Conference Paper

Use of Industrial Waste for the Optimization of Ceramic Construction Materials

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Received: 20 March 2020
Accepted: 30 April 2020
Published: 13 April 2020

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Selection and Peer-review under the responsibility of the RICON19 - REMINE International Conference Conference Committee.

OPEN ACCESS

Abstract

The massive growth of the ceramic industry and the consequent demand for construction materials worldwide has motivated the search for alternative solutions aimed at reducing the use of mineral / natural resources as the main source of raw materials. One of the strategies frequently adopted by the scientific community is the reuse of industrial waste. It is beneficial not only to reduce the overexploitation of mineral resources, but also to reduce the environmental, economic and social impacts resulting from their incorrect disposal / treatment and consequent deposition on land unsuitable for that purpose. Due to considerations such as physico-mechanical characteristics and the high production rate, two different types of industrial waste were selected for this work, ashes resulting from the burning of coal in a thermoelectric power plant, commonly known as bottom ash (B), and the Marble Powder (MP). It was intended to test the technological feasibility of the manufacture of ceramic materials produced from clay mixtures containing these two residues. For this purpose, the fine fraction (<63 μm) obtained from the sieving of the marble residue (MR) and slag (Bf) was used, as well as a coarser grain slag fraction ranging from 63 -125 μm (Bg). The resulting test samples were subjected to a firing of 950 °C under an oxidizing atmosphere, following a primary drying process. Faced with the standard values, the new ceramic materials obtained from MP have seen their mechanical and porous characteristics decrease and increase, respectively. At the same time, although the addition of B in no way influenced the mechanical characteristics, a significant improvement of the porous characteristics was observed. The incorporation of these residues produced a color very close to the original sample material.

Keywords: industrial waste, ceramic, construction materials, bottom ash, Marble Powder

1. Introduction

It is well known that the construction industry is an important consumer of resources and is also responsible for its processing and consequent application in a wide range of areas. All this process presents as main inconveniences the use of exorbitant amounts of energy, resulting in superfluous expenses and, derived from the abusive use of natural resources, the disappearance of multiple reserves of raw materials. Associated with this issue is a large number of waste from a wide range of manufacturing industries which, due to their individual properties, do not present any economic viability. This waste is usually stacked in waste dumps, causing multiple visual, environmental and even financial impacts.

Clay, the main raw material used in the manufacture of ceramic building materials, which includes products such as bricks, tiles and ceramic tiles, is one of the most used resources in the construction sector. For other hand, in order to try to solve the excess of residues, which Man produces on a day basis, a need to take advantage and make them useful for society arises. The ceramic industry is one of the largest exploiters of the planet's natural resources, often opening open-pit explorations for the extraction of clays and other industrial minerals that are critical to their livelihoods, causing environmental impacts and the squandering of areas that despite of being recovered environmentally will not be what they initially were. Thus, the incorporation of ashes resulting from the incineration of mineral coal from power plants still working, or marble powder that comes from the industry of transformation of natural stone, result of the sawing of this type of rock [1], or even other types of residues like coffee waste [2], rocks and minerals processing waste [3, 4], municipal solid wastes is intended to reduce their amounts that will have to be landfilled and to provide a substitute for natural raw materials, contributing to the environmental sustainability and making possible the reduction of the manufacturing cost of ceramic material. Energy efficiency in construction materials is one of the major research concerns in the building sector nowadays, aiming to reduce energy consumptions and promote sustainability.

By 2050, the world is expected to generate 3.40 billion tons of waste annually, increasing drastically from today's 2.01 billion tons [5]. Waste generation is a natural product of urbanization, economic development, and population growth. As nations and cities become more populated and prosperous, offer more products and services to citizens, and participate in global trade and exchange, they face corresponding amounts of waste to manage through treatment and disposal.

It was estimated that only 55.2% of produced coal combustion residues are reused, mainly by the cement and mining industries [6], this solution cannot consume all the produced waste, thus, the necessity of environmentally friendly solutions for coal combustion residue disposal is still a concern.

