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Thermal activation of coal tailings

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Abstract

This paper reports on a study of the thermal activation of coal tailings in order to increase the intrinsic reactivity of this material.

Thermal treatment was carried out in a furnace with an inert atmosphere in the temperature range from 100 to 1000°C. The changes in the material following the treatment were determined by X-ray diffraction, thermogravimetric analysis and differential scanning calorimetry.

In order to verify the true reactivity of the activated material, leachings with standard solutions of HCl were made and the yield of the aluminium extraction process was determined.

INTRODUCTION

Coal tailings are complex clay materials, mainly composed of silicates with a high aluminium content and a considerable coal fraction [1].

The recovery of these waste materials which derive from the exploitation of coal employing aluminium-containing compound ores, can be achieved by acid leaching and subsequent separation of the other metallic cations present [2–4]. The yield of the acid-leaching process depends on the thermal treatment of the initial material.

This paper reports a study of the thermal activation of the coal tailings which was undertaken in order to determine the calcination temperature required to obtain the best material for the acid-leaching process.

Thermal energy can totally or partially break down the crystalline arrangement of a mineral. Nitrates and carbonates are decomposed during thermal activation, producing gasification of the constitution water of the clay components of the tailings, and giving rise to mineral phases which are much more active in the leaching process.

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This study has determined the best activation temperature and has followed the changes of each mineral phase during the thermal activation.

EXPERIMENTAL

Chemical analysis

To verify the chemical determinations of the tailings, the material was first heated at 950°C to destroy the organic matter present, and then an alkaline fusion was performed using lithium metaborate, the resulting solutions being analyzed (Al^{3+}, Fe^{2+}, K^+, Ca^{2+}) using a Varian atomic absorption spectrometer model 300 (Table 1).

Mineralogical analysis

A semi-quantitative mineralogical analysis was carried out using a Philips X-ray diffractometer model PV 1050/23 (Cu Kα radiation), following the Shultz method. Amounts of quartz and clay minerals were determined quantitatively by a study of the relative intensities of the characteristic reflections of the mineral species which make up the tailings. The intensity factors determined by Schultz for this kind of material were used for this purpose (Table 2).

Calcination of tailings

Activation was carried out by calcining the tailings in a furnace in an inert atmosphere; N_2 atmosphere was used in order to avoid autocombustion of the material so that the furnace temperature could be exactly controlled.

Coal tailings were calcined for 1 h at temperatures from 100 to 1000°C. Table 3 shows the mass losses at temperature intervals of 100°C.

Thermal analysis

The thermal behaviour of the tailings was followed by thermogravimetric analysis (TGA), and differential scanning calorimetry (DSC) in a Mettler TA 4000 (TG 50, DSC 30, rate of heating 10°C min^{-1}).

TABLE 1

Chemical analysis (%)

C loss	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	TiO_2
20	47	22	3.8	1.3	1.1	0.65	3.2	0.5

TABLE 2

Mineralogical analysis (%)

Quartz	Kaolinite	Illite	Montmorillonite	Other minerals
20	30	20	10	5

Leaching of the calcined tailings

The leaching conditions of the tailings calcinated at different temperatures were selected from the results of a study reported in a previous paper [5]: 3 h of lixiviation at reflux in 9.5 M HCl; 10 ml : 1 g ratio solution : solid. After leaching, the solid waste was separated by filtration and washed several times with small amounts of boiling water in order to dissolve the extracted cations. Aluminium and iron were determined by atomic absorption spectrometry.

RESULTS AND DISCUSSION

The results of the chemical and mineralogical analyses of the initial material are compiled in Tables 1 and 2. It should be noted that the high siliciate content found in the mineralogical analysis is in agreement with the chemical analysis. Chemical analysis also reveals significant amounts of Fe and C (organic and inorganic) in addition to the main component (Si).

Figure 1 shows the X-ray patterns of the solid waste obtained from calcination of the tailings at different temperatures, where the evolution of the different mineral phases with temperature can be observed.

The patterns for the tailings treated at 100, 200 and 300°C are similar. Up to 300°C, the material loses only moisture, its crystalline structure remaining unaltered. However, from 400°C, significant changes are observed.

