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Use of Geopolymerized Fly Ash with GGBS as a Barrier for Waste Containment Facilities

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ABSTRACT

The present paper reports the results of experimental investigations performed to examine the feasibility of using fly ash (FA) and ground-granulated blast furnace slag (GGBS) geopolymers as barrier materials for waste containment facilities. The alkaline geopolymer is a blend of FA and GGBS with sodium hydroxide in concentrations varying from 1 to 5. The important properties of most barrier materials include strength and hydraulic conductivity. While FA can develop compressive strength through pozzolanic reactions, polymerized FA develops tensile strength. For the construction of barriers for landfills with higher heights, tensile strength assumes importance. To further improve the strength, FA can be amended with GGBS. Results indicate that the FA-GGBS mixture in the ratio of 40:60, when cured, exhibited higher strength at any molar concentration. Further, the hydraulic conductivity of the material, which is predominant for barriers in waste containment facilities, is studied. To examine the impact of the presence of heavy metals in the leachates, batch adsorption studies were executed on a 40% FA- 60% GGBS mixture. Leachate with nickel and lead were adapted for their retention within the barrier. It has been observed that the geopolymerized FA and GGBS can retain ionic metals. The retention capacity of heavy metals is due to their precipitation in the voids of the barrier material enabling further reduction in the hydraulic conductivity making geopolymer a sustainable barrier material.

INTRODUCTION

Landfilling is the widely followed approach for solid waste disposal. Leachate generated from these landfills is becoming a major threat to the surrounding environment (Ganjian et al. 2004a). To protect the adjacent sites from groundwater contamination, landfill liners were proposed in the past. Many conventional barrier materials used for facilities releasing leachates with ionic contaminants, such as heavy metals, increase their hydraulic conductivity.

Strength characteristics and the retention capacity of contaminants are the two primary parameters to be analyzed in a landfill design (Deka & Sreedeep 2016). Barriers play a very important role in the operation of any landfill.

Generally, attention is paid to either reducing the permeability of the liner or immobilizing contaminants, but the strength properties have not yet been examined (Shankar & Phanikumar 2012). In landfills with greater heights, a higher settlement and slope failure are expected (Sheng et al. 2021). Engineered landfills are designed aiming for enhanced strength and better performance by retaining contaminants in the liner system (Herrmann et al. 2010). In recent years, research has especially centered on the utilization of industrial by-products in waste containment facilities (Devarangadi & Shankar 2020).

Conventional mortar with polymerized FA with alkali is used to partly replace different amounts of sand in mortar and is extensively studied to minimize the environmental effects of disposal FA besides the environmental issue of extensive mining of river sand. Industrial by-products such as FA and GGBS, when added together as a mixture, resulting in good strength due to hydration, forming cementitious compounds (Yawale & Patankar 2023). Polymerization of these components by alkali activation can further enhance the bonding effect of these materials (Wattez et al. 2021). Unlike Portland cement, where water is formed by hydration, geopolymerization eliminates water (Wilińska & Pacewska 2018). A further advantage is that this polymerization occurs at room temperature. Geo-polymerization does not form calcium silicate hydrates like those present in cement but develops strength by polycondensation of alumina and silica precursors at high concentrations of alkali (Qaidi et al. 2022).

Apart from strength, permeability is the major factor that affects the long-term performance of a liner (Ganjian et al. 2004). Landfill liners should be less permeable and are essential in waste disposal systems to control the flow of leachate and the migration of contaminants into the surrounding environment (Fall et al. 2010). In general, a clay liner following compaction should possess a permeability of 1×10^{-9} m/s or less (Cossu & Garbo 2018).

The primary aim of this study is to generate a barrier system with good strength in both compression and tension by using polymerized FA with GGBS mixtures, besides satisfying the liner requirements for hydraulic conductivity.

