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Influence of the addition of waste graphite powder on the physical and microstructural performance of hydraulic lime pastes

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HIGHLIGHTS

- Graphite powder by EDM can be used as filler in hydraulic lime pastes.
- EDM graphite is compatible with hydraulic lime pastes.
- Graphite addition increases 3 times the compressive strength.
- Graphite reduces the open porosity and the absorption coefficient.
- Thermal conductivity increases up to 80% with the graphite addition.

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ABSTRACT

The use of waste materials contributes to the improvement of the life cycle assessment of building materials, while traditional binders with a low carbon footprint enhance building sustainability. Isostatic graphite powder waste has not been used in the industry in spite of the huge quantities of it around the world. However, hydraulic lime is a traditional binder which requires low energy during manufacture. Hence, the aim of our research was to investigate the possibility of incorporating graphite waste powder into hydraulic lime pastes. Batches with different hydraulic lime NHL-5 replacements by graphite, ranging from 0% to 50%, with and without plasticizer were blended. Their mechanical characteristics and physical characteristics, namely, water absorption and thermal properties, together with the microstructure were analyzed. Among other effects, the addition of EDM graphite increased the compressive strength of the samples by almost three times while reducing the open porosity and the water absorption coefficient, enabling a wider use of these limes in building applications.

1. Introduction

Sustainability is an abstract term which is used to designate the balance between human activity and its consequences in the environment. Society has become progressively aware of the implications of human activity in climate change, and a reduction in the environmental impact of the production processes has become an urgent necessity. From this point of view, the use of waste materials has rocketed in recent years as it contributes to an improvement in the life cycle assessment. In this regard, several guidelines and legal frameworks have been approved by the European Union since 2002 [1,2]. The aforementioned strategies have been set to reduce landfill waste and to re-cycle it as a valuable resource for industry. Graphite is a material that combines metallic and ceramic properties. It is extremely hard, and can be used in roads and thin ceramic plates [3,4]. Its great heat and chemicals resistance makes it useful for many applications in industry such as plastic insulation, thermal-conductors or in silicon semiconductor elements, among others. As regards this, one of its main uses is in the production of Electric Discharge Machining (EDM) whose procedure was previously explained in detail by the authors [5]. As a consequence of this industrial appliance, about 14,600 tons/year are produced worldwide [5]. This waste was not included into

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the European Waste Catalogue and Hazardous Waste List [6] nor in the catalogue of recycled products in the Spanish Standard [7], nor in the Environment Protection Agency [8]. In spite of its importance, no other studies, apart from the aforementioned research into gypsum pastes, have been found in the literature to propose revalorization, reevaluation and recycling of this type of EDM graphite waste as an addition to building materials [9,10]. Indeed, there is a lot of research into the different types of graphite such as expanded graphite [11–15], exfoliated graphite [16], carbon nanotubes [17] or oxidized carbon nanofibers [18] but all of them are in the nanoscale. Apart from this, their use was proposed in a cement matrix [13,17,19,20] or concretes [18,21] in order to enhance the bonds with calcium silicate hydrate. As well as these uses this research deals with a type of crystalline isostatic synthetic graphite coming from Electrical Discharge Machining (EDM), which is classified as «biologically inert material and producing dust classified as a nuisance» [22].

On the other hand, lime has been the most important binder in the history of construction. However, it became obsolete during the 1970's [23] due to the greater and faster strength of Portland cement. The incompatibility of this cement in traditional masonry as well as a greater environmental awareness in the recent decades has given rise to increased interest in this binder in the building sector. Not only as regards the properties of the material itself, but also its lower energy demand and CO_2 emissions compared to cement manufacture. In fact, the latter is one the key issues in the construction sector for sustainability. It is based on the large sources of limestone and the low temperature required to burn the raw material. Hydraulic lime is mainly made up of calcium silicate and portlandite although, depending on the raw material, it could also contain aluminate of calcium. Its calcination temperature should be under 1250 °C to generate the calcium silicates which are responsible for strength of the hydraulic lime. This binder had been used not only in exterior plasters but also in mortars and grouts in the fields of bioconstruction and restoration as an alternative to cement. In the last decade, some research came up with the idea of the partial replacement of the lime by additions which are better for the environmental and energy consumption associated with the production of the lime [24] while increasing the strength [25–28] and durability of the lime-based composites [25,29,30]. Furthermore, additions were included to analyze their effect on the rheology [31,32], mechanical properties [33,34] or water absorption [30,35], among others.

Some of the additions have been successfully used in other binders, such as cements. They boosted the composite strength through the densification of the matrix and the increase in its contact surface. The additions commonly used in hydraulic lime composites are metakaolin [25,26,29,36–38] which is used even in lime concretes [24,26,39], pozzolans [25,40], silica fume [25,27,41], fly ash [31], pulverised fuel ash [42], ground granulated blastfurnace slag [42–43], cement kiln dust [44], calcined clay [42], red and yellow bricks [42], ceramic wastes [45], expanded clay residue [28], rice husk [37,42,43], diatomite [24], microsilica [42], nanosilica [46–48] and binary and ternary batches with ground granulated blastfurnace slag, fly ash and metakaolin and red-brick dusts [27,39]. However, no previous research has been found using isostatic graphite powder from the milling production of molds for Electrical Discharge Machining (EDM) in hydraulic lime pastes.

