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Low embodied energy cement containing untreated RHA: A strength development and durability study

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HIGHLIGHTS

- Lower carbon cement of acceptable strength can be produced with up to 15% untreated RHA.
- *k* of 0.5 implies a low (<10%) addition potential for coarse RHA in concrete.
- Untreated RHA improved mortar sorptivity by 50%. Resistivity was retained to baseline levels.
- Carbonation depth doubled with 15% RHA use. This also occurs in most treated SCMs.
- A less pronounced negative effect was recorded against chloride ingress.

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ABSTRACT

In contrast to common approach – that often involves extensive pre-grinding – the utilization potential of rice husk ash (RHA: by-product of rice industry) as a supplementary cementing material, in its untreated form was examined. This is to meet industry's increasing awareness for lower embodied energy cement and concrete. Given the availability of current raw materials in cement manufacturing, what is needed is to be able to achieve an optimum approach, between sustainability and durability when designing concrete structures. Under this frame, as-produced RHAs (D50 of approx. 70 μm) were examined with respect to their inherent characteristics and their impact on the performance of cement-based mortars. Strength, hydration and durability properties were considered. Untreated RHA exhibits moderate pozzolanicity and a low efficiency factor of approximately 0.5–0.6. Its very high chemical reactivity (active silica ratio of approx. 90%) cannot be exploited since the lack of adequate specific surface slows down its engagement in hydration reactions. Taking into consideration their untreated nature, it can be supported that they exhibit an acceptable strength potential after 28 days, complying – even marginally – with the strength class (42.5) of the used cement. Durability results revealed that there is no need to pre-treat RHA in order to achieve equal or even better performance than mortar with plain cement in terms of sorptivity (50% increase for 15% RHA usage) and resistivity. Untreated RHA however fails – as most SCMs do – to improve carbonation resistance since for a 15% cement replacement, carbonation depth was almost doubled. Overall, results indicate that usage of untreated RHA in cement-based systems is an alternative to decrease their carbon profile, as long as cement replacement is kept relatively low (below 15%).

1. Introduction

Concrete is recognized to be the most widely used construction material, second only to water in total volumes consumed annually by society. It has been estimated that its average consumption is more than 1 m^3 per year per every person on the planet [1]. However, production of cement and concrete is associated with a significant environmental burden. For instance, the cement manufacturing industry accounts for 5–7% of the total CO_2 anthropogenic emissions [2]. Similarly, concrete production embodies significant carbon and energy footprint. Embodied carbon (energy) has been defined as the total carbon released (energy consumed) from direct and indirect processes with a product or service and within the boundaries of cradle-to-gate [3]. With respect to concrete, cement is its ingredient that contributes most to its embodied energy. It has been proved that sustainable development of the

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cement and concrete industries can be achieved by utilizing supplementary cementing materials (SCM) (fly ash, silica fume, ground granulated blast furnace slag, etc.), without minimizing the long term safety and durability of the structure.

The link between durable design and sustainability is also emphasized, on the newly imposed EN Standards on the sustainable assessment of buildings [4], where a combination of the assessments of environmental and economic performance taking into account the technical and functional requirements of a building is approached and on the next generation structural codes (fib Model Code 2010), where repair and maintenance of concrete structures, will be subjected to strict requirements both with regard to environmental, economical and service life constraints [5]. One of the most urgent challenges that construction industry has to face is the deterioration of reinforced concrete structures. Thus, the scientific community must promote industrial ecology (utilization of industrial by-products) and establish the principles of sustainable management in concrete production, in order to achieve a "green" mix design and a new rigorous approach towards construction of robust durable structures (for a given service life) with the minimum environmental burden.

Rice husk ash (RHA) is a promising SCM; it is actually the solid residue derived from controlled burning of rice husks. RHA has shown to contain significant amount of reactive silica that could contribute chemically to the development of pozzolanic reactions. In many cases – depending on the nature of husks and burning/cooling conditions – the total silica of RHA exceeds 90% with most of it being non-crystalline, thus reactive under alkaline conditions [6]. Pozzolanic activity of RHA depends on silica content, silica crystallization phase, and size and surface area of ash particles. Ideally, ash must contain limited amount of unburnt carbon. RHA that has amorphous silica content and large surface area can be produced by combustion of rice husk at controlled temperature [7] and these factors are mainly responsible for its high reactivity [8,9].

Despite the fact that a lot of work has been done on treated RHA, results on the direct use of untreated RHA are scarce in the literature and are usually limited to strength contribution and not durability aspects. Feng et al. [10], for example, examined the pozzolanic properties of both untreated and treated RHA but did not attempt to assess their behavior in terms of durability. Sensale [11] analyzed the effect of two types of RHA (amorphous and partially crystalline optimized by dry-milling) on durability of cementitious materials by proper testing. Other relevant studies [12,13] have also examined the influence of untreated RHA in certain durability properties; however, more investigation has to be conducted concerning its valorization in cement-based mortars and concrete systems and especially properties like resistivity and resistance to carbonation.

The study presented here focuses on the use of untreated RHA in mortar and provides an integrated approach of the three main mechanisms which control durability of reinforced concrete: capillary absorption, permeability and diffusion. Results of strength, absorption by capillarity, chloride ion penetration, accelerated carbonation and resistivity tests are discussed in order to identify the potential added value of untreated RHA as a supplementary cementing material.

