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Characterization of Moroccan Coal Waste (Jerada Mine): Impact on Physical Properties of Mortars Made of Coal Waste

RajaeAddou[1], Kinda Hannawi[2], William Prince Agbodjan[2], Mohamed Zenasni[1]

[1]*Laboratory of Mechanics and Digital modeling 'LM2N, National School of Applied Sciences, Oujda, Morocco.*
[2]*Laboratory of civil engineering and mechanical engineering, 'LGCGM', INSA of Rennes, Rennes, France.*

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- ✓ ultrasonic wave propagation

R. ADDOU
rajaeaddou@gmail.com
+212672886496

Abstract

The exploitation of the mine of Jerada in Morocco generates significant quantities of waste rock stored in heaps. This study aims to exploit these mining wastes, as constituents in building materials. A first characterization work has been performed on coal waste, using mineralogical, thermal and physical tests on samples in powder form. The experimental results lead us to the possible recovery of such waste as partial replacement of cement or aggregates in mortars and concretes. An initial exploratory work was the use of coal waste as partial substitution in cementitious mortars, but the low index of activity showed that the waste does not present pozzolanic reactivity. Thus, the following study will be made on mortars with the addition of this waste as a volume replacement of sand with different percentages. In the second part, physical tests were conducted to characterize the mortars before and after heat treatment at 400°C and 600°C, by measuring the apparent density, porosity, gas permeability and the ultrasonic wave propagation. We have observed that porosity increases with the addition of waste and even with the heat treatment, resulting in a remarkably lightweight mortar. On the other hand, the measurement of the apparent gas permeability shows a decrease in permeability with the incorporation of waste. Furthermore, there is an increase of the gas permeability in mortars after thermal treatment, which is probably due to pore volume liberated by the combustion of coal. Measuring ultrasonic wave propagation shows that the sound velocity decreases with the addition of waste and with the heat treatment. This is attributed to the increase in porosity that will limit the waves' velocity. Thus, the thermal processing of these composites is interesting to obtain lightweight and insulating materials.

1. Introduction

The Jerada coal mine had being operated from 1927 to 2001, which generated significant amounts of rock waste estimated between15 to 20 million tons, which remain stored in the region as heaps. The coal from this region is of high quality as it contains primarily the anthracite with 2-5% pyrite. These dumps create an environmental impact manifest both in terms of landscape and contamination of aquifers (i.e. endangerment of living species and destabilization of soils).On the other hand, the construction sector requires ever increasing withdrawals of natural resources and faces in some cases, shortages of materials. Moreover, the rise in energy bills encourages builders and stakeholders to find efficient solutions for alternative materials, which are in line with sustainable development concerns.

Several studies [1 - 2] have already been conducted on the harmful environmental impact of the waste piles to evaluate the impact on air, soil and groundwater in the region. An acid mine drainage was proven in the study of Bendra and al. [1] with high rates of sulfates, chlorides and nitrates, more than the average specified by the World Organization of Health. Another study by M. Battioui and al. [2] on the groundwater quality in the province of Jerada shows strong chemical contamination and a high degree of pollution.

In addition and in the context of sustainable development, many researchers are interested in the recovery of such wastes and their exploitation in civil engineering operations. In Spain and in other countries such as Germany, the UK or France, they are used as armed land or as plugging material in road embankments, expressways, highways and railways in addition to the sea dikes. It was the aim of a study conducted by Gonzalez Cañibano and Madera Fernandez [3]. This waste can be recycled in the cement industry by saving energy due to exothermic phenomenon linked to the burning of coal, based on the thesis of D.Belkheiri and al. [4].

Finally, Darmane et al.[5] studied another way of valorisation of these wastes. Indeed, the abandoned slagheaps of the coal mine of Jerada have greatly evolved, particularly the contained pyrite has been oxidized. In some parts, the only remains are iron oxides, which are concentrated within the fine particles of the heap. The following ore processing: sifting, gravimetry and flotation have resulted in a marketable product that provides 35% iron oxide (Fe2O3) for use in painting, enamelling and bio-construction.

Our present project aims to valorize mine tailings from the Jerada region, reusing them as additional ingredients in building materials. This project will provide an effective solution to the triple challenges of performance and durability of materials, energy efficiency and reduction of environmental impact.

2. Materials and Methods:

2.1. *Materials:*

- *Coal Waste of Jerada:*

The test samples were collected from the large conical heap located just outside the city of Jerada towards Hassi Blal (Northeastern Morocco), 70 m of height and measuring 450x412 m at the base (see ***Figure 1***). First, coal tailings were finely milled because the fineness has an impact on their reactivity [6].

Figure 1: Grand waste heap of Jerada

- *Cement:*

The cement used is an artificial Portland cement type CEM I 52.5, specific gravity $\rho_c = 3.15$.
View size distribution curve of the cement used in Figure 1, made by laser granulometry, by a liquid (ethanol).

