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Beneficial use of boiler ash in alkali-activated bricks

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ABSTRACT

This research incorporates waste boiler ash into masonry construction materials using alkali-activation. The boiler ash, derived from three different Indian pulp and paper mills, has many undesirable characteristics for alkali-activation, including varying shape, large particle sizes ranging from 5 to 600 μm , loss on ignition between 8 and 35%, and less than 4% alumina. When combined with supplementary materials in the form of clay and lime, high compressive strengths are observed in the bricks made with all three ashes, demonstrating the robustness of the proposed mix design. A brick formulation with a solids phase weight ratio of ash(70):clay (20):lime(10), liquid to solid ratio of 0.45, and 2 M NaOH produces bricks with compressive strengths between 11 and 15 MPa after 28 days curing at 30 °C. Furthermore, early strength development is observed, as more than 55% of the 28 day strength is achieved after one day curing. An economic and environmental analysis indicates that these bricks can be produced for similar costs as the clay fired brick with reduced environmental impact, making them a viable alternative in the market.

1. Introduction

Significant rapid growth in both population and industrial activity in India has offered great development opportunity but has been accompanied by substantial environmental challenges. With this research we aim to address one challenge, namely the unprecedented demand for building materials stemming from population growth, by leveraging another, the production of vast quantities of non-hazardous waste from industrial activity.

On the population side, India is home to over 1.26 billion people, making it the second most populous nation in the world with studies predicting that India will surpass China for the largest population by 2030 (Chandramouli, 2001; James, 2011). This spike in population leads to an inevitable increase in the demand for buildings and infrastructure at an annual growth rate of 7% thereby increasing housing stock by five times the current quantity from 2005 to 2030, leading to extraordinary materials use (Maithel et al., 2012).

Historically, the fired clay brick is the most commonly used building material in India due to its low manufacturing cost and the availability of clay throughout the country (Maity, 2015). Despite the fired clay brick's long standing dominance as the building material of choice for housing, a number of environmental concerns surrounding its production have raised concern about future use (Maithel et al., 2012). The

manufacturing of fired clay bricks is an energy intensive process as the bricks are fired at temperatures over 1000 °C, making this industry the third largest consumer of coal in India as upwards of 150 billion bricks are produced annually (Maity, 2015). Furthermore, the frequent use of outdated kiln technology for firing leads to significant air pollution in the form of carbon dioxide, carbon monoxide, sulfur dioxide, nitrogen oxides, black carbon, and particulate matter (Maithel et al., 2012). Another serious environmental concern associated with the brick making industry is the degradation of topsoil extracted to make the bricks, which is quickly reducing the amount of irrigable land in India (Kathuria and Balasubramanian, 2014). The above mentioned problems, among others including extreme, labor-intensive working conditions, have prompted the search for alternative brick manufacture solutions, such as concrete or cement stabilized earth blocks.

In parallel with the growing population, there has been significant growth in industry within India. To date, this rapid industrialization has generated vast quantities of industrial wastes or byproducts. For instance, due to the lack of energy access in rural India, (Balachandra, 2011; Bhattacharyya, 2006; Srivastava et al., 2012) many small to medium sized factories produce their own energy by burning a variety of raw materials in industrial boilers, generating a byproduct called boiler ash. The raw materials these factories use fluctuate because their goal is to use the cheapest materials on the market. Some of the

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materials burned include petroleum coke, coal, and biomass/agricultural residues in the forms of rice husk, bagasse, and mustard straw (Cardoen et al., 2015) where materials may be burned individually or co-combusted (Kalemkiewicz and Chmielarz, 2012). The ongoing changes in the quantity and quality of raw material sources produce ash with high variability in its physical and chemical properties. Furthermore, the inefficiency of the boiler where the raw materials are combusted produces ash with a large amount of unburnt material. Also, due to small-scale local production of boiler ash (as compared with coal-derived fly ash production from large thermal power plants), there has been low interest among entrepreneurs to make use of it, despite its large collective impact. To date, these issues have been a barrier to finding a beneficial use for boiler ash (Pappu et al., 2007). Therefore, much of this boiler ash is being dumped into landfills or disposed of illegally, occupying valuable farmland and posing serious hazards to both the environment and human health. Furthermore, landfilling this ash comes at an expense to factory owners who need to purchase land, transport the ash to the site, and finally wet and level the ash.

One potential method of addressing both of the aforementioned problems (increase demand for housing materials and increased volume of industrial waste) is to use industrial ash byproducts as raw materials in bricks. Zhang defines three technological strategies for doing this: firing, cementing, and alkali-activation of waste (Zhang, 2013). Firing follows the same procedure necessary for the traditional fired clay brick, the only difference being a partial substitution of industrial waste for clay. However, studies show that as the waste substitution percentage increases, the strength of the brick decreases (Velasco et al., 2014). Moreover the bricks still need to be fired at high temperatures using traditional kiln technology, thus the energy consumption and air pollution are equal to that of traditional fired clay brick production (Zhang, 2013). The second strategy, cementing, avoids the use of a kiln for high firing temperatures (Abdalqader et al., 2016; Antunes Boca Santa et al., 2013). Still, with the use of cement in the mix design, the carbon footprint of these bricks is greatly increased as around 0.92 tons CO₂ are released for each ton of clinker produced (Komnitsas, 2011). Previous work has explored the use of coal bottom ash as a sand replacement in concrete where it does not have significant reactivity (Singh and Siddique, 2013). The third strategy, alkali-activation, depends on a chemical reaction between amorphous alumina and silica rich solids and an alkaline activator (Provis and Van Deventer, 2014). This strategy uses a low energy curing process allowing the bricks to gain strength at ambient temperature. Due to the increased interest in the field of alkali-activation, the research presented here focuses on this third strategy to make beneficial use of ash in bricks.

