Research on opportunity of recycling coal ash in ash dumps for power generation

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Abstract: - Before United Nations Framework Convention on Climate Change Treaty, power generation was relying mainly on coal. As a result, large amounts of ash resulting from coal combustion are occupying permanently land throughout the world causing air and soil pollution issues. Such an example is Valea Ceplea (Romania) ash dump, which occupies approximately 50 hectares with a volume of 13 million cubic meters. The ash dump poses critical air pollution issues to the neighbouring populated areas. The ash in this particular dump comes from low quality lignite with high content of humidity and sterile. As a result, the ash contains a significant amount of unburned carbon, which makes it unsuitable for coal ash conventional recycling directions. The paper explores the possibility of using such ash in mixture with coal in conventional steam generation boilers, achieving a triple objective: I. Recycling ash, which becomes suitable for other recycling directions II. Saving coal, which results in significant economic savings related to coal mining, transport and processing and III. Closure of ash dumps, eliminating the environmental impact.

Key-Words: - Coal ash, Recycling, Unburned carbon.

1 Introduction

Although coal has been the main fuel for power generation and the key triggering factor of the Industrial Revolution, its usage produced environmental issues that only recently have become apparent. Coal usage for power generation has multiple negative effects on the environment with different irreversibility degrees. Climate change due to an increased carbon dioxide in the atmosphere is the most concerning because it is virtually irreversible and can have devastating long term consequences. Coal combustion by-products such as ash and slag are produced at huge rates, proportional to the coal combustion. Recently, several recycling directions for ash resulting from coal combustion have been identified. The range of such recycling directions is wide but it requires several fundamental conditions for the ash:

- The content of unburned matter is limited by the application
- Particle Distribution Size (PDS) uniform and adequate to the specific application

Such conditions are applicable in general to Fly Ash (FA) collected in dry state from the electrostatic precipitators. In contrast, Bottom Ash (BA) collected at the bottom of the boiler furnace contains variable amounts of unburned matter and a PDS spread over a large interval of values.

1.1 Literature review and state-of-the-art

Research on coal ash recycling technologies was stimulated by environmental regulations, which have become increasingly restrictive in the attempt to limit the environmental impact of coal combustion for power generation.

Effects of coal ash dump sites on the environment have been extensively investigated. Herrando-Pérez and Frid [1] conducted an analysis of the effects of macrobenthos and sediments caused by closure of an ash dump site. Quantitative sampling of the sediment and the macrobenthos was undertaken in the summer 1993 over 27 square km of seabed covering the Blyth Fly-ash Dumpsite and the surrounding area. It was found that 9 months after dumping cessation significant negative effects on the specific marine fauna were still present.

Closure and reclamation of ash dump sites is a long and expensive process, which cannot always guarantee full recovery of the environment factors.

Kostić et al [2] investigated the influence of weathering and vegetation development on changes in physical and chemical properties of FA, as well as environmental risks of FA at three deposit lagoons at the "Nikola Tesla-A" thermal power plant in Obrenovac, Serbia. It was found that weathering and revegetation has resulted in an overall improvement in the physical and chemical characteristics of raw fly ash through increasing the clay and silt fraction, decreasing alkalinity and salinity, and increasing the cation exchangeable capacity and N, P and K content, particularly in the surface layer. These changes have resulted in the creation of more favorable conditions for the further development of vegetation, as well as the accumulation and decomposition of organic material.

BA has a very limited range of recycling directions. The main reasons that limit the possibilities of recycling are the content of unburned (or partially burned) coal, content of slag (molten ash due to the high temperature in the furnace) and a non-uniform PDS.

The effect of unburned matter content in BA was investigated by Trifunovic et al [3], assessing the potential of recyclability of BA for road pavement materials. The study considered four samples with different contents of unburned carbon as follows: raw bottom ash, two size fractions obtained from it (2–5 and <2mm) and bottom ash treated by the "float–sink" method. The content of unburned carbon was determined by simultaneous DTA/TGA. Materials were then used in two types of mixtures, as follows: When these materials were used as a component in the mixture: fly ash–Portland cement and bottom ash–water for road construction, it was found that only mixtures containing bottom ash with lower carbon content (size fraction <2 mm and treated) presented mechanical properties suitable for the application.

