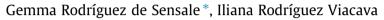
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A study on blended Portland cements containing residual rice husk ash and limestone filler



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HIGHLIGHTS

- Blends of Portland cement, residual rice husk ash and limestone filler are studied.
- Blended cements prepared using ternary blends satisfy standard requirements.
- Alkali-silica reaction studies in the ternary mixtures show an innocuous behavior.
- The ternary mixtures lead to a reduction in CO₂ emissions compared to OPC.
- Effects of microstructure (DRX and SEM) on BPC properties are studied.

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ABSTRACT

The cement industry is responsible for large CO₂ emissions. Residual rice husk ash (RRHA) can contribute both to a reduction of the environmental impact of cement manufacturing as well as to the agro-energy chain. The objective of this study is to obtain Blended Portland Cement (BPC) with up to 35% substitution of cement by RRHA and limestone filler. On the analysed mixtures, properties required by BPC standards, alkali-silica reaction, and effects of mineralogy and microstructure on BPC properties are discussed. The mixtures satisfy standard BPC requirements and lead to a reduction in CO₂ emissions when compared to Portland cement.

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1. Introduction

The cement industry is responsible for 5–7% of CO₂ emissions in the world [1–5]. The production of cements is based on the use of clinker. By using a kilogram less of clinker when making one ton of cement, CO₂ emissions could be reduced by 1 kg [6]. Taking into account environmental and economic factors, a reduction in carbon dioxide emissions is highly desirable in the cement industry.

For this reason, the replacement of clinker by other materials, especially if these are agro-industrial waste or by-products, is a highly attractive option to reduce the problem significantly, and, consequently, to promote energy savings and reduce the environmental impact [2,7,8].

During the rice production process, the husk is separated from the grain. Husk accounts by weight for about 20% [9]. The disposition of this agricultural residue is a problem [10–13].

* Corresponding author. *E-mail address:* gemma@fing.edu.uy (G. Rodríguez de Sensale). On the other hand, in the same way as other agro-industrial residues, rice husk (RH) offers great potential for its use as biomass [9]. The burning of the RH generates a new residue: the rice husk ash (RHA). Several investigators [9,10] argue that the burning of the rice husk can generate approximately 20% ash as a residue. If the entire RH was used as a source of biomass for energy generation, the volume of RHAs generated worldwide would be in the order of 19.26 million tons by 2016/2017. The RHA has low bulk density, very high percentage of silica, and its open field disposition in large volumes can create environmental problems [9,14,15]. Currently, RHA has no specific destination in many countries, being one of the most abundant residues of the agro-energy process taking place in the world.

Research and development of products using RHA has been carried out worldwide so as to give a destination to this residue, being [9,16] examples of recent publications. There are many investigations regarding the RHA pozzolanic activity can have as well as its use as a supplementary cementitious material in cement and concrete; different authors present the state of the art





[10,17–23]. In this regard, most of the published studies refer to the RHA influence on the mechanical and durability properties of different concretes [10,19,21,25–27].

The physical and chemical properties of RHA depend on the combustion method used for the burning of the RH [28]. When produced by controlled incineration, it is a highly active pozzolan, [24,29]; if it is incinerated in open field or from non-controlled combustion in industrial furnaces, usually contains a high proportion of non-reactive silica minerals, it is scarcely pozzolanic and is named residual rice-husk ash (RRHA). On the other hand, acid pretreatments studies have been carried out to rice husk to obtain amorphous rice husk ash [30–32], and more recently studies analyze the pozzolanicity of a combination of RHA and kaolinitic clay obtained in simultaneous calcination to obtain a high performance supplementary cementing material [33].

According to Mehta [34], satisfactory hydraulic cements can be made from rice husk ash (RHA) containing silica in a highly reactive form, obtained by patent [35,36], by simply blending previously ground ash, or by intergrinding the ash with calcareous material. For this reason, the focus of the present work is the study of Blended Portland Cement (BPC) employing RRHA. The use of these residues [37], for BPC has enormous potential in countries where there is great rice production, as it can contribute to the reduction of the environmental impact of the agro-energy chain, to the reduction of energy demand in the manufacture of Portland cement and its costs, as well as to the reduction of CO_2 emissions.

Residual rice-husk ash (RRHA), when properly crushed, can acquire pozzolanic characteristics [24,38,39]. Its reactivity depends on the BET specific surface and the average particle size independently of the amorphous content of the sample [40].