In order to mitigate this problem, and create an ecological ceramic material, possible to insert in the ceramic industry, it were produced ceramic test samples, due to attributes such as physico-mechanical characteristics and high production rate, two different types of industrial waste were selected for this work: ashes resulting from the burning of coal in a thermoelectric plant, commonly known as bottom ash (B) and the Marble Powder (MP). This powder comes from the industry of transformation of natural stone, result of the sawing of this type of rock. It was intended to test the technological feasibility of the manufacture of ceramic materials produced from clay mixtures containing these two residues.

It is possible to predict the negative impacts resulting from the worldwide production of coal combustion (B) and sewage powder (MP). Because there is no solution to mitigate this problem, the authors studied slag-embedded ceramic materials resulting from this residues, with the aim of reducing both the volume of waste to be landfilled and the use of natural raw materials used for its manufacture. Improving this knowledge is a key challenge to avoid wasting or destroying the resources available. According to the data contained in the last two reports published [7], regarding the world production of ceramics, it can be seen that over the last decade, the world production of this type of material has shown a strong growth trend (Fig.1).

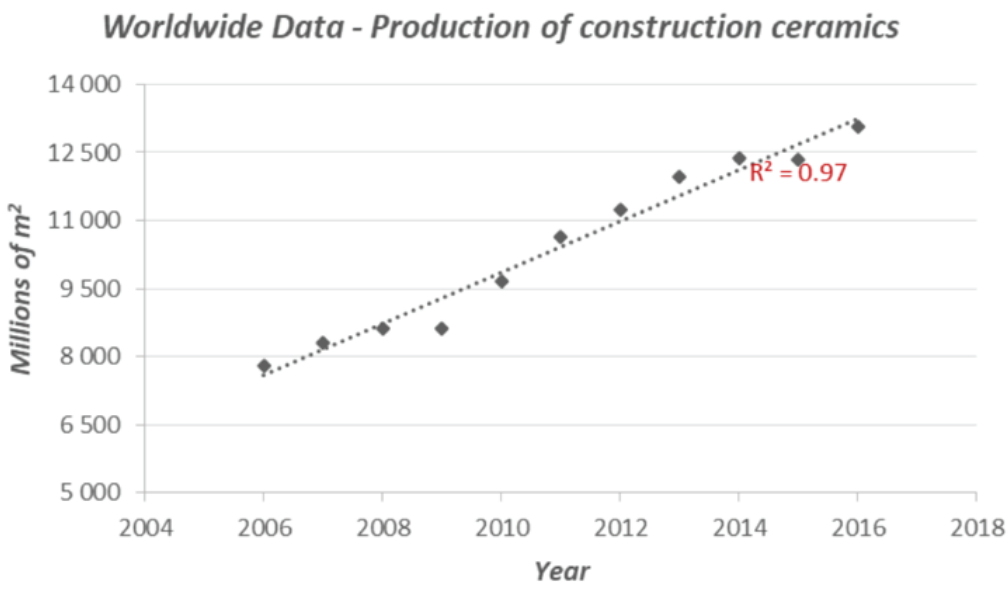

Figure 1: World production of ceramics between 2006 and 2016.

For this purpose, the fine fraction (<63 μm) obtained from the sieving of the marble residue (MR) and slag (BF) was used, as well as a coarser grain slag fraction ranging from 63 -125 μm (BG). In all, eleven different mixtures were made: a standard mixture, composed solely of clay, two containing 5% and 10% by weight of MP and the remaining weight with 5%, 10%, 15% and 20% BF and BG, respectively.

These powder mixtures were analysed for their extrusion feasibility from plasticity characterization. An equivalent amount of water was then added to all the mixtures and formed by means of a plunger extruder. The resulting test pieces were subjected to a firing process at 950 °C under an oxidizing atmosphere, following a primary drying process.

2. Materials and Methodology

As mentioned, one of the objectives of this work was the valorisation of two by-products which, given the existing quantities, caused problems of a different nature. As they were embedded in a clay matrix, it was possible to assign them a new utility, reducing not only their volume but also the amount of clay used in the manufacture of ceramic materials. As such, the realization of this work included the use of three basic raw materials: red clay, residues resulting from the sawing of Estremoz marble and bottom ash from the Sines Thermoelectric Power Plant. The clay used in this work belongs to a company from the building ceramics sector that has been in the Iberian market for several decades, while the ashes are from the Portuguese Power Plant company, and the Marble power was collected in Estremoz region (South of Portugal), from a Marble exploration factory (Fig.2).