- *Sand:*

The sand used is a normal silica sand, specific gravity$\rho_s = 2.65$.

2.2 *Methods:*

2.2.1. Tests on mortars with mass replacement of cement: Measurement of the activity index

In this part of the work, the tests were conducted on prismatic specimens (40x40x160 mm^3) of mortars: standardized according to EN 196-1, with finely mashed coal waste in mass replacement of cement (0% to 25%). The aim was to study its reactivity in the mixture, and to decide its use in concrete.

These samples were first crushed at bending (three points) and thus divided into two cubic pieces. Then, each piece was crushed to compression. See figure 2.

The activity index describes the degree of reaction time or reaction rate between a pozzolanic material and the $Ca(OH)_2$ in the presence of water. It is given by the ratio between the compressive strength of a mortar with the addition of p% of coal waste and the compressive strength of a reference mortar with 100% cement [7].

The activity index measuring on a standardized mortar is done by the following formula:

$$\mathbf{I = fc_{90}\ (with\ 25\%\ of\ waste)\ /\ fc_{90}\ (without\ addition)}$$

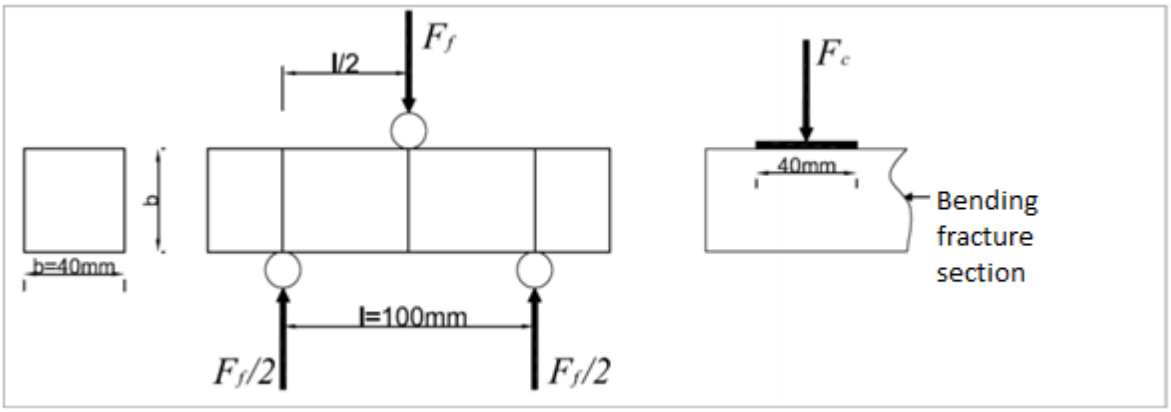

Figure 2: A flexure-compression on 40x40x160 mm^3 samples

2.2.2. Tests on mortars with coal waste as sand replacement:

In this second part, we reformulated a mortar with coal waste as sand replacement (in volume), taking only the granular fraction 0/5 of waste by sieving, then grinding the largest fraction and sieving.

Below, in table 1 and figure 3, we represent the particle size analysis of the mixture of waste reduced to the fraction of sand 0/5:

Table 1: Particle size distribution

Sieve Size (mm)	Cumulative passing (%)
5	100%
2,5	74%
2	66,80%
0,63	31,20%
0,315	20%
0,16	11,20%
0,08	6,20%

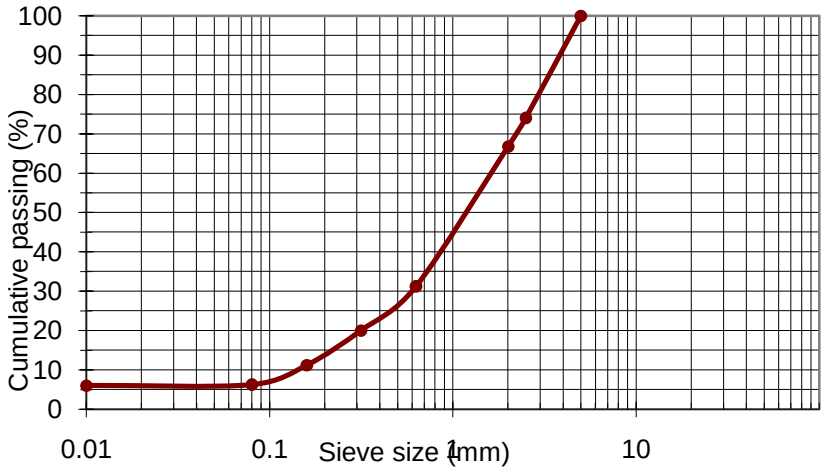

Figure 3: The particle size curve

For the following tests, we made normal mortars, in cylindrical pieces 40x60 mm^2 with different coal waste percentages as sand replacement (in volume): 0%, 10%, 20% and 50%. Composition of the different mixtures is given in ***Table 2.*** After 24 hours, samples are removed from the molds and cured in the wet room during 90 days at 20°C and 100% of relative humidity, we let them dry in the oven at 105°C until a constant mass was obtained, and then we divided them into three parts: one part of the specimens was left at 105 °C, the second underwent a heat treatment at 400 °C and the third at 600 °C. Thus, we could observe the impact of heat treatment on the mortars.