Anghelescu et al [4] studied applicability of BA as a mixture component in various construction materials. It was found that unburned coal present in the BA limits significantly the mechanical performances of the final products.

Colonna et al [5] investigated the possibility of using BA for pavement binder course. Bottom ash may contain pyrites or lightweight, porous particles which are not an ideal aggregate source in bituminous mixes, especially for applications in which wearing is important. The presence of pyrites causes many issues to the hot mix asphalt when it is in contact with water and air. This is one of the reasons that made the use of bottom ash advisable only for intermediate courses. The test results indicate that there is no worsening in mechanical characteristics of the asphalt mix in comparison to the conventional reference mix. It means that BA could be added among the recoverable materials.

Recycling ash can be categorized in two different directions:

A. FA collected from the electrostatic precipitators is considered "clean" in the sense that it contains a low amount of organic matter in the form of unburned carbon. Recycling of such product is a mature technology. Construction materials industry if one of the main customers for fly ash. Singh et al [6] performed a comprehensive review of fly ash use in fabrication of self-compacting concrete. Rodriguez [7] studied the benefits and drawbacks of using fly ash as raw material for manufacturing Portland cement or PCC pavement. It was pointed out that more than 50% of the concrete produced in the U.S. contains fly ash. Xu and Shi [8] reviewed the applications of FA in various industries with a focus on sustainability and recycling.

B. Ash deposited in landfills has properties different from FA. It consists of a non-homogenous mixture of FA, BA and slag. The time spent in the landfill influences significantly the chemical and physical properties of the ash. Landfills pose special environmental problems and identifying landfill ash using directions is mainly a matter of landfill closure and not necessarily re-using the waste material. Xu et al. [9] carried out a chemical and geotechnical study on a FA landfill site in China. It was found that physical and chemical properties vary significantly across the landfill area. Significant modifications of the physical and chemical properties of the material occur over time. A large number of experimental studies exist on the issue of reclaiming ash landfills Cooke [10], Fine and Mingelgrin [11], Ayers [12], etc. There are also studies on integrating the bottom ash as a raw material for construction industry. Singh and Bhardwaj [13] reviewed the role of bottom ash as an alternative for cement. The study reviewed the chemical, microstructure, physical, mechanical, durability properties of concretes made with grinded BA. Some environmental concerns associated with using grinded BA deposited in landfills are discussed. Singh and Arya reviewed the properties of concrete containing various types of BA. Mechanical and durability properties of Normally Vibrated Concrete (NVC) and Self-Compacting Concrete (SCC) obtained with incorporation of BA as replacement of fine aggregates were investigated. Most of the studies considered show that the use of BA reduces the overall performance of NVC/SCC. However, several studies have reported its successful use in reaching similar or better performance compared to that of the concretes obtained with traditional aggregates.

Gentry [14] investigated economic and management issues related to ash handling and recycling. Several case studies and success stories were presented along with new promising directions for FA and BA recycling.

1.2 Ash dump site description

Valea Ceplea (Romania) ash dump is an ash collection facility that was in service for more than 25 years. The ash dump site covers a surface area of 52.64 hectares and a volume of approximately 13 mil m^3. The ash resulting from coal combustion for power generation at Turceni power plant 8x330 MW situated approximately 2 km from the ash, has been deposited by means of hydraulic conveying in the ash dump. FA and BA were mixed and transported in an aqueous solution with the ash/water ratio 10/1 (wt). This type of transport and deposit caused secondary environmental issues:

- Large volume of water required for transport
- Infiltration of contaminated water in the groundwater

The ash dump site was in operation before selective collection of ash has been implemented in order to apply recycling technologies. As of 2005, a FA collection installation has been implemented with the purpose of recycling FA as an additive for construction materials. A new process flow for collection and transport of the BA was also implemented in order to reduce the water consumption for transport. The continuous accumulation of ash in the Valea Ceplea ash dump extended its limits in the vicinity of inhabited areas. Serious environmental concerns and issues caused the closure of the ash dump. Even in its closed state, the ash dump poses serious environmental issues. Once the water used for transport has infiltrated or was lost through evaporation the ash dump surface becomes exposed to wind causing dust contamination of the inhabited areas during periods with certain wind intensity and directions.