Thus, the intensity of the diffraction peaks corresponding to the kaolinite decreases as the treatment temperature increases; the kaolinite peaks disappear at 800°C [6]. Something similar happens with the illite, although in this case the evolution is less clear because some of the peaks overlap with those of other components (mainly silica). Both kaolinite and illite are clay minerals, i.e. aluminium silicates of lamellar structure. Nevertheless,

TABLE 3

Loss in mass (%) on calcination of the material and different temperatures

100	200	300	400	500	600	700	800	900	1000
2.0	2.5	2.7	9.0	15.2	17.5	18.3	19.4	20.5	20.50

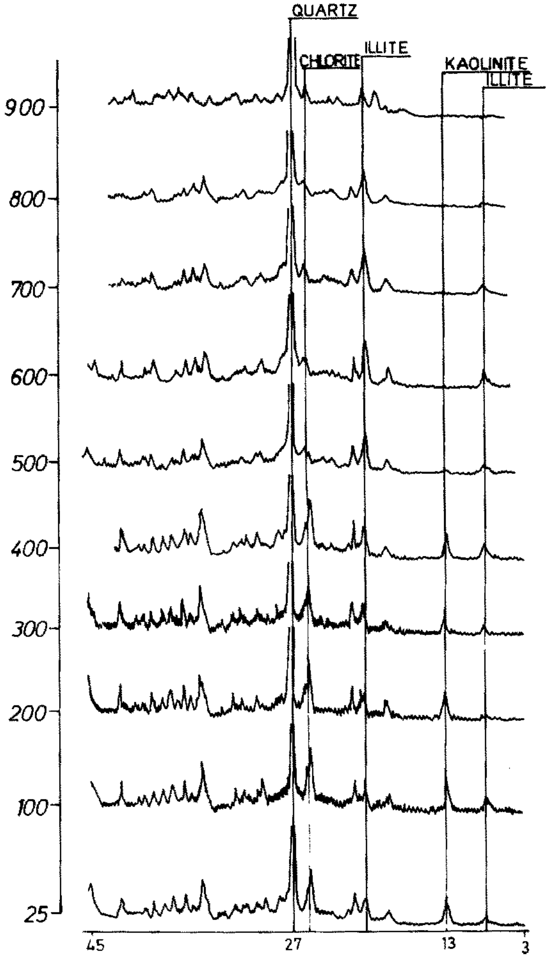

Fig. 1. X-ray patterns of the solid waste obtained from calcination of the tailings at different temperatures.

they are noticeably different because the kaolinite layers are bonded only by Van der Waals forces, while in the illite, strong electrostatic interactions resulting from the existence of interlayer cations stabilize the layered structure. The temperature needed for illite activation is about 150°C higher than that required to activate the kaolinite. In both cases, once the process is achieved, solid phases that are more easily lixiviated are obtained: metakaolinite, illitemonohydrate, etc. Higher treatment temperatures lead to the breakdown of the material and formation of amorphous silicate structures.

Siderite is another important component of the material which, in an oxidizing atmosphere, transforms into iron(III) oxide which may then crystallize as haematites. Siderite remains unaltered in the nitrogen atmo-

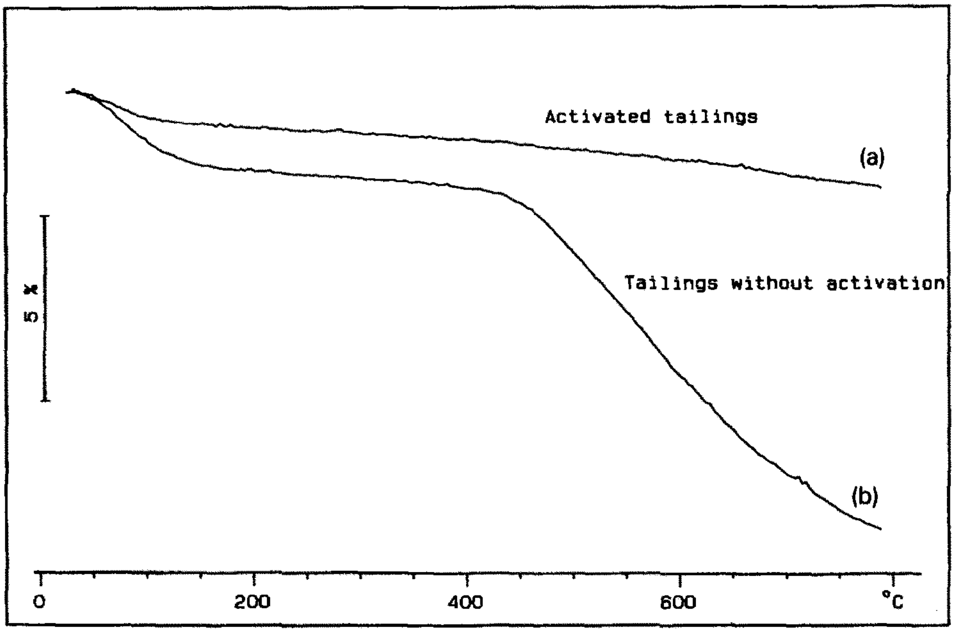

Fig. 2. Thermal analysis (TGA) of the tailings: curve a, non-activated tailings; curve b, tailings activated at 800°C.

sphere, helping the subsequent iron extraction, because iron oxides less readily lixiviated than siderite. This effect is reduced if the thermal treatment is carried out in air.