A technical feasibility of using one of the RRHA employed in the present work as a supplementary cementitious material in concrete has been reported in Ref. [18,20,41]. Other residual rice husk ashes were studied as supplement in Refs. [42–46]. Recent publications introduced the use of a ternary blend of Portland cement, RRHA and limestone powder to improve the properties of self-compacting concrete [47], and the use of a mixture of RRHA, limestone and wood fibers to improve the properties of lightweight concrete blocks [48].

There are multiple extensive studies on different properties of binary and ternary combinations of Portland Cement with RRHA discussed in the literature [8,22,41,49–55], but there is a lack of an overall study discussing all the standard properties required to produce Blended Portland Cements (BPC) with a single ash.

According to Nedhi et al. [56], since limestone is available in all cement plants, partial replacement of cement by combined pozzolanic industrial byproducts and ground limestone may provide a more efficient and ecologically friendly use of cement without compromising the fundamental characteristics of the cementitious binder.

In this paper, properties of Blended Portland Cement (BPC) employing RRHA and limestone filler were investigated. Limestone is used as filler to improve the workability of the BPC due to the high-water demand of the RRHA. RRHA from two industrial processes were used. Different percentages of cement substitution by RRHA and limestone filler were studied with regards to compliance of normative properties required for BPCs and their effect on the mineralogy and microstructure. The alkali-silica reaction (ASR) was also studied; the ASR is a potential problem when the RHA is used in large quantities, exceeding the amount that can react with the calcium hydroxide (CH) of the cement in the hydration. At industrial scale, the percentage of CO₂ emissions of the BPC production was estimated and contrasted with the emissions of the ordinary Portand cement (OPC) production. In addition, this work develops a model to predict the BPC compressive strength at 28 days from OPC compressive strength at 28 days and the percentages of RRHA and limestone filler to be used.

2. Materials and methods

2.1. Materials

The materials used in this study are:

- Ordinary Portland Cement, whose characteristics are listed in Table 1.
- Rice husk ash: to address the problem of the use of RRHA for cements, as they may come from different industrial processes, two residual ashes of different origin were used. One of them is a residue from the use of rice husk as fuel to generate steam in the process of parboiling of rice (RRHA1). The other one is a residue from the use of rice husk as biomass for electric power generation (RRHA2). To prepare them for their use in cement it was necessary to undergo simple drying operations (since both RRHAs had humidity greater than 3%), sieving (to eliminate impurities such as unburned rice husks, aggregates, etc.), and grinding (to acquire pozzolanicity according to [24,40]).

The ashes originally had humidities of 4% and 25%, respectively, so they were dried until constant weight was attained. As RRHA1 and RRHA2 contained unburned rice husk and fine aggregates respectively, both ashes were sieved with a 1.18 mm sieve (to remove these impurities).

Then, the optimum grinding time for the use of RRHA in cement was studied. Both RRHA samples were subjected to dry-milling at different times; the particle size distribution of the samples obtained were studied by laser light Coulter LS 230 equipment. Through milling process, the particle size decreased in grinding times from 5 to 30 min for both RRHAs with the type of mill used; when the grinding duration was over 30 min the mean particle size increase: this is due to the particle agglomeration associated with excessive grinding time [57,23]. Strength activity indexes with cement obtained at the age of 28 days [58], were performed at the different milling times, only with grinding time of 30 min were obtained results higher than 0.75 [59]. The mean particle size and the higher strength activity indexes with cement obtained with grinding times of 30 min were 7.53 µm and 0.90 for RRHA1, and 4.12 μm and 0.81 for RRHA2, respectively. The physical and chemical characteristics of both RRHAs are shown in Table 1. For the determination of the chemical components were used a Energy Dispersive X-ray Fluorescence (ED-XRF) traditional methods. Both RRHAs studied had the same content of silicon dioxide. A rapid analytical method to evaluated the content of amorphous silica in the rice husk ashes has been used [60]; the percentage of reactive silica contained in RRHA1 was 39.39% and in RRHA2 was 34.83%. In order to detect the crystalline phases present, the RRHAs were studied by X-ray diffraction (XRD), using the powder method with a RIGAKU-UltimaIV diffractometer. The X-ray diffraction diagram of each sample, is presented in Fig. 1. Crystalline peaks were identified in both ashes, which were identified as SiO₂ cristobalite in RRHA1 while in RRHA2 they are SiO₂ quartz and SiO₂ cristobalite. According to Mehta [36], it has been shown experimentally that attrition grinding of crystalline quartz can activate the silica by rupturing the chemical bonds at the surface. Fig. 2 shows the scanning electron microscopy (SEM) of both ashes and the employed limestone filler. The images (a) and (b) clearly show that more impurities are present on the surface of RRHA2 than in RRHA1. According to Soares et al. [52] when these impurities are present on the surface of silica they decrease it pozzolanic activity,

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