For each mixture and for each test, three samples are tested and the mean value is considered.

Table 2: Composition of different mixtures.

Mixture – Coal waste aggregates content*	0%	10%	20%	50%
Water (kg/m^3)	256.5	256.5	256.5	256.5
Cement (kg/m^3)	513	513	513	513
Sand (Kg/m^3)	1539	1385	1231	769
Coal waste (Kg/m^3)	0	154.28	308.56	771.41

* Volume replacement of sand by plastic aggregates.

a- Porosity Accessible to Water:

The porosity may be opened (i.e. accessible to water) or closed. Its measurement indicates the percentage of voids within the mass of the mortar, which are connected with the surface. This test is performed according to RILEM recommendation 49TER [8].

The apparent porosity (Pa) is calculated using the following formula [9]: $\boldsymbol{P_a = \frac{M_{sat,air} - M_d}{V}}$

Where: M_d : the dry mass (g), $M_{sat,air}$:the saturated mass in air (g)

b- Apparent density:

The apparent density ρ_d (or dry density) is calculated with the formulas below [9]:

$$\boldsymbol{V = \frac{M_{sat,air} - M_{sat,water}}{\rho_{water}}} \quad \text{and} \quad \boldsymbol{\rho_d = \frac{M_d}{V}}$$

Where: $M_{sat,water}$: the immersed saturated mass in water (g) and ρ_{water} : water density.

c- Gas Permeability:

The permeability of a material reflects its ability for passing through a fluid under a pressure gradient. It depends on the porosity, the pore geometry, the pore tortuosity, and most importantly the pore connectivity of the material [10].This physical property is governed by Darcy's law:

$$\boldsymbol{Q = \frac{k}{\mu} A \frac{dP}{dx}}$$

With Q: gas volume flow, μ: Viscosity, A: Section of the test material and dx: thickness crossed.

To calculate the apparent permeability, the containment pressure is set at 8 bars, and the percolation pressure is taken equal to 2 bars. After stabilization of the gas flow, the value of Q (m^3/s) is taken to determine K_a (m^2), which depends on P_i (gas percolation pressure) according to the following formula: $\boldsymbol{K_a = \frac{2QL}{A} \left(\frac{Patm}{Pi^2 - Patm^2}\right)}$ where:

- L and A are respectively the length (m) and the section (m^2) of the test piece;
- μ is the gas viscosity at 20 °C (μ_{helium} = 0.000194 Po = 19.4 *10-6 Pa.s)**;**
- $P_{atm}$is the atmospheric pressure at the gas outlet (= 1bar)
- P_i is the inlet pressure in bars.
- Q is the measured gas flow (m^3/s).

This test was conducted according to recommendation standard RILEM TC 116-PCD [11].

d- Ultrasonic Wave Propagation:

Measuring ultrasonic wave propagation is certainly an easy and non-destructive process to determine the material quality [12], yet very demanding since it depends on the shape, distribution, crystallographic orientation and size of the defects (pores and cracks) as well as the presence of interstitial fluids.

For our study, the sound velocities were measured by the transmission method, by arranging two transducers (transmitter and receiver) on either side of a test piece of known length (40mm x 60mm) to measure the time the wave takes to pass through.

The waves were produced by an ultrasonic device (Sofranel 350M), see figure 4.

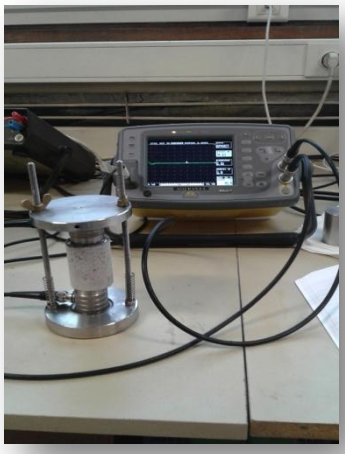

Figure 4: Experimental device for measuring ultrasound

3. Results and Discussion:

3.1.Characterization of Coal Waste of Jerada:

A granulometric analysis to the laser provides the granular distribution of the mining waste powder in Figure 5. Comparing it with that of cement CEM I 52.5 ***(Figure 5)***, we note that the coal waste powder size is slightly finer than that of the Portland cement specimen used.