Originally, the material deposited in the ash dump consists of mixture of approximately 80% FA and 20% BA. While FA is known to have a low content of unburned matter and a uniform PDS with the order of magnitude 10^{-5} – 10^{-4} m, BA has a PDS varying over several orders of magnitude and significant amounts of unburned matter in the form of coal particles.

Recycling of ash in Valea Ceplea ash dump is not possible in the traditional recycling directions for FA due to the following particularities:

- FA has been mixed with BA, the latest having characteristics described above
- Contamination over time with organic material

2 Methods

The ash in the Valea Ceplea ash dump has a number of special particularities, deriving from the combustion process and from the characteristics of coal. The fuel used at Turceni power plant is lignite from Oltenia open pit mine. Lignite has a low quality with calorific values ranging from 1200 to 1700 kcal/kg, high ash and humidity content. Another major issue of this type of coal is the impossibility to achieve full separation of coal from the sterile material before entering the fuel processing flow. The coal-fired boiler uses pulverized lignite, which is obtained through the following processing sequence:

- Crushing to the particle size 10-30 mm
- Milling combined with drying in coal mills to particle size 100-1000 mm

The low quality of the lignite and the presence of sterile material mixed with the coal do not ensure the desired milling size. As a result coal particle with large dimensions (2-10 mm) are injected in the furnace. This results in an incomplete combustion process, with larger coal particles being evacuated partially or not at all burned. Usually, the higher density of such particles compared to ash, makes them drop on the furnace bottom and are evacuated in mixture with BA. It has been noticed that the ash deposited in the ash dump contains variable amounts of unburned coal, which suggest a significant loss exists resulting in lowering the overall efficiency of the *power generation process.

This study will assess the technical feasibility of re-using the ash deposited in Valea Ceplea ash dump in mixture with coal for combustion in the coal-fired boiler to achieve the following objectives:

- Ecological closure of the Valea Ceplea ash dump site
- Replacing partially the lignite requirements for power generation
- The resulting ash with unburned carbon eliminated re-combustion will have characteristics suitable for recycling (absence of organic matter and uniform PDS)

2.1 Separation of ash particles by dimensions

The most important characteristic of a fuel in power generation industry is the calorific value. Therefore, before performing an elementary analysis of the ash to detect combustible elements, it was considered critical to estimate the combustion characteristics.

In order to ensure properties uniformity and relevance a number of 20 ash samples were collected from various areas of the ash dump. Each sample consisted of 2 kg of raw ash. Samples were mechanically mixed and a final sample was collected for further analysis. Visual inspection of the sample suggests a significant humidity and heterogeneous distribution of particle size.

The sample was subject to drying to remove the humidity by placing it in a drying chamber at 105 °C for 24 h. The appearance of the sample is presented in Figure 1. Drying was followed by particle size analysis, which was performed by means of mechanical sieving. A Retsch AS200 vibratory sieving machine using a throwing motion with angular momentum was employed (Figure 2).

A number of nine sieves were used, as follows: 4 mm and higher, 2-4mm, 1-2 mm, 0.500-1 mm, 0.250-0.500 mm, 0.125-0.250 mm, 0.090-0.125 mm, 0.063-0.090 mm and 0.063 mm and less.

Fig. 1 Physical appearance of the raw ash sample after humidity removal

Fig. 2 Sieving machine used for separation of particles by size

Particle size separation was necessary in order to determine if combustible particles concentrate in one or several particle size ranges or are uniformly distributed over the whole particle size spectrum. PDS is presented in Figure 3 and cumulative PDS is presented in Figure 4.

