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Research article

Use of oxyfuel combustion ash for the production of blended cements: A synergetic solution toward reduction of CO_2 emissions

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ABSTRACT

In this paper, it is investigated the possibility of reusing ashes, issued by an oxyfuel combustion process aimed at mitigating CO_2 emission, as substitutes for natural pozzolan in the production of low-CO_2 blended cements. To this end, the oxyfuel plant (a 95 kW_{th} pilot-scale fluidized bed reactor) was operated under controlled conditions by feeding blends of anthracite or lignite and biomass corn stover. Characterization of fly and bottom ashes revealed that the latter showed properties able to make them considerable for obtaining blended cements by mixing them with Portland clinker and natural gypsum. The cements were subjected to pozzolanicity and hydration tests for curing times ranging from 2 to 28 d at 20° and 40 °C. X-ray fluorescence and diffraction, differential thermal–thermogravimetric analyses and scanning electron microscopy were employed as characterization techniques. With reference to a standard blended cement, and with particular eye on the blended cement containing bottom ashes obtained from the lignite–biomass mixture combustion, it was observed a good similarity in the ability of the silico-aluminous fraction to react with $Ca(OH)_2$ produced by Portland clinker hydration, to yield the desired calcium silicate hydrates among the hydration products.

1. Introduction

1.1. Oxyfuel combustion as a mean to reduce CO_2 emissions in the atmosphere

The economic growth during the industrial age has brought an exceptional wealth to the humankind, but it has also come at a cost, mainly associated with the current global warming (GW; list of abbreviations reported at the end of the manuscript) [1], representing the main environmental and economic menace in our time [2]. This phenomenon is caused by the emissions of greenhouse gases (GHGs) which are mainly released by the burning of fossil fuels, land clearing, agriculture-related and other human activities.

Among GHGs, carbon dioxide is the most important and abundant gas giving the largest contribution to GW (65%) [3]. Since the beginning of the industrial revolution (around year 1760), the mean concentration of CO_2 in the Earth's atmosphere has increased tremendously, growing from around 280 ppm (the same value as the one estimated around the year 1000 through recent analyses on Antarctic glaciers) to 402.25 ppm (August 2016) [4]. In particular, in the decade 2001–2011 the annual average increase in CO_2 emissions was about equal to ca. 4% per year; this value decreased to about 1% per year in the following two years and to about only 0.5% in 2014 when the global CO_2 emissions accounted for 35.7 Gt. Almost 61% of CO_2 emission is caused by industrial activities (electricity, heat generation and other industries) [5]. Therefore, searching for promising approaches to mitigate CO_2 emissions represents the priority of studies aimed at mitigating the threat of climate change.

Among the currently developing technologies having the potential to almost completely eliminate CO_2 emissions from power plants and other industries (including cement factories), 'carbon capture and storage [sequestration]' (CCS) processes are of particular relevance [6–9]. CCS indicates a group of technologies developed to obtain CO_2-rich flue gases ready to be stored by injection into geological strata with specific features. Among the different CCS processes, such as e.g. chemical absorption, chemical and calcium looping, oxyfuel combustion (OFC) is of great interest due to its conceptual simplicity [10]. In OFC, a blend of nearly pure oxygen and part of exhaust gas, as O_2 diluent for safety reasons, is used for combustion, thus lowering both nitrogen and NO_x contents in the exhaust gas [11–13]. In absence of the most relevant diluent for CO_2 (namely N_2) and after further treatments, it is possible to

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obtain streams >90% CO_2-rich. The CO2-rich stream is thus ready for final processing and geological storage.

1.2. Low-CO_2 blended cements

Cement production is one of the most raw materials - as well as energy-intensive manufacturing processes and represents one of the major industrial sectors giving rise to CO_2 emission. In fact, the contribution to the global anthropogenic CO_2 emission is estimated as high as 7% (namely, about 26% of the industrial CO_2 emission), deriving from a global cement production equal to about 4.5 Gt in 2015 [14,15]. Portland cement is the most widely used binder all over the world and is obtained by intergrinding Portland cement clinker (PCC) with a few percent of calcium sulfates (mostly gypsum). PCC is produced by heating and making it to react, in a rotary kiln at about 1450 °C, a mixture commonly composed by limestone (about 75%) and clay. For each kg of PCC produced, about 0.87 kg of CO_2 are released [16]; this comes from both limestone thermal decomposition (about 60% of the total CO_2 emission) and fuel combustion (mainly fossil and pet coke) [17]. Therefore, the main challenge of the cement industry is focused on the CO_2 emission reduction to 1.55 Gt per year (about −45% of the current value) by 2050 [16]. To this end, since the beginning of the third millennium, both cement producers and scientific community suggested several approaches [18–21], as: a) the use of non-traditional fuels; b) a higher utilization of low-CO_2 Portland cements; c) the development of alternative low-CO_2 binders obtained from non-Portland clinkers and d) the application of the CCS technology to cement factories.