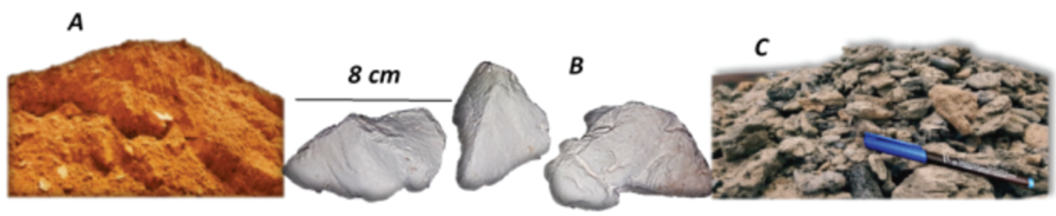

Figure 2: Raw materials used in this research: red clay, residues resulting from the sawing of marble and bottom ash.

The preparation of the ceramic samples began with the processing (milling and sieving) of the three basic raw materials, resulting fractions of less than 250 μm. Subsequently, the clay - ”residue(s)” mixtures were made, in percentage weight, and a specific amount of water was added for each one of them. A total of 25 different types of ceramic mixtures were formulated, differentiated by the individual addition (single mixtures) and combined (composite mixtures) of each of the by-products under analysis. For this purpose, these products were processed and individualized in: ash <63 μm (BF); ash 63 - 125 μm (BG) and marble powder <63 μm (MP). In a first phase, clay-ash (F and G) was mixed, in the percentages of 5%, 10%, 15%, 20%, 25% and 30%, and marble – clay (M) in the percentages of 2.5%, 5% %, 7.5% and 10%. A standard sample composed solely of clay (0% mixture) was also performed. In a second step, these elements (GF, GM and GFM) were combined in the percentages of 12.5%, 15% and 17.5%. Subsequently, the paste obtained was extruded through a manual extruder, sectioning it into 10 cm long specimens. After one day at room temperature, the generated samples were dried in an oven at 110 °C for 72 hours. The manufacturing process of the ceramic samples culminated with their firing in an oxidizing atmosphere (electric muffle) at a single temperature (950 °C). In total and finally, 580 specimens were extruded, dried and fired at 950 °C.

From all the preparatory stages until the obtaining of the ceramic samples, the elaboration of the mixtures was the one that required the greatest commitment and accuracy of execution. According to some authors [1], the addition of marble powder was beneficial for incorporation percentages around 5%, and can even be extended to values in the range of 15% [8], on the other hand, analysed the effect of the incorporation of ashes up to percentages of 20%, and established that the optimal percentage values for firing temperatures between 900 °C and 1000 °C, are around 10%; however, this author did not mention the maximum limit that clay can tolerate by including this type of material.

Figure 3 shows the visual dissimilarities, at the moment of the overlap of BF in clay, in the percentages of incorporation: 5%; 10%; 15%; 20%; 25% and 30%.

Figure 3: Contrast obtained when F is added to make the different mixtures of A - F.

The most used material in the development of this work was, without any doubt, clay (A), with about 37 kg used. The other components that follow are, respectively, Ash 63 - 125 μm (BG), with 3 kg used and, to a lesser extent, Ash < 63 μm (BF), with approximately 2.33 kg. By exclusion of parts, the component that had the lowest contribution in this study was marble powder < 63 µm (MP), with approximately 725 grams applied (Table 1).

Table 1: Quantity (g) of materials involved in the formulation of the compound mixtures.

		%	Clay (A)	Marble Powder < 63 µm (MP)	Ash < 63 µm (BF)	Ash 63 - 125 µm (BG)	TOTAL
BLENDED MIXTURES	G + M	12,5	875	25	–	100	1000 g
		15	850	50	–	100	1000 g
		17,5	825	75	–	100	1000 g
	G + F	12,5	875	–	25	100	1000 g
		15	850	–	50	100	1000 g
		17,5	825	–	75	100	1000 g
	G + F + M	12,5	875	12,5	12,5	100	1000 g
		15	850	25	25	100	1000 g
		17,5	825	37,5	37,5	100	1000 g
	Total		7650	225	225	900	

After 24 hours at rest of the mixture of the pulps, it was proceeded gradually, the unpacking of the ceramic pulps and its extrusion was performed immediately. For this purpose, after unpacking, the samples were inserted inside the extruder, and then compacted through the pressure exerted by a piston (Fig.4).