MATERIALS AND METHODS

Materials

The present investigation utilizes FA and GGBS as source materials, with sodium hydroxide (NaOH) pellets as an alkali activator. The FA utilized in the present investigation is procured from the Raichur thermal power plant in Karnataka, India. The percentage of lime (CaO) in FA used in this research is less than 2 and is categorized as Class F (Mishra & Ravindra 2015). The physical properties of the FA and GGBS used are provided in Table 1.

GGBS and sodium hydroxide used in the present research are procured from local distributors in Bangalore, Karnataka, India. The chemical compositions of FA and GGBS are outlined in Table 2.

It is seen from grain size analysis that the particles of FA are within the limits of fine sand and silt, while almost all the particles of GGBS are silt-sized.

Surface texture plays a crucial part in achieving density even with the same compactive effort (Hu et al. 2018). Hence, the microstructural properties were assessed by scanning electron microscopy (SEM) (Mukri et al. 2018). Table 1: Physical properties of FA and GGBS.

| Properties | FA | GGBS |
|---|--------|---------|
| Color | Grey | Whitish |
| Specific Gravity | 2.14 | 2.80 |
| Specific surface area(m ² /kg) | 339.00 | 480.00 |
| Liquid Limit (%) | 27.00 | 30.00 |
| Sand size (%) (4.75 - 0.075 mm) | 58.55 | 25.51 |
| Silt size (%) (0.075 - 0.002mm) | 39.82 | 70.49 |
| Clay size (%) (<0.002mm) | 0.63 | 4.00 |
| Optimum Moisture Content (%) | 15.60 | 22.70 |
| Maximum Dry Density (kN/m ³) | 14.00 | 15.90 |

Table 2: Chemical composition of FA and GGBS.

| Constituent | Proportion by Weight (%) | | |
|--------------------------------|--------------------------|-------|--|
| | FA | GGBS | |
| Al ₂ O ₃ | 39.64 | 20.85 | |
| SiO_2 | 50.44 | 32.81 | |
| Fe ₂ O ₃ | 4.18 | 0.36 | |
| CaO | 1.26 | 35.51 | |

Fig. 1 illustrates the microscopic images of FA and GGBS. From the SEM image, it is observed that FA particles are hollow cenospheres, whereas GGBS particles are angular with clear edges.

Experimental Program

The specified work investigates the impacts of FA-GGBSbased geopolymers concerning the geotechnical properties of landfill liners. The experimental program is categorized as material collection, casting, and curing, followed by laboratory testing and data evaluation. A set of trials was performed on FA-GGBS mixtures to figure out the optimum FA-GGBS ratio and the required amount of alkali for effective liner requirements. The paper discusses mixtures with 20%, 40%, 60%, and 80% FA blended with GGBS after activating with alkali for synthesizing into geopolymers. The alkali concentration is varied from 1 to 5 Molar (M).

The compaction characteristics of various FA-GGBS mixtures are determined using the minicompaction test apparatus (Sharma & Sivapullaiah 2016).

Specimens with standard size (3.8 cm \times 7.6 cm) were cast with respect to their maximum dry density (MDD) and optimum moisture content (OMC) for unconfined compressive strength (UCS) in a manner that the same impact energy is maintained for every sample (Palmer 2000). The mixtures were prepared on a dry-weight basis by adding the requisite quantity of water. Then the extruded samples were sealed in airtight bags, and the effect of time was



(a) Fly Ash



(b) GGBS

Fig. 1: SEM pictures of FA and GGBS.

investigated by curing the samples for 3, 14, and 28 days in a desiccator while maintaining 100% relative humidity at room temperature. The influence of alkali on FA-GGBS mixtures has been examined using unconfined compressive strength (UCS) as per IS 2720 (Part 10) standards.

Specimens with 100 mm in diameter and 200 mm in height were cast under molar concentrations of 1 to 5 to determine the tensile strength. The stiffness or tensile strength is studied by testing the specimens using a digital compression testing machine as per IS 5816 (1999) standards (Bellum et al. 2019).

Cubic specimens of 100 mm in size are cast and cured for 28 days to determine the permeability. The casted specimens were permeated with water and tested in terms of penetration depth as per IS 516-Part 2-Sec1 (Ibrahim & Issa 2016).