In particular, low-CO_2 Portland cements, namely hydraulic binders whose production process is associated with a reduced CO_2 generation, can be obtained following two main different approaches [22,23]: 1) the use of a non-carbonated CaO source instead of limestone as a constituent of the Portland clinker-generating raw mix [24–28]; 2) the increased production of blended cements, obtained by mixing Portland clinker with significant amounts of supplementary cementitious materials based on silica and alumina (e.g. natural pozzolans, coal fly ashes, blast-furnace slags), to alleviate the environmental impact related to the clinker production [29–34].

In particular, coal fly ashes (CFA) are generated during the combustion of pulverized coal in electric power stations. The main components of CFA are silica, alumina, ferrous oxide and calcium oxide with varying amounts of unburnt carbon. Their chemistry, depending on the type of coal burned, has traditionally been the basis for assessing their

Table 1
Proximate and ultimate analyses for B, A and L, mass %.

	B-corn stover biomass	A-anthracite	L-lignite
Proximate analysis			
Moisture	6.18	2.42	13.57
Volatiles	70.68	6.71	25.72
Fixed carbon	17.64	59.57	30.41
Ash	5.50	31.30	30.30
Lower heating value, kJ kg^{-1}	15,438	21,620	14,434
Ultimate analysis			
C	43.3	59.27	40.53
H	5.82	2.01	3.18
N	0.57	0.91	0.28
S	0.11	1.58	6.65
Cl	0.35	–	–

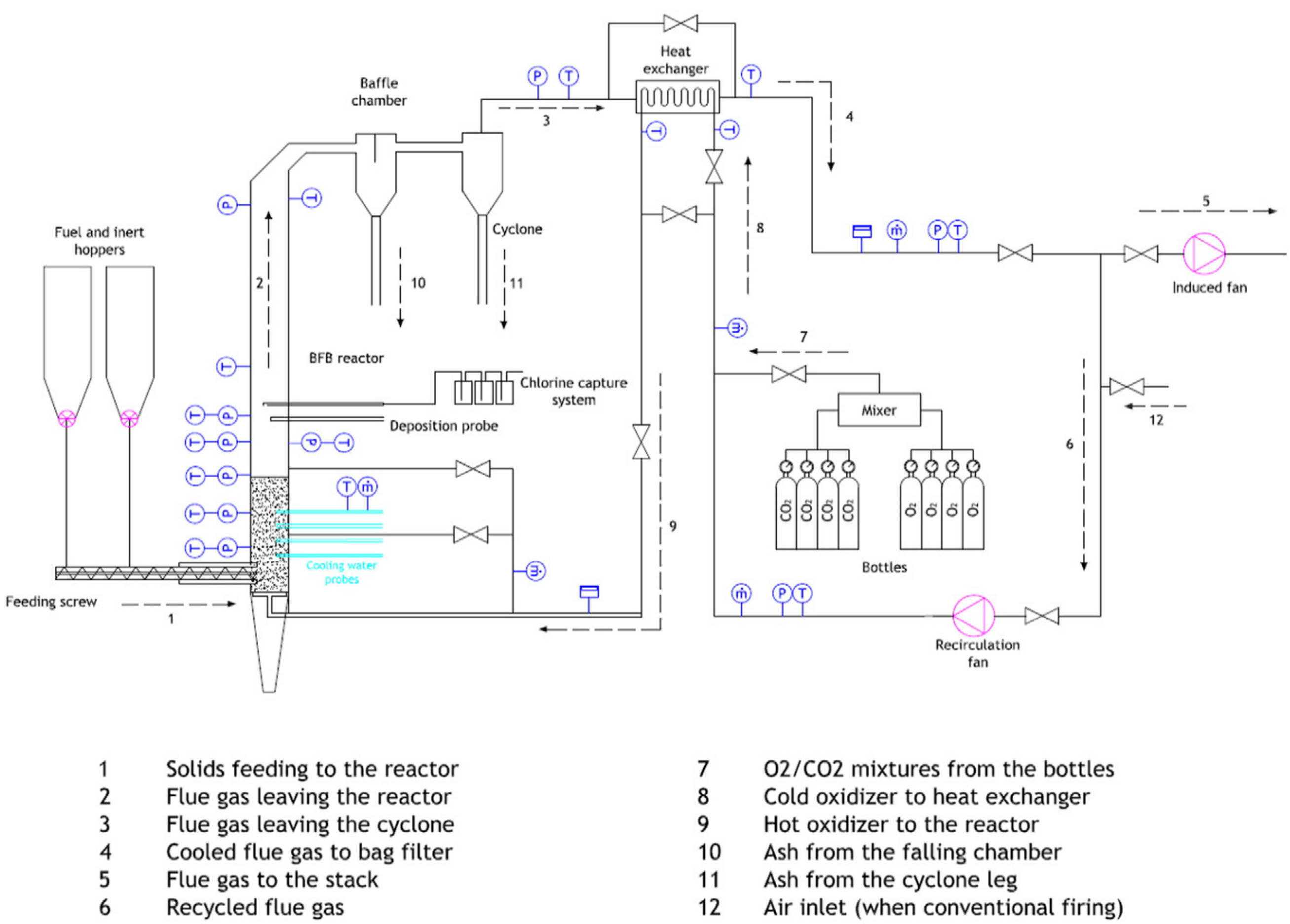

Fig. 1. Oxyfuel fluidized bed pilot plant installed at CIRCE, Spain.