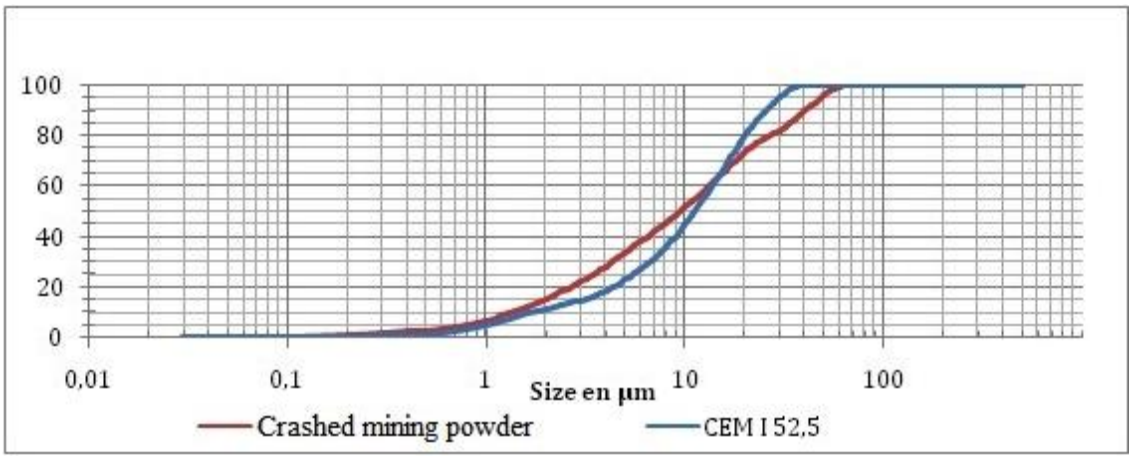

Figure 5: Comparing the size of mineral powder and cement CEM I 52.5

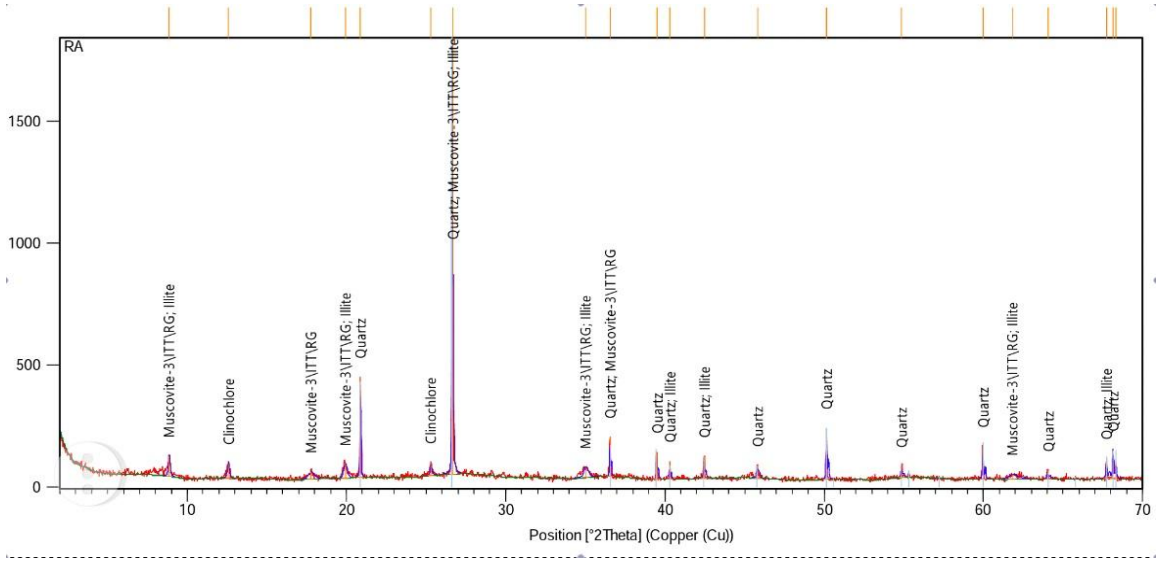

Figure 6: Diffractogram of the sample of Coal Waste of Jerada.

Table 3: Chemical analysis of the spectrum 51

Element	% Mass	% Atomic
C	**34.40**	**46.21**
O	**40.69**	**41.03**
Al	**7.64**	**4.57**
Si	**9.81**	**5.63**
S	**0.40**	**0.20**
K	**2.29**	**0.95**
Ca	**0.20**	**0.08**
Ti	**0.13**	**0.05**
Fe	**4.45**	**1.28**

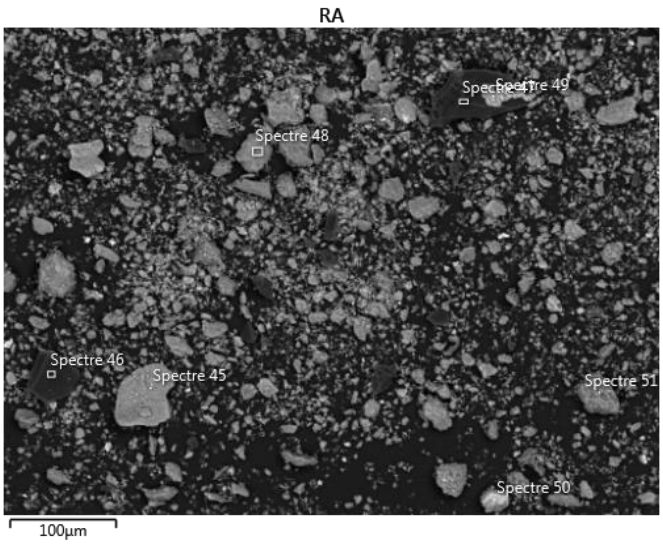

Figure 7: MEB image of waste coal

It has a density of 2.66 t/m3 and the nitrogen adsorption-desorption test gave a BET surface area [13-14] equal to 16.1633 m²/g, against 1,47m²/g for cement, which would allow a fast interaction with water in the cement paste.

