Ing. William Castellanos for supplying the material for its characterization, and the Low Dimensional Advanced Materials Group of the "Instituto de Ciencia de Materiales de Madrid" (ICMM), "Consejo Superior de Investiaciones Científicas" (CSIC), Spain.

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