Batch equilibrium tests have been undertaken on a polymerized 40% FA and 60% GGBS mixture. 50 ppm each of lead (Pb) and nickel (Ni) solutions were prepared and used as leachate in the present study. 10 grams of adsorbent collected from the tested UCS samples were added to 100 mL of heavy metal solution and kept in a rotary shaker, maintaining a contact time of 8 hours. Later, the samples were filtered and tested for the presence of metal contaminants through atomic absorption spectroscopy.

A representative sample from the tested UCS specimens was collected and the morphology of the compounds was analyzed through microstructural studies.

RESULTS AND DISCUSSION

Compaction Characteristics

In this section, the compaction characteristics of FA blended with various percentages of GGBS were determined. The compaction curves are as shown in Fig. 2 and the optimum moisture content (OMC) and maximum dry density (MDD) are noted and presented in Table 3.

It is noticed from Fig. 2 that a rise in the percentage of GGBS, and MDD has increased whereas OMC is not following any particular trend. The enhancement in MDD is owing to the particle size, as it is seen from grain size analysis that GGBS is finer with more silt-sized particles. These finer particles fill the spaces left in the FA particles, following a dense mix, thereby increasing MDD.

UCS of FA-GGBS Mixtures Cured for Various Periods

UCS of FA and GGBS mixtures after various curing periods are presented in Fig. 3. Blending FA with GGBS will twin effect the strength both by improving the ratios of reactive silica and lime for the production of cementitious composites and by improving the silica-alumina ratio for effective polymerization with alkali (Sasui et al. 2019).

It is observed that the strength of FA-GGBS mixtures increases with the curing period. Cementitious compounds such as calcium silicate hydrate (CSH) formed are noticed over curing because of the pozzolanic reactions that are time-dependent.

| Table 3: Compaction | characteristics | of FA-GGBS | mixtures |
|---------------------|-----------------|------------|----------|
|---------------------|-----------------|------------|----------|

| GGBS - FA (%) | OMC (%) | MDD (kN/m ³) |
|---------------|---------|--------------------------|
| 0:100 | 15.6 | 14.0 |
| 20:80 | 18.1 | 14.4 |
| 40:60 | 20.5 | 14.7 |
| 60:40 | 20.3 | 15.2 |
| 80:20 | 20.5 | 15.5 |
| 100:0 | 22.7 | 15.9 |



Fig. 2: Compaction curves of various FA-GGBS mixes.



Fig. 3: Variation in UCS on FA and GGBS mixtures with curing period.

The escalation in strength of these mixtures is almost linear for FA with GGBS up to 60%. But for FA with 80% GGBS, the strength boosts steeply between 7 to 14 days and remains constant beyond 14 days. For GGBS alone, a marginal strength is noticed with curing. The increase in strength is because of the reactive silica and lime in FA/ GGBS that undergoes pozzolanic reactions. It was noticed that FA with 60% GGBS showed the highest strength, which means the mixture of 40% FA and 60% GGBS has enough lime and silica essential to promote the pozzolanic reaction (Singh 2018). This may be attributed to the optimum ratio of lime and reactive silica in the combined mixture of FA and GGBS. For mixtures with higher GGBS, the rise in strength with curing is less, implying that the pozzolanic reactions are not proceeding owing to one of the constituents, i.e., reactive silica or lime, which is not in the right proportion. This justifies using mixtures rather than either of them.

The reaction of sodium hydroxide on FA-GGBS mixtures followed by a 28-day curing period is shown in Fig. 4. Pozzolanic reactions proceed finer in the existence of alkali due in account to better solubility of silica from FA and/or GGBS. Apart from better pozzolanic activity, sodium hydroxide is an alkaline activator that helps to form inorganic polymers which increase in association with the alkali concentration.