Chemical and mineralogical analyzes of this coal waste carried out to characterize these sterile showed the presence of low levels of heavy metals (lead, chromium, iron) and a small percentage of Sulphur.

The diffraction test X-ray performed with a Bruker D8 Discover diffractometer, showed the presence of a significant amount of quartz in addition to clays (muscovite, illite and clinochlore) which explains the schistose nature of the waste ***(Figure 6)***.

Observation by scanning electron microscope of a sample of sterile waste coal shows the presence of a porous structure with an angular grain morphology***(Figure 7)***, and analysis of elementary spectra shows a strong presence of carbon and oxygen, a significant amount of titanium, aluminum and silica with some traces of iron, potassium, calcium and sodium ***(Table 3).***

Thermogravimetric analysis was performed using a TGA / DSC on a sample of coal waste previously dried in the oven at 60 ° C.

This analysis gives the variation of the mass of the sample as a function of temperature, as shown by the curve in ***figure 8***.

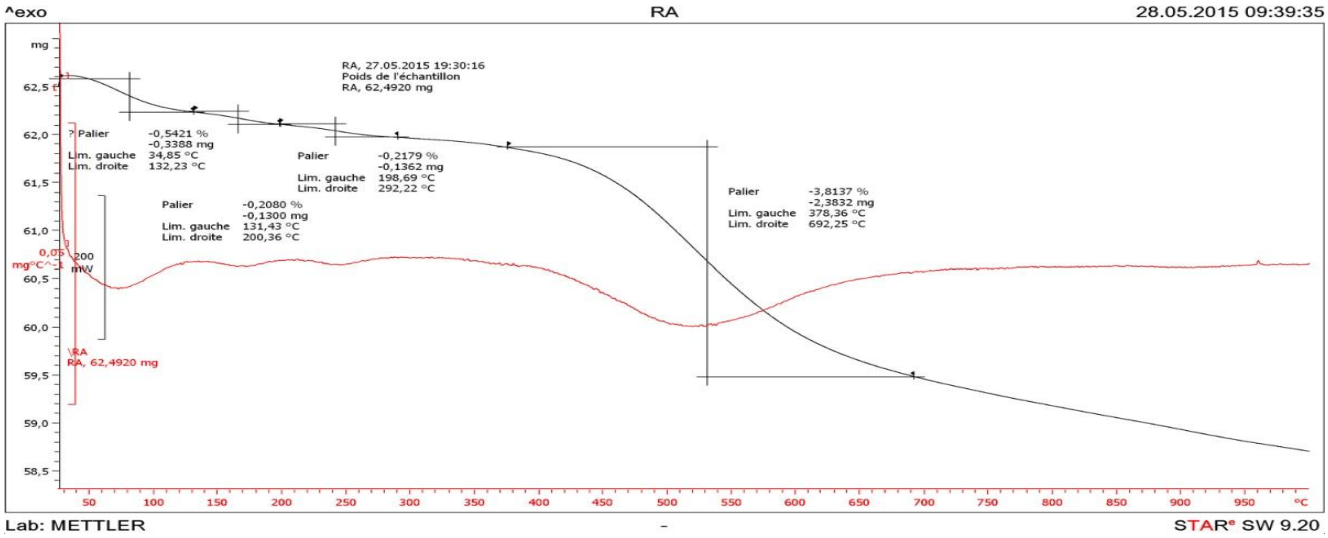

Figure 8: Curve of the sample thermal analysis.

This curve shows a first peak between 100 and 150 ° C, attributed to the departure of the water contained in the sample; there is subsequently a more significant peak at 535 ° C, relative to the combustion of the coal contained in the sample portion.

This led us to study the thermal behavior of a mortar with these coal waste additions before and after this observed peak to predict degradation at temperatures exceeding 460 ° C, due to the transformation of portlandite into CaO.

Above this temperature, the combustion of coal waste will induce a loss of mass, thus the possibility to form a lightweight material with high porosity and good thermal insulation properties.

3.2. Activity Index:

The norm ASTM C618 provides a minimum of pozzolanic activity index equal to 67% at 28 days, with a 25% substitution percentage. In our case, we obtained an activity index I_a = 0.56, less than the normative limit of 0.67.We conclude that this sterile coal waste does not exhibit pozzolanic reactivity.
Therefore, the use of this coal waste in mass replacement of cement mortars is not interesting and rather unfavorable for its poor mechanical strength. This is why we will explore its use as partial substitution of the sand in the mortar.