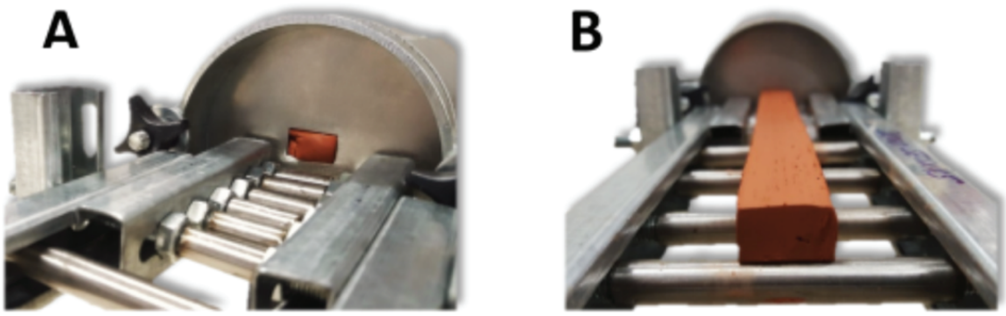

Figure 4: Extrusion of ceramic pastes: A - Compaction of the paste against the spinner. B - Shaping.

Each sample was double referenced on its upper surface, specifically at both ends, entering the acronyms F, G and/or M and the respective percentages of incorporation, according to the components present in the mixture (Fig.5).

Finally, the samples obtained were weighed in order to evaluate their mass loss in the different temperature stages. After being exposed to the open air for about a day, the drying of the specimens began with their transfer to a ventilated oven, where they remained for approximately 72 hours at a constant temperature of 110 °C. Finally, the

Figure 5: Referencing of ceramic specimens after their being smoothed.

specimens were then placed inside an electric muffle and submitted to progressive heating up to a maximum temperature of 950 °C during 1 hour. After first drying at 110 °C followed by sintering in an oxidizing atmosphere at 950 °C, the ceramic samples were subjected to several technological tests (physical and mechanical) in order to evaluate their behaviour in the presence of different quantities of M, F and G. Initially, as the specimens came out of the oven and the muffle, they were successively measured and weighed in order to determine their linear shrinkage and loss in mass, respectively. The flexural strength test was then carried out and each specimen was inevitably divided into two distinct halves. Afterwards, these two halves were selected and the water absorption tests were carried out (by capillary effect, at atmospheric pressure and by vacuum).

3. Results & Discussion

In order to understand the behaviour of the ceramic samples manufactured, it was fundamental to analyze the raw materials that were at its origin. In addition to having the function of characterizing each of the raw materials involved, these analyses allowed a better understanding of the results obtained by the specimens when subjected to the most varied tests.

When analysing Table 2, it can be seen that, for each of the raw materials used in the production of the ceramic test-pieces (A, F, G and M), the oxides of the respective main elements were determined, as well as their values of loss on Ignition (LOI).

In a first approach, it is observed that both clay and ash (in both configurations) share as majority elements SiO_2, Al_2O_3, Fe_2O_3, CaO and K_2O. The marble, as one would expect for its mineralogical constitution, predominantly calcitic ($CaCO_3$), exhibits a very high concentration of CaO.

TABLE 2: Oxide content (% by weight) and loss on Ignition (LOI) - Obtained by XRF

	Clay (A)	Ash < 63 µm (F)	Ash 63 - 125 µm (G)	Marble < 63 µm (M)
SiO_2	49,425	48,433	48,007	2,404
Al_2O_3	20,664	16,098	16,321	0,625
Fe_2O_3	7,995	5,891	6,574	0,194
CaO	5,601	2,514	2,747	52,199
K_2O	4,247	1,290	1,202	0,169
MgO	2,894	1,126	1,197	0,631
TiO_2	0,877	0,796	0,818	0,024
Na_2O	0,292	1,130	1,151	0,031
P_2O_5	0,162	0,229	0,259	0,011
MnO	0,119	0,059	0,039	0,008
SO_3	0,042	0,027	0,017	0,017
LOI	7,470	22,080	21,360	43,660

With regard to the loss of weight, figures 6 and 7, relating to ashes (F and G) and marble dust (M), respectively, show the variability of mass trends, with the increasing addition of these same elements.