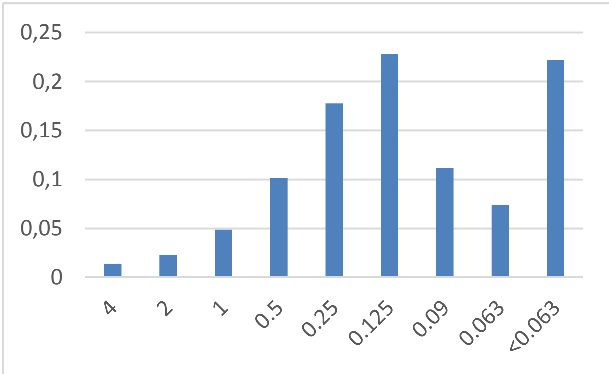

Fig. 3 Raw ash – PDS

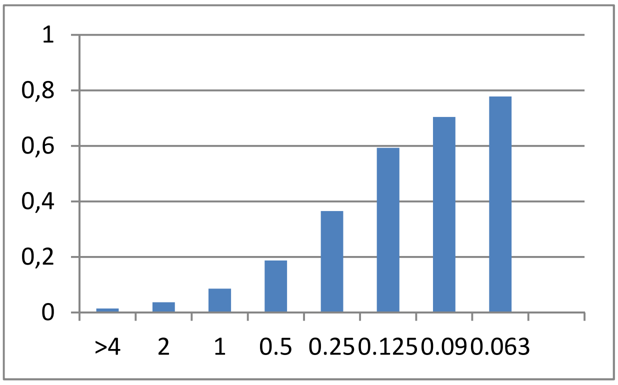

Fig. 4 Raw ash – cumulative PDS function

Two key observations can be extracted from analysing Figures 3 and 4:

I. Almost 50% of ash particles consists of particles from 90 mm to 250 mm

II. More than 20% of the ash particles are less than 63 mm in size.

The two observations above suggest that FA is the dominant component of the raw ash, considering the specific size of FA particles. However, almost 40% of the raw ash consists of particles with size 250 mm or higher, which could be an indication that significant amounts of unburned carbon is captured in this particle size range.

2.2 Determination of calorific value

Calorific value determination was carried out by means of an E2K Oxygen Bomb Calorimetric System. (Figure 5). Since it is not expected that FA component would contain unburned matter, the calorific value determination was carried out for each particle size interval in order to identify the particle size interval that concentrates the highest amount of unburned matter.

Ash samples were weighed using a high accuracy scale (10^{-4} g) and placed in the calorimetric bomb crucible (Figure 6). Determination of the calorific value for each particle size interval was carried out for 5 samples. An amount of approximately 1 g of ash was placed in the metallic crucible and the calorimetric vessel was filled with pure oxygen at 30 bar. All ash samples with particle dimensions above 0.125 mm were subject to milling in a mechanical vibrating mill in order to ensure particles with dimensions small enough for combustion process to occur.

Fig. 5 Oxygen Bomb Calorimetric System and pressurized vessel

Fig. 6 Scale used for weighing the ash samples and calorimetric bomb vessel

Determination of the calorific value was carried out without spiking material in order to assess the capacity of the ash sample to undergo combustion in the absence of a highly flammable material. It was found that only 3 particle size intervals are capable of combustion in the absence of spiking material: 2-4mm, 1-2 mm, 0.500-1 mm.

3 Results and discussion

Calorific values for the nine particle size intervals are presented in Figure 7.

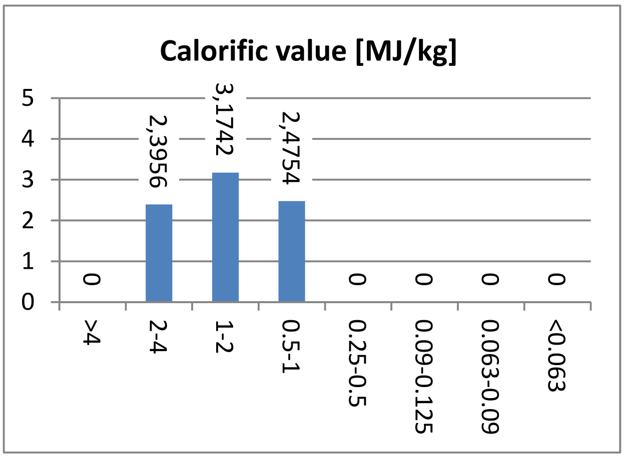

Fig. 7 Calorific values determined for each particle size interval

For all other intervals the calorimetric bomb instrument did not detect combustion (the instrument returned *misfire* error). Visual inspection of the samples revealed no detectable modifications that would indicate occurrence of combustion. In order to confirm non-flammability of the samples under calorimetric bomb conditions (highly oxygen saturated atmosphere, 30 bar) a single test was performed for each sample using spiking material.