3.3. Porosity accessible to water and apparent density:

The evaluation of the effect of coal waste on the porosity accessible to water, shows after 90 days of treatment that the reference mortar (0%) is less porous than mortars containing coal waste. Indeed, it has a porosity of 20.43%, against 20.7%, 21.01% and 21.41% for mortars with 10%, 20% and 50% additions respectively.

This porosity increases even more with the heat treatment of these mortars at 400 °C, but particularly at 600 °C, to 23.4% for the composite of 50% coal waste, treated at 400 °C and 25.69% for mortars (50%), treated at 600 °C. This increase in porosity is mainly due to the combustion of coal content in the waste, at around 530 °C (See ***Table 4*** and ***Figure 9***)

This increase in porosity with the percentage additions results in lower apparent density(more emptiness, thus more lightness), due to the density of the waste which is almost equal to that of the sand used (ρ_{waste} = 2.66 and ρ_{sand} = 2.65). (See ***table 5*** and ***Figure 10***)

Table 4: Pore percentage in the mortar pieces

Percentage of waste in mortars	Interconnected porosity		
	Without heat treatment	Heat treatment at 400°C	Heat treatment at 600°C
0%	20.43 %	20.52 %	24.94 %
10%	20.76 %	21.22 %	25.41 %
20%	21.01 %	22.09 %	25.46 %
50%	21.41 %	23.47 %	25.69 %

Table 5: Apparent density of the pieces.

Percentage of waste in mortars	Apparent density in Kg/m^3		
	Without heat treatment	Heat treatment at 400°C	Heat treatment at 600°C
0%	2038.23	2032.66	1970.81
10%	2037.79	2030.13	1967.44
20%	2028	2020.4	1964.33
50%	2028.9	2003.44	1940.74

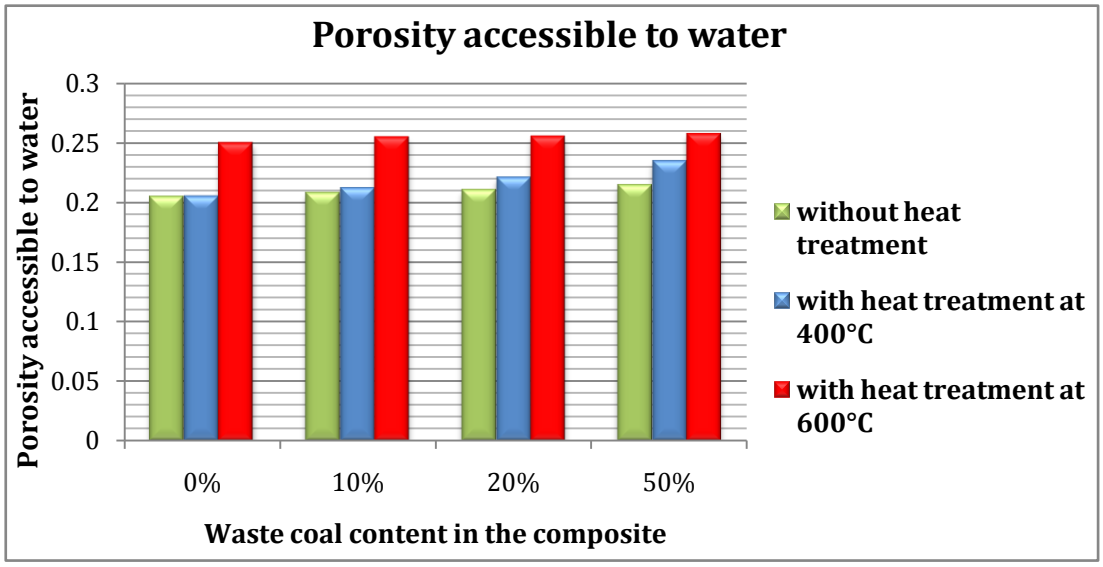

Figure 9: Evolution of porosity in different composites with heat treatment

3.4. Gas Permeability:

For each composition, the samples were tested under a percolation pressure P = 2 bars to identify the apparent permeability. ***Table 6*** summarizes the mean values of apparent permeability obtained for each composite, with and without heat treatment at 400 ° C and 600 ° C. See ***Figure 11***.

These results demonstrate that the incorporation of coal waste tailings causes a reduction in the apparent permeability. This leaves us to assume that despite the composite porosity, the pores are not interconnected and therefore the passage of gas is not warranted. Moreover, after heat treatment of the mortars studied, the value of the apparent permeability increases for each composite. For example, in composites with 50% of coal waste, K_a changes from 1.34.10-15 without heat treatment to Ka = 1.89.10-15 after exposure to temperatures of 400 °C. This increase is more significant after heat treatment at 600 °C

where Ka = 9.54.10-15. This can be explained in two ways. On one hand, the burning of coal in the waste, which occurs at around 530 °C, releases a pore volume in the mortar and subsequently increases its gas permeability. On the other hand, this increase is due to the decomposition of portlandite in the cement paste beyond 450 °C.