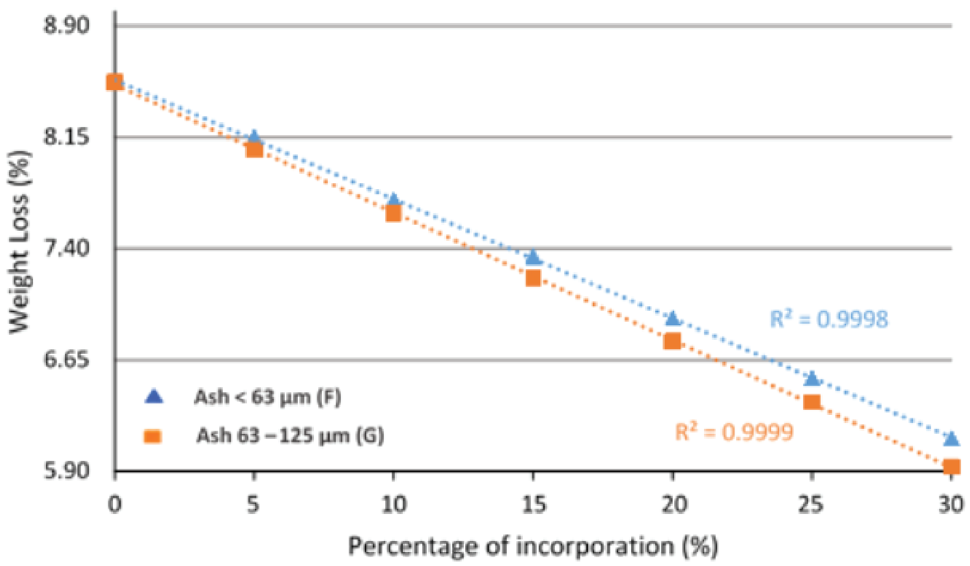

Figure 6: Change in mass (%) of test pieces made with F and G after sintering at 950 °C.

Through the analysis of the previous graph, it is concluded that, regardless of the granulometry used, the gradual addition of ash promotes an attenuation of the effect of loss in weight of the specimens after their firing, when compared with the standard value.

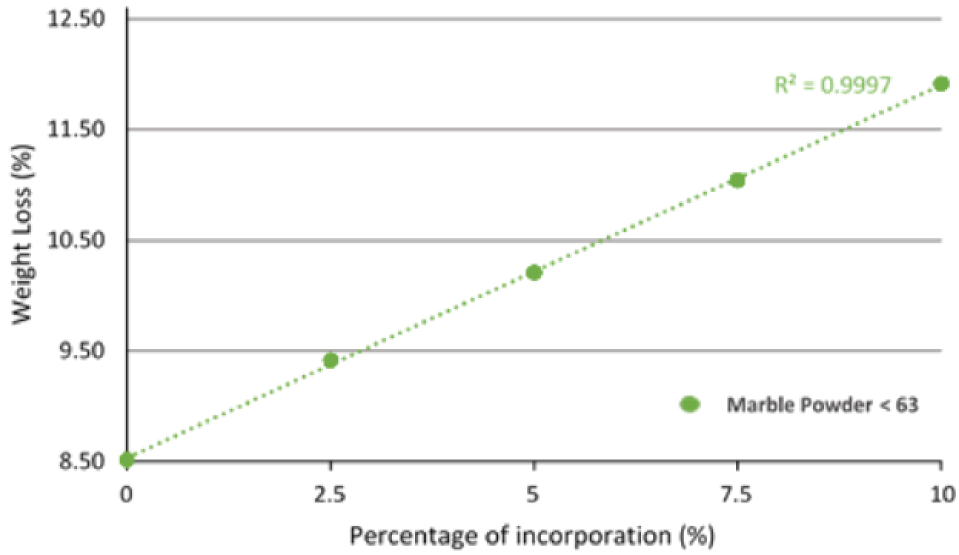

Figure 7: Change in weight (%) of test specimens made with M after their sintering at 950 °C.