Fig. 4 portrays that at a subsequent curing of 28 days; a clear trend is evident in all FA and GGBS mixtures and at all alkali concentrations. A gradual increase in UCS is observed for the mixture with 60% GGBS. Beyond 60%



Fig. 4: Effect of alkali concentration on UCS of FA and GGBS mixtures after 28 days of curing.

GGBS, UCS is reduced with any concentration of alkali. The maximum strength is obtained at about 60% of GGBS at 28 days of curing at any concentration of alkali, but the optimum alkali concentration can be considered as 3M. The reduction in strength at a higher GGBS percent is larger for mixtures with increased concentrations.

Blending FA with GGBS can enhance the properties required to use them as a component of a landfill liner. Enhanced production of cementitious compounds by reactions amidst lime and reactive silica available in them, apart from their production by hydration reactions of GGBS and FA, is noticed. The number of cementitious compounds formed can increase with alkali concentration, and the participation of sodium in the formation of these compounds enhances the hydration reaction by forming cementitious compounds such as CSH gels over an increased curing time (Thakur et al. 2022). The extent of these compounds formed can be advantageous when lime and reactive silica are together in the required proportion, and this can be evaluated by calculating the strength of various FA and GGBS combinations cured for various ages and with different concentrations of alkali. The mechanism becomes clearer if the variation in tensile strength influenced by these parameters is compared.

Tensile Strength of FA-GGBS Mixture

The optimized FA-GGBS mixture, which gave the highest compressive strength, is considered for tensile strength analysis. The tensile strength of a mixture of 40% FA and 60% GGBS with respect to alkali concentration at 28 days of curing is determined. Polymerization of silica in FA with alkali can be more advantageous in the development of tensile strength. Fig. 5 shows that the tensile strength progressively increases with an increase in the alkali



Fig. 5: Variation in tensile strength of 60% GGBS 40% FA mixture after 28 days of curing with alkali concentration.



Fig. 6: Effect of permeability for optimized FA-GGBS mixture cured for 28 days.

concentration over a curing period of 28 days. This may be because of the polymerization of FA and GGBS with sodium-based alkaline activators.

In general, for cementitious materials, when the compressive strength increases, the tensile strength fails to increase proportionally, or rather, it may decrease as well. It is good to see that both compressive strength and tensile strength are increasing. The steep increase in tensile strength compared to UCS confirms the polymerization of FA and/ or GGBS. Unlike cementation by pozzolanic compounds, which substantially enhance the unconfined compressive strength (UCS), polymerization can increase tensile strength.

A closure examination of Fig. 4 and 5 reveals that the percent increase in tensile strength is higher than the corresponding increase in compressive strength at any curing period.

Permeability of FA-GGBS Mixture with Varying Alkali Concentration

Fig. 6 shows the permeability of an optimized mix of 40% FA and 60% GGBS with varying concentrations of alkali from 1 to 5 molar cured for 28 days. It is observed that a rise in the alkali concentration reduces the permeability. Geopolymerization of reactive silica obtainable in fly ash in the presence of alkali, which confirms a significant lowering of permeability with alkali concentration, can be a reason. However, the role of more cementitious compounds in the reaction of reactive silica with lime over-curing cannot be ignored.

Batch Adsorption Studies

Batch adsorption tests were performed using an optimized FA-GGBS mixture as an adsorbent to a synthetic solution prepared in a laboratory at a known initial concentration of 100 ppm. Lead (Pb) and nickel (Ni) were the selected heavy

metals for these studies. 10 grams of adsorbent are added to 100 ml of synthetic solution and kept in a rotary shaker, maintaining a contact time of 8 hours. Later, the collected solutions were filtered and tested for heavy metals through atomic absorption spectroscopy (AAS), as shown in Table 4.