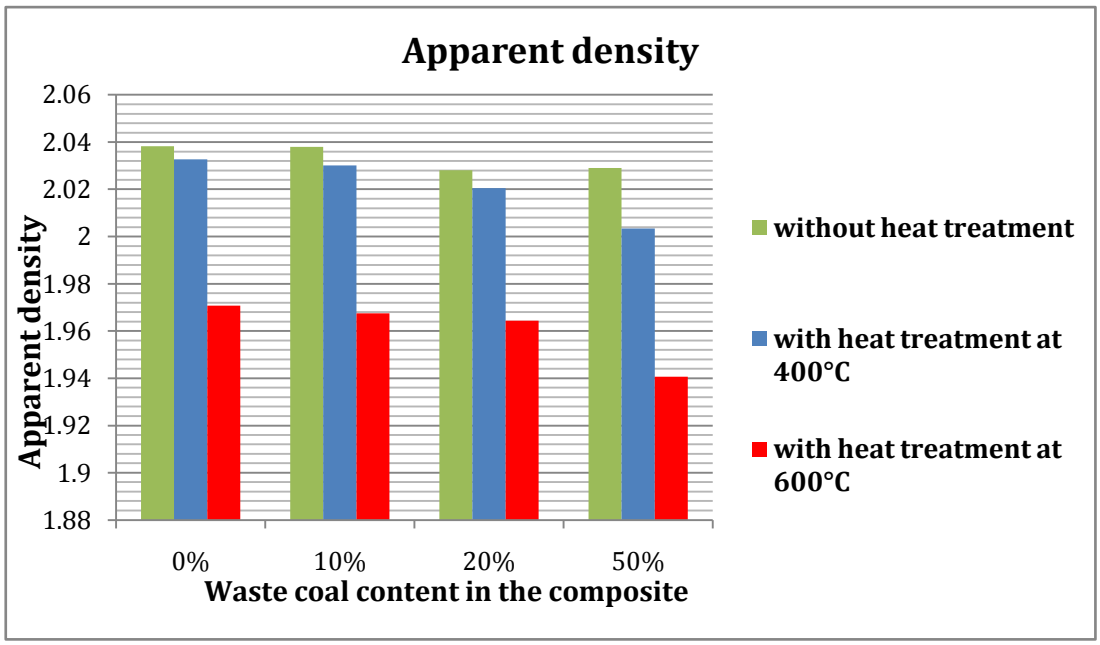

Figure 10: Evolution of density in different mortars with heat treatment

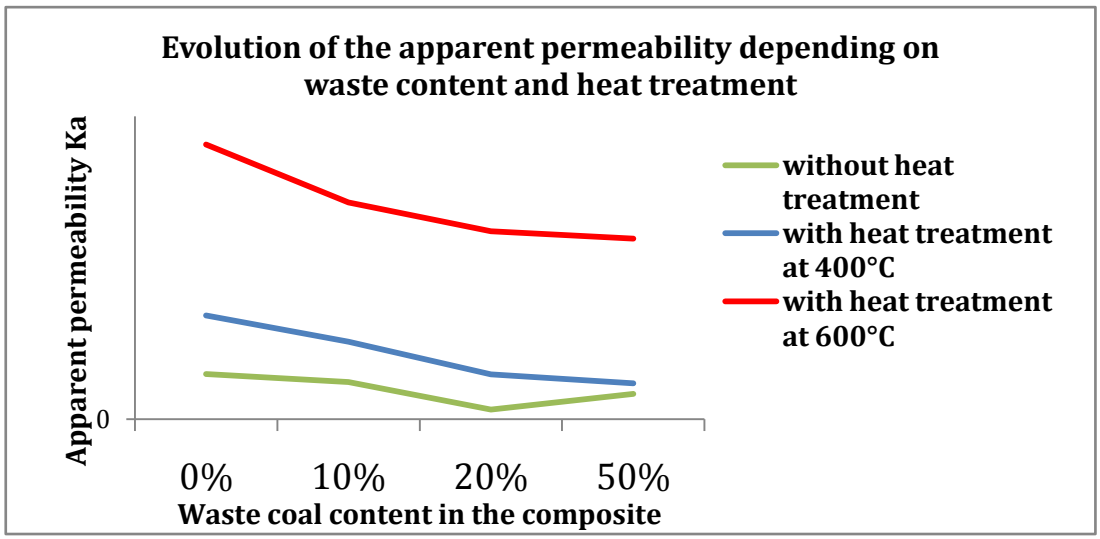

Figure 11: Evolution of the apparent permeability in mortars with heat treatment

Table 6: Apparent permeability of the composites

Percentage of waste in mortars	K Apparent, P= 2 bars		
	Without heat treatment	Heat treatmentat 400°C	Heat treatmentat 600°C
0%	2.39214E-15	5.47877E-15	1.45024E-14
10%	1.96808E-15	4.0941E-15	1.1449E-14
20%	5.04419E-16	2.3627E-15	9.92899E-15
50%	1.34044E-15	1.89809E-15	9.54368E-15