Alkali-activation has been the subject of intense study across a broad range of wastes including those with relatively controlled compositions such as slag, lime kiln dust and fly ash (Arulrajah et al., 2017a,b; Bernal, 2016). Broader wastes have been targeted such as those derived from the following four categories: 1) energy generation (i.e., coal and biomass-derived fly and bottom ash), 2) material and fuel production (i.e., mining (Ahmari and Zhang, 2012; Castro-Gomes et al., 2012; Jiao et al., 2013), metallurgical wastes such as red mud (Badanoiu et al., 2015; Dimas et al., 2009; He et al., 2013; Kumar and Kumar, 2013), and paper production wastes (Santa et al., 2013; Yan and Sagoe-Crentsil, 2012)), 3) waste treatment (i.e., incineration ash (Chen et al., 2016; Garcia-Lodeiro et al., 2016) and water treatment sludge (Geraldo et al., 2017; Guo et al., 2010)), as well as 4) end-of-life flows (i.e., glass waste (Novais et al., 2016; Wang et al., 2016) and ceramic waste (Reig et al., 2013; Sun et al., 2013)).

An issue hindering the implementation of alkali-activation is the inability to reliably predict the mechanical performance of the products formed based on the properties of the ash (Aughenbaugh et al., 2015; Provis et al., 2012). The reactivity of the ash will directly impact the final properties of the product formed (Diaz et al., 2010; Van Jaarsveld et al., 2003). The overall reactivity of the ash is dependent on a number of factors including the amount of reactive amorphous silica and

alumina, calcium and iron content, unburnt material, as well as particle size and morphology. Since boiler ash has yet to be used in significant quantities in the application of alkali-activated materials, the desired properties of the boiler ash will be compared with the characteristics of fly ash, as the latter has been successfully used in the production of alkali-activated materials (Shi et al., 2011). Characterizing the ash will determine its suitability for use in alkali-activated materials and identify solutions to improve its suitability if needed. Therefore, the contributions of this work are to characterize boiler ash from the pulp and paper industry, identify whether it can be incorporated in bricks through alkali-activation, and measure the properties of the resulting bricks in comparison with fired clay alternatives along dimensions of technical, economic and environmental performance.

2. Materials and methods

Alkali-activated bricks were made out of boiler ash, clay, hydrated lime, sodium hydroxide pellets, and municipal water. The boiler ash samples were obtained from three different paper mills in the city of Muzaffarnagar, India. The paper mills were Bindlas, Silverton, and Siddhbadi, and we identify the boiler ash throughout this manuscript by the name of the paper mill from which it was obtained. A map of the study region including the paper mills of interest is provided in the appendix. The percentages by weight of raw materials combusted to produce the Bindlas boiler ash was 63% bagasse pit, 27% rice husk, and 10% petroleum coke. The Silverton and Siddhbadi ash were byproducts of combustion of 100% rice husk. Each of these energy sources was chosen by the paper producers within each mill and the mix will vary seasonally depending on their availability. The clay was also obtained from a field nearby the paper mills in Muzaffarnagar. Both the boiler ash and clay were placed in an oven at 105 °C to remove any moisture from the specimens. The clay was then ground and sieved to pass the #35 sieve (0.5 mm) to break up any large agglomerates. Hydrated lime was a Graymont product obtained from Madigan Lime Corporation in Ayer, MA. The lime was also sieved to pass the #35 sieve to remove any large agglomerates. Laboratory grade > 97% sodium hydroxide pellets were acquired from Sigma Aldrich. Municipal water was used throughout to attempt to represent field conditions.

The products formed using alkali-activation technology are heavily dependent on the boiler ash properties. The chemical composition of the ash is related to the raw materials burned, while the mineralogical properties depend on the design and operation of the boiler (Williams and Van Riessen, 2010). Common tests which were used to characterize ash include X-ray fluorescence (XRF), X-ray diffraction (XRD), particle size distribution (PSD), and scanning electron microscopy (SEM). A semi quantitative elemental chemical analysis was obtained via XRF using a Bruker S4 Explorer. A Leco SC632 Carbon Analyzer was used to determine carbon content in the ash. XRD data was collected using high speed Bragg-Brentano optics on the PANalytical X'Pert Pro MPD. The crystalline peaks were identified using the ICDD PDF4+ database. Data was obtained between 15° and 70° (2 θ). Particle size measurements on the boiler ash were conducted using a Horiba LA920 laser scattering particle size distribution analyzer. SEM images were taken on a Philips XL30 FEG ESEM to observe the morphology of the boiler ash particles.

Tests were performed to determine the mechanical characteristics of the bricks made using alkali-activation. To prepare the samples, first the sodium hydroxide pellets were dissolved in water by stirring until a homogenous solution was formed. Due to the exothermic reaction, this was done a day in advance to allow the solution to cool down to room temperature. Next, the dry materials (boiler ash, clay, and hydrated lime) were weighed, added to a bowl, and mixed using a planetary mixer for 3 min to attain a homogenous composition of the solids phase. The NaOH solution was then weighed and added to the bowl. This was mixed with the solids phase at maximum speed until a homogenous wet consistency was formed. This typically took between 20 and 30 minutes to achieve. The mixture was then transferred into 2" cubic molds where

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