Batch experiments were performed to evaluate the sorption capacity and retention of selected heavy metals (Pb and Ni). The concentration of Ni was below the detectable limit (BDL) at all alkali concentrations of 1-4 molar except 5 molars, whereas for Pb, a gradual abatement of the residual ions is observed, and at 5 molars the existence of Pb ions is below the detectable level. The solubility products of nickel and lead hydroxides are in the range of 10⁻¹³ and 10⁻²⁰ mol/L (Scholz & Kahlert 2015). Thus, nickel is more strongly precipitated and retained at any given concentration. Theoretically, the retention can increase with a rise in the concentration of alkali. However, in the range of concentrations of 1 to 5 M alkali solution used, the differences can be very small. Lead precipitation shows some sensitivity to the alkali concentration and decreases over an increase in the concentration of alkali and can become completely precipitated at 5 molar alkali concentration. The precipitated hydroxide can redissolve if the alkali concentration is greatly increased. Thus, Ni precipitation decreases at higher concentrations of 5M alkali, and a small amount appears in soluble form. Despite all, both metals are retained to a great extent. This confirms the retention of Pb

Table 4: Heavy metal analysis.

| Concentration of alkali | Nickle (Ni), mg/L | Lead (Pb), mg/L |
|-------------------------|-------------------|-----------------|
| 1M | BDL | 1.11 |
| 2M | BDL | 0.60 |
| 3M | BDL | 0.42 |
| 4M | BDL | 0.34 |
| 5M | 0.70 | BDL |



(a) 1 Molar



(b) 2 Molar





(c) 3 Molar







Fig. 7: SEM images of 60% GGBS and 40% FA mixture with different alkali concentrations after curing for 28 days.

and Ni in the developed barrier material, predominantly by precipitation.

Micro Structural Investigations

An attempt is made to support the above explanations for variations in UCS and tensile strength with varying GGBS content and alkali concentration. An observation on SEM images of various molars at 28 days of curing on the optimized mix shows that at low concentrations of alkali, the gel formation of FA-GGBS mixtures is less, and spherical and clear-edge shaped particles are observed, whereas, at higher molarity, major percent of FA and GGBS particles react and result in gel formation. The new mineralogical formation is maximum for 60% GGBS and 40% FA

at 28 days of curing with higher molarity, as exhibited in Fig. 7.

CONCLUSIONS

Based on the detailed experimental studies conducted on polymerized FA-GGBS with alkali and curing for 28 days for its possible application in the construction of barrier systems in waste containment systems, the following findings are drawn:

- The maximum dry density (MDD) of FA increased with the incorporation of GGBS but the optimum moisture content (OMC) decreased. Generally, it is presumed that higher MDD is preferred, as it generally indicates higher strength and lower settlements.
- Curing the FA-GGBS mixture increases its strength with the length of the curing time. Among all the mixtures, the mixture with 60% GGBS and 40% FA showed the highest strength after curing for 28 days. This has been attributed to the optimal utilization of reactive silica and lime present in FA-GGBS mixtures.
- While the concentration of alkali increases, the UCS increases for all mixtures, which is also the maximum for FA with 60% GGBS.
- The tensile strength of the mixture with 60% GGBS and 40% FA increased by alkali activation using sodium hydroxide due to the polymerization of FA in the existence of alkali.
- The permeability of the barrier material is much lower than the minimum permeability recommended for liners of waste disposal facilities. Permeability declines when the concentration of alkali enhances and the reason behind this is the effective formation of geopolymers.
- Relatively, the rise in tensile of the FA-GGBS mixture is greater than in UCS, this trend becomes particularly vivid at higher concentrations of alkali.
- A substantial decrease in the hydraulic conductivity confirms the predominant effect of polymerization in FA-GGBS mixtures with alkali solutions.
- The formation of cementitious compounds to bind the particles of FA and GGBS to increase the unconfined compressive strength and the polymerization of FA in the presence of sodium hydroxide to enhance tensile strength are qualitatively in tune with the changes observed in both forms of strength and permeability, as indicated from detailed microstructural studies through SEM.
- Batch equilibrium studies revealed effective retention of ionic metal contaminants by precipitation at high alkaline conditions prevailing in the barrier system.

• The studies established the feasibility of FA-GGBS admixture polymerized with alkali as a potential material in barrier systems for waste containment facilities, making it a more sustainable barrier material than conventional natural soil barriers.

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