3.5. Ultrasound Propagation:

Ultrasound pulse velocity (UPV) values of different mixes are given in ***Figure 12***. Solid materials transfer the sound faster than porous materials [15].The values taken at the measurement of the propagation velocity of ultrasonic waves show that it is greater for the samples untreated with heat, and reduced with the thermal treatment at 400 ° C then further decreased under 600 ° C. This is due to the porosity of mortars which increases with heat treatment..

On the other hand, it is found that the ultrasonic wave propagation velocity decreases with the addition of coal waste in composite mortars. We attribute this to the increase in porosity due to the addition of such waste, which will mitigate the waves and reduce their spread.

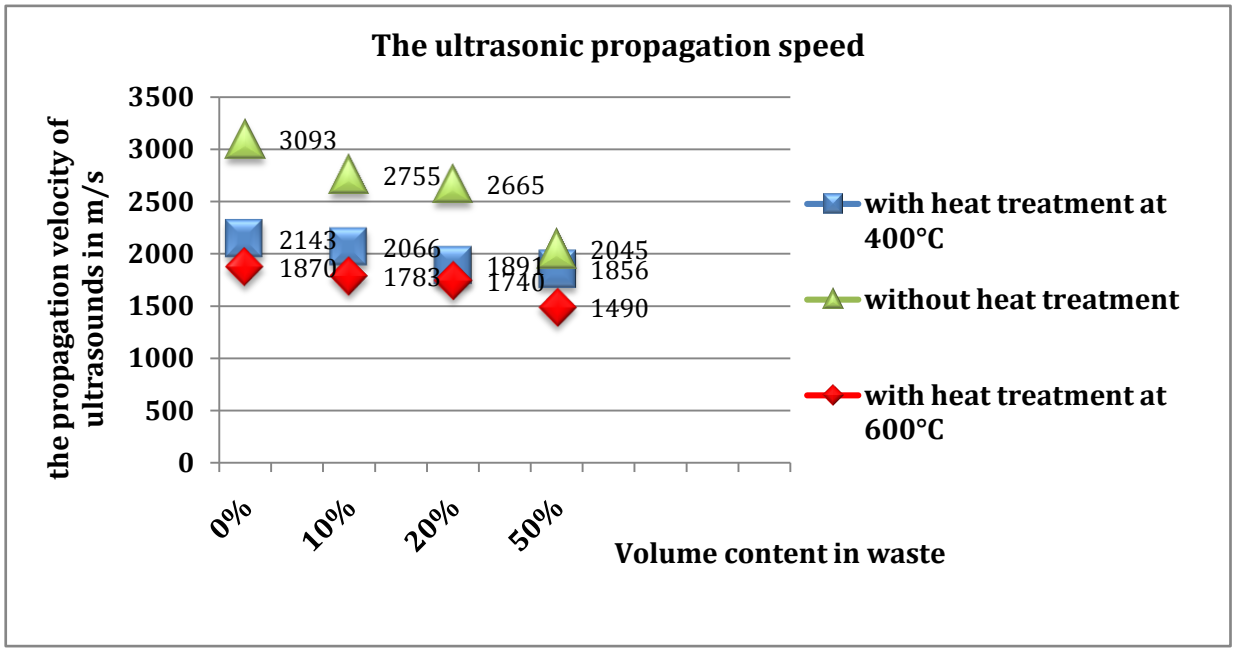

Figure 12: Evolution of the ultrasonic propagation speed in the composite with heat treatment.

Conclusion:

In the global energy context, one of the challenges is to cope in some cases with local shortage of materials, and the growing demand for natural resources in the field of construction. Thus, research for efficient solutions of alternative materials in line with sustainable development concerns are increasingly popular.

This work of recovery of coal waste tailings will provide an effective solution to the dual problem of saving resources and reducing environmental impacts.

At the end of this work, the track of re-using this waste as mass replacement of the cement in mortars was ruled out, giving way to the possibility of valuing the waste as aggregates in the concrete. The experimental results show that porosity increases with the addition of waste and even with the heat treatment, resulting in a remarkably lightweight mortar, it passes from 21.41 % for mortars consisting of 50% of waste, to 25.69 % for those treated in 600°C. However, the gas permeability decreases with the percentage of the additions, but it increases with the heat treatment, which is probably due to the creation of new cracks in the cement paste and to the pore volume liberated by the combustion of coal under the effect of high temperature. Finally, we notice that the propagation of the ultrasounds decreases with the addition of waste as well as with the heat treatment. Thus, the thermal processing of these composites is interesting to obtain lightweight and insulating materials.

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