By observing the graph relative to the mass variation of the samples with M (Fig.7), the opposite of what happened in the case of the ashes (F and G) is observed. Since this element (M) is mostly made up of $CaCO_3$, it is natural that, with the increase of the burning temperature, the volatile elements associated with this component "volatilize" leaving empty spaces (pores) in their place.

The measurement of the flexural strength of simple ceramic samples has acquired additional importance. The purpose of this test was to evaluate the influence that the different components would have on the strength of the formulated ceramic materials, as well as to understand the maximum tolerance of the clay when including these products in its composition. Take a look at the graphs shown in figures 8 and 9, which illustrate the different behaviours of ceramic bodies formulated from ash (F and G) and marble dust (M), respectively.

In a first approach, from the analysis of the previous figure it is concluded that the clay used in this work reveals a great tolerance to the addition of ashes, even supporting percentages of up to 25% of this product without excessively conditioning the mechanical strength of the specimens made. Considering the cases in which percentages of 25% were incorporated, the variation between the resistance evidenced by the reference samples and the samples conceived with this percentage level is - 0.64 MPa (≈ - 2.5%), thus guaranteeing an excellent compromise between the quantity of raw

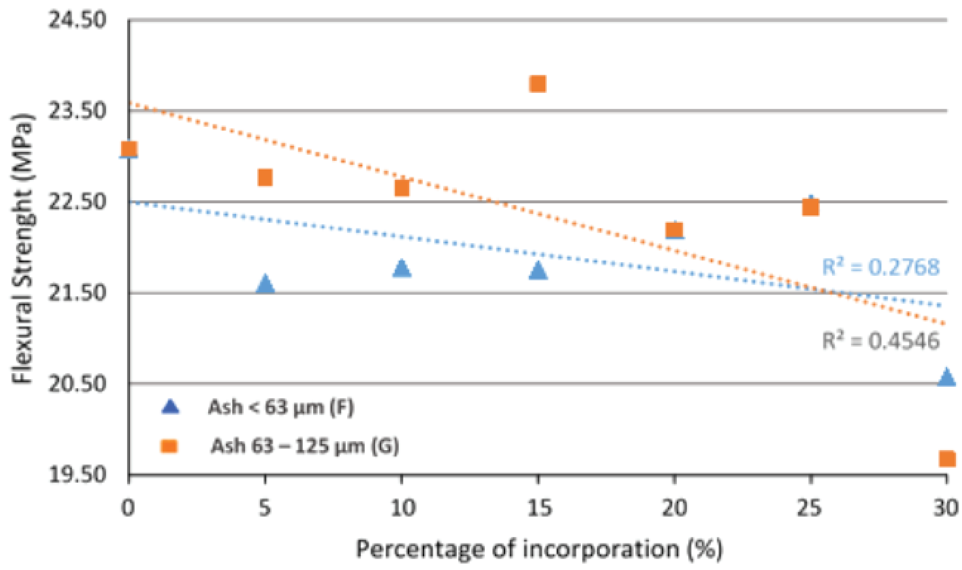

Figure 8: Value of flexural strength (MPa) evidenced by samples made with F and G, after its sinterization at 950 °C.

material used and the reused industrial products. Therefore, 25% of the incorporation of this material allows to reach the best ratio between the total quantity of ash included and the resistance of the ceramic specimens. When carrying out the realization of ceramic bodies with this percentage level of ash, considerable amounts of clay will be saved, attributing a new functionality to a product of high potential, existing in very significant quantities.

Regarding the addition of M (Fig. 9), this element, in percentage terms, generally promotes a more noticeable reduction in strength values when compared to ash. When calcium carbonate is added in small quantities, it may act as a melting agent; the percentage of incorporation at 5% of M supports this statement, with a value practically identical to the resistance shown by standard samples (0%).