Furthermore, the use of chemical admixtures in lime pastes was recently analyzed by different authors to reduce, among other things, the water content while increasing strength and flowability in mortars and grouts. They were based on polycarboxylate [38,41,46,48], polynaphtalenesulfonate [47], lignosulfonate [47], hydroxypropylmethyl [32] cellulose derivative and perlite [38] and others [32]. Differences have been found in them depending on their nature as well as the binder and additives.

Hence, the aim of this paper is to investigate on the possibilities of isostatic graphite waste as an addition in the productive chain for lime-based products, and their enhancement in the building industry. Both the fresh and hardened properties were tested to analyze their effectiveness.

2. Experimental studies

2.1. Materials

The materials necessary to produce the lime pastes were natural hydraulic lime and graphite waste. A commercial natural hydraulic lime type NHL-5 in accordance with EN 459-1:2001 was used. According to the company, it was made up of portlandite (38%), belite (28%) and calcite (21%) while its fineness and bulk density are 0.9837 m^2/g and 769 kg/m^3, respectively.

Graphite waste is synthetic pure graphite, a specific form of carbon [5]. Its properties as waste are 441 kg/m^3 of bulk density, a BET surface area of 26.3078 ± 0.2622 m^2/g and a micropore area of 1.7052 m^2/g. Furthermore, the main particle size determined by laser diffraction and ranged from 1–10 μm, in which the peak from 35–150 μm was caused by the agglomeration of particles due to electrostatic forces [5].

At the same time, the plasticizer effect was analyzed through the slump test analysis and the Z potential of the samples through three different chemical admixtures. Firstly, a commercial plasticizer-air entraining agent, in accordance with ASTM C 457-71 and EN 934-3, was used. It was an aqueous solution of alkali products with minority components such as sulphuric acid and sodium salts which were used to stabilize several types of polymorphs [49]. Its bulk density was 1050 kg/m^3 and it will be known as p hereafter. A significant variation in the workability was observed during the tests which depended not only on the type of lime but also on the additions and type of additive and polymer used, among others [48–50]. Consequently, two other plasticizers were used. The former was an aqueous solution of magnesium lignosulfonate with deairants based on butilphosphates plasticizer, known as p1. While the second was an aqueous solution of ether-polycarboxilate polymer with deairants and a viscosity modulator based on organic polymers with a high molecular weight, known as p2. Their bulk densities were 1160 kg/m^3 and 1032 kg/m^3, respectively.

2.2. Mixture proportions

A control mix of hydraulic lime without graphite was made with a water/binder ratio (w/b) of 0.60. In order to analyze the effect of the addition of graphite, this ratio was maintained in all the samples, although it resulted in pastes of varying consistence. Natural hydraulic lime was replaced by graphite in five different percentages, namely, 5%, 10%, 15%, 20% and 25%, by weight. Complementary to those, batches with higher amounts of graphite were tested to analyze their effect on the mechanical strength. the w/b ratio in them was established according to the suitable flowability in the control samples (slump within 155 ± 5 mm). The graphite content was increased by four different percentages, namely, 25%, 30%, 35% and 50%.

All the series were known as ch_0%g, chp_0%g, chp1_0%g or chp2_0%g, where the letters *ch* refer to natural hydraulic lime; *p* stands for plasticizer, *p1* for plasticizer type 1, *p2* for type; 20%g means graphite and its percentage of replacement which varies from 0% to 50%, as previously mentioned;. The letter *v* at the end indicated a variation of the water content in accordance with the second criterion (the slump was maintained while w/b depended on the graphite replacement). The component contents of the different batches tested are specified in Table 1.

To facilitate the casting and mixture procedure, the graphite powder was added to the water to acquire a homogeneous distribution. Lime was incorporated at the end and mixed for five minutes at low speed. Due to the fine particles of the graphite, the kneading time had to be extended to guarantee the homogeneity of the blend.

Three samples of each batch were cast in standard prismatic moulds $40 \times 40 \times 160$ mm^3 for flexural and compressive strength as well as $40 \times 100 \times 160$ mm^3moulds for thermal conductivity tests. The blends were poured in two layers and each of them was compacted with 60 strokes from 15 ± 0.3 mm (EN196-1 2005). Samples were maintained under laboratory conditions at 20 °C and 60% of relative humidity. They were turned out of the mould after 7 days of curing and stored up to 28 days, 90 days and 360 days, under the same laboratory conditions, when the mechanical and physical tests were carried out.

2.3. Test procedures

2.3.1. Flow diameter

To determine the workability of fresh hydraulic lime pastes, the flow diameter test was carried out in accordance with the European standard EN 13279-2:2014. In this case, a cone of 50 mm top diameter, 100 mm base diameter and height 151 mm was used. The results were the average of three tests of each composition.