2. Experimental part

2.1. Raw materials characteristics

Two Greek RHAs – designated hereafter as A and Θ – each from the two production plants of a producer (Greek rice industry) were used in their raw state. Their chemical analysis along with information on their particle size distribution and mineralogy are given in Table 1. A Cilas 1064 Laser granulometer was used for measuring the particle size distribution of the raw materials, while mineral phases were detected with the aid of a Siemens X-ray diffractometer (Cu Ka radiation, 40 kV, 30 mA, in a scanning range of 5–65° in 2θ scale) equipped with a Diffrac-At Database. Morphology of RHAs (Fig. 1) was evaluated using a Scanning Electron Microscope (SEM-EDS) JEOL JSM 35C microscope system NORAN, Voyager and semi-quantitative standardless analysis. A CEM I 42.5 R was used for preparing the mixtures and commercially available silica fume (Sikafume S 92 D) completes the list of the raw materials used in this study. Main physicochemical characteristics of the cement and silica fume (SF) are also given in Table 1.

No apparent differences can be seen in the chemical and mineralogical characteristics of the two RHAs. They primarily consist of SiO_2 most of which is amorphous, thus potentially reactive in alkaline environment. The predominant mineral phase is cristobalite, while some limited trydimite was also detected. Even though cristobalite is a relatively hard mineral (6–7 according to Mohs scale), the absence of even harder and abrasive quartz (7 in Mohs scale) is expected to have a lower effect on mill capacity reduction when RHA is to be co-ground with the rest of the cement ingredients. The only differences between the ashes are limited to the silica content -both total and reactive- as well as their particle size distribution. In general RHA A can be considered an ash of slightly higher quality due to higher reactive silica content and finer particle size.

In terms of morphology RHAs demonstrate many cavities of various sizes leading to an interconnected porous network. Both ashes exhibit a structure resembling rolling hills (the morphology of rice husk) while areas of fragmented morphology were also identified, especially in RHA Θ. This morphology is probably the result of the burning process of the original husk in which the initial coverings retain their shape and give a teeth-like outer surface. Surface hairs are also preserved resulting in long silica fibers while partial burning of carbon leads to large porosity [14].

2.2. Pozzolanic reactivity

Before inserting RHAs into mortars and pastes their potential pozzolanic activity was assessed using the Chapelle test [14]. In this accelerated test, a gram from each pozzolan is added into dilute slurry of calcium hydroxide (reagent grade) and is treated under hydrothermal conditions (100 °C) for 18 h. The solution is subsequently filtered and the remaining quantity of lime in suspension is determined by titration. Results are expressed in grams of lime reacted per gram of pozzolan tested.

2.3. Cement mortars and pastes

2.3.1. Workability and compressive strength

Mortar specimens were produced according to EN 196-1 as shown in Table 2. The mix design consists of a control mortar with no ash addition, one mortar with 10% SF and mortars containing 10% and 15% of RHA A and Θ respectively. All SCM additions replaced equal weight of cement. Before molding, all mortars were brought to similar workability with the use of a modified polycarboxylate-type superplasticizer (SP). Workability target was set at 210 ± 10 mm to resemble the rheology of the control mortar (CTL). It was measured following the procedure described in ASTM 1437 [15] and average flow diameters are also given in Table 2. Compressive strength development was monitored at 7, 28 and 90 days.

2.3.2. Efficiency factor

The concept of the efficiency factor (simpler k-value) has been introduced as a way to predict the effect of SCMs on the properties (e.g. compressive strength) of Portland cement systems that utilize them. In other words, the efficiency factor is defined as the part of the SCM, which can be considered as equivalent to Portland cement, having the same properties as the concrete without SCM (obviously $k = 1$ for Portland cement) [16]. In this work, the efficiency factors were determined in order to draw conclusions regarding the effectiveness of the RHA-cements. Moreover, the authors aimed at determining whether previously developed analytical expressions, correlating active silica of artificial SCMs with k-values, could be also applied in the case of untreated RHA-cements.

2.3.3. Non evaporable water content

For evaluating the hydration process, paste specimens with a 10% and 20% RHA addition were prepared following a similar procedure to mortar preparation. The higher replacement ratio of 20%, instead of 15%, was chosen for the hydration study in order to fully examine and comprehend the small differences appearing in the hydration products of the cement pastes. The specimens were cast in plastic vials after intensive shaking to remove any air content. 2, 7, 28 and 90 days after mixing hydration was stopped with the use of organic solvents and overnight drying in a low pressure chamber connected to a vacuum pump. Fragments from the core of each dried sample were taken and were further pulverized to assure that they all pass through the 125 μm sieve. The pulverized material was then kept sealed into plastic bags and stored in a dessicator until testing. For monitoring the hydration process, the non-evaporable water contents and gel/space ratios were determined. Comparisons were made in all cases with the control specimen (CTL). To determine the non-evaporable water content (W_n) of the hydrated samples, 1 g of the hydrated sample was first dried at 70 °C overnight (dried paste weight) and was afterwards ignited at 950 °C in an electric furnace for 1 h (ignited paste weight). Then the W_n