Usage of spiking material is recommended for samples with very low calorific value or materials that burn reluctantly. As spiking material, benzoic acid C_6H_5COOH tablets were used with standard calorific value 26.44 MJ/kg. Standard benzoic acid tablets delivered with the calorimetric bomb were used. The machine returned calorific value 0 (misfire) confirming that ash samples with particle size other than 2-4mm, 1-2 mm, 0.500-1 mm are not capable of combustion, even under calorimetric bomb conditions with 30 bar and highly oxygen saturated atmosphere.

The appearance of one ash sample after firing in the calorimetric bomb with spiking material is presented in Figure 8. In the upper image the sample is presented after removal from the calorimetric bomb crucible. Two visually distinct, concentric zones can be identified, with the central zone presenting characteristics specific to non-flammable material exposed to high temperatures. The outer zone does not exhibit any modification specific to high-temperature exposure or combustion. In the lower image the sample was removed from the crucible in order to detect physical modifications underneath the surface.

It can be noticed that a slight sintering occurred around the zone where the spiking material was placed. This suggests that the high temperature resulting from the combustion of the spiking material caused melting in the sample immediate contact region, sintering in the vicinity of the contact zone and no other effect in the rest of the sample, which maintained its original appearance of a powder material. It is obvious that the burning of the benzoic acid tablet did not initiate combustion of the ash sample.

It can be concluded that ash samples with particle size other than 2-4 mm, 1-2 mm, 0.500-1 mm are not capable of combustion. Particles larger than 4 mm are expected to occur due to ash contamination over time, since the ash dump is almost 40 years old. A significant percentage of the particles (~22%) fall below 0.063 mm. These dimensions are typical to FA and correspond to coal particles that underwent complete combustion; therefore, they do not contain organic matter and have no flammability properties, or the content of organic matter does not ensure flammability properties.

Contamination in the landfill due to exposure to environmental factors and vegetation is expected to contribute to the content of organic matter, however it does not explain flammability properties, which can only be attributed to unburned coal particles.

Fig. 8 Appearance of the ash sample after being subject to combustion with spiking material

4 Conclusions

Ash deposited in a historic ash dump site Valea Ceplea (Romania) was characterized in terms of combustible matter content in order to study the technical feasibility of re-burning it in mixture with coal. It was found that organic matter in the form of unburned coal is concentrated in particles with dimensions 0.5 – 4 mm. This size interval corresponds to approximately 20% in terms of mass, the rest consisting of particles with dimensions below 0.25 mm and a negligible percentage, particles with dimensions higher than 4 mm. Ash particles with dimensions 0.25-4 mm are capable of combustion and their calorific value ranging from 2.4 to 3.1 MJ/kg is approximately half or less of the lignite used in the boiler (ranging from 5.0 to 7.1 MJ/kg).

Although such calorific value is relatively low, in mixture with lignite it can burn properly eliminating completely the unburned carbon. With actual ash separation and recycling technologies, it would be possible to obtain ash with proper characteristics for recycling. The process flow requires the following phases:

I. Removal of humidity from the raw ash
II. Separation of ash particles by size
III. Transport of the fraction 0.5-4 mm to the power plant and mixing with lignite on the process flow

Phases I and III require a significant energy consumption, which is expected to reduce the overall efficiency of the process. A secondary positive effect results from separation of particles is that the remaining fractions (i.e. fractions that do not contain combustible material) can be used in other recycling directions (additive for construction material, amend the soil pH in agriculture, etc.).

Although no major economic benefit can be expected from ash re-burning, it can be the only feasible way to close the ash dump site and reclaim the area currently occupied. If environmental regulations will be enforced to close and reclaim such ash dumps, separation followed by re-burning of the fraction containing unburned carbon can be the only viable solution. For existing installations where no recycling technologies for ash were implemented it is recommended to separate FA from BA and deposit the products in different ash dumps.

The presence of flammable components in the ash deposited in the landfill, even after prolonged exposure to environmental factors, is an indication of the low efficiency of the combustion process. As a secondary conclusion that can be derived from this study, measures to increase the efficiency of the coal combustion must be examined.

The multi-objective recycling process presented has the potential to achieve feasibility conditions given the triple benefit it can bring.

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