Figures 2 and 3 show the thermal analysis (DSC and TGA) of the activated and non-activated tailings at 800°C. The DSC curve of tailings that have not been activated shows three endotheric processes: the first, at 90°C, corresponds to the loss of moisture from the material; the second one, at 120°C, results from the gasification of the hydration water of the clays; the third peak is sharper and wider, and it corresponds to the loss of the constitution water of the material; it appears at 450°C, in agreement with literature data. The material calcined at 800°C shows only a small band between 50 and 100°C due to the moisture taken up by the material during its cooling (the samples were cooled to 200°C in the inert atmosphere and subsequently in air).

Leaching of the tailings treated at different temperatures proceeded according to the description above.

Figure 4 shows the aluminium recuperation plotted against the activation temperature of the tailings. At ≤ 400°C, the yield was about 10–15% of the aluminium extracted. From this temperature, yield increases noticeably, reaching a maximum value in the tailings calcined at 800°C.

From the results obtained, the following conclusions can be made: the thermal activation of the material is effective from 450°C when the kaolin-

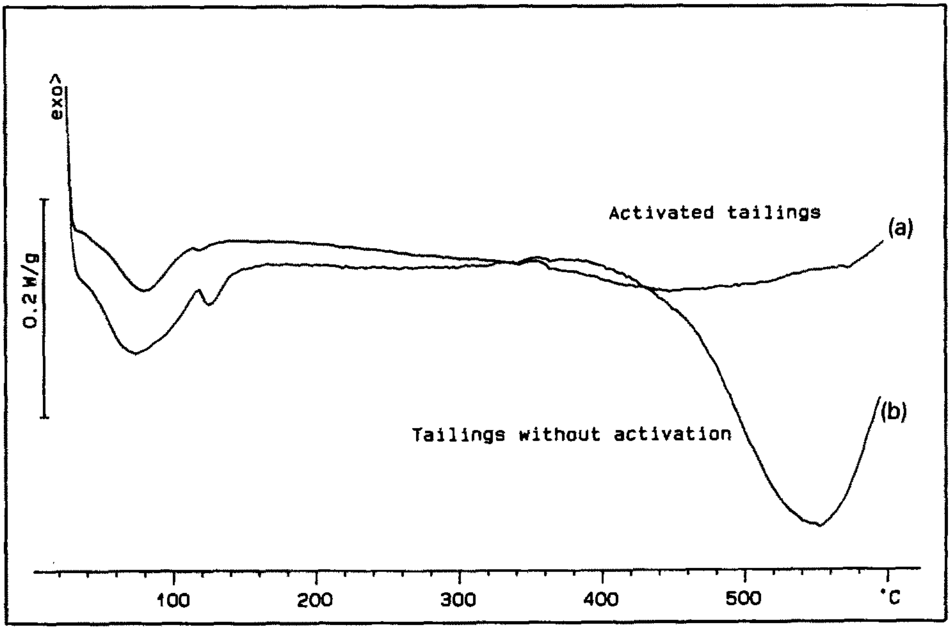

Fig. 3. Thermal analysis (DSC) of the tailings: curve a, non-activated tailings; curve b, tailings activated at 800°C.

ite structure begins to break down; illite activation is completed at 800°C, and therefore, this is the best activation temperature; above 800°C, the material progresses to the formation of amorphous silicate structures which are less readily lixiviated, so that the yield of the aluminium extraction decreases; activation is favoured by the kaolinite in the material owing to its lower thermal resistance.

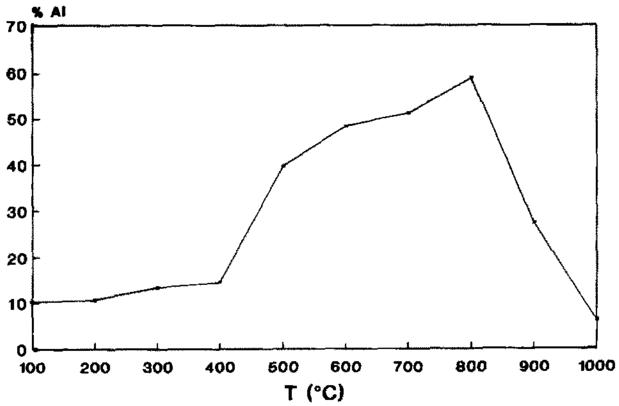

Fig. 4. Aluminium recuperation plotted against the activation temperature of the tailings.

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