4. Conclusions

Given the enormous quantities of ceramic materials produced worldwide, it is urgent to adopt processes that lead to their optimization, not only in order to improve their technological characteristics, but also to create measures aimed at reducing the recurring environmental impacts of their massive production.

In short, the addition of ashes and marble dust promotes a slight decrease in the mechanical resistance of ceramic specimens; however, if added in the right quantities,

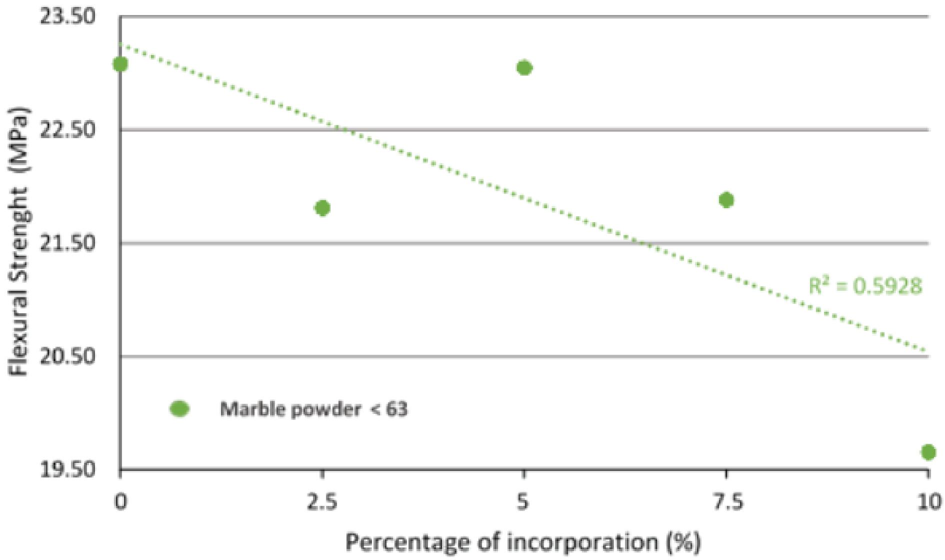

Figure 9: Value of flexural strength (MPa) evidenced by samples made with M, after its sinterization at 950 °C.

these elements have the capacity to slightly increase these values. In this sense, ash G (63 - 125 μm) was the element that presented the best individual results.

It is concluded that the samples containing amounts of M $\leq$ 5% were those with the best results. This fact proves the application value of small percentages of $CaCO_3$, demonstrating the validity of this as a structural "binder". Although the results are encouraging, in order to take full advantage of the addition of ashes, they should be combined with the element M.

Another important characteristic that must be taken into account is the loss of weight after the firing of the different ceramic materials. This can dictate that an entire batch is not viable due to non-compliance with requirements such as the weight of the material itself. In this sense, the loss in weight of the different samples was evaluated, and it was concluded that both ashes reduce the effect of the loss in weight, while the addition of M increases these values. The specimens with lower mass loss were formulated using the individual addition of F and G, this reduction being proportional to the percentage of incorporation of these elements.

Regarding the mechanical resistance of ceramic samples designed at the sintering temperature used, it is important to note that, except for the mixture of G at 15%, all simple formulations obtained resistance values slightly lower than those found in standard samples. These values present an additional highlight for the mixtures of F and G at 30% where there was a clear decrease of this property. From this result it was possible to conclude that, both for the clay and for the forming method used, incorporation percentages higher than 25% are not viable.

Regarding the addition of M it is concluded that the maximum percentages of incorporation should not exceed 5% in order to generate additional benefits.

In short, it is possible to conclude that the objective set has been fully achieved. From the combination of three materials without apparent utility, it was possible to substantially improve the technological properties of ceramic materials. This work proves the countless valences of the use of these by-products and challenges the ceramic industries to study and later use them. In this way, in addition to the quantities of raw materials saved, ecological ceramics will be produced, assuming a clear alternative to the use of traditional building materials.

Authors would like to thanks to Diogo Seixas for making this work available. To Geobiotec research Center where the tests were performed and the materials analyzed. To Engª Erika Davim and Filipe Davim for their support in this research. To Sines Thermoelectric Power Plant for the ashes.

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