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Effect of Alkali-activation on Elaboration of a Binder Based on Ground Coal Gangue

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Abstract

The storage and accumulation of coal waste have adverse effects, including the utilization of valuable land resources, air pollution, and the potential for groundwater contamination. To address these challenges, this study investigated the feasibility of utilizing coal gangue as a raw material for alkali-activated binders, focusing on the influence of different sodium hydroxide (NaOH) concentrations. The coal gangue samples were subjected to various activation conditions using sodium hydroxide (NaOH) solutions of different concentrations (5 M and 12 M). The resulting binders were characterized using X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FT-IR), and thermogravimetric-differential thermal analysis (TG-DTA). XRD analysis confirmed successful activation, with higher NaOH concentrations resulting in increased aluminosilicate gel (N-A-S-H) and reduced quartz peaks. FT-IR analysis showed the formation of aluminosilicate, with higher NaOH concentrations leading to more significant gel formation. TG-DTA analysis revealed multiple stages of thermal decomposition and improved thermal stability with higher NaOH concentrations. Overall, the results highlight the influence of NaOH concentration on the properties of alkali-activated binders and emphasize the potential of coal gangue as a sustainable binder alternative.

Keywords

Coal gangue, Alkali-activated binder, Aluminosilicate gel, Alternative binder

Introduction

Coal gangue, a hazardous solid waste produced during coal mining, presents various drawbacks. Its accumulation and storage consume valuable land resources and contribute to the release of harmful gases into the atmosphere [1-3]. Furthermore, the presence of heavy metals like Pb^{2+}, Zn^{2+}, Cu^{2+}, and Cd^{2+} in coal gangue poses a risk of groundwater contamination through advection and diffusion processes, leading to secondary pollution [4]. Nonetheless, the distinctive physical, chemical, and mineralogical properties of coal gangue allow for its utilization as an additional aluminosilicate material, facilitating the formation of amorphous phases and less crystalline products in alkali-activated materials. These materials exhibit promising potential as eco-friendly alternatives to conventional construction materials [5, 6]. Quartz, muscovite, and kaolinite are among the primary mineralogical constituents of coal gangue [6]. In the chemical composition of coal gangue from Jerada city, the combined weight percentages of SiO_2 and Al_2O_3 account for over 70% of the total weight, as demonstrated in table 1. However, raw coal gangue has a relatively stable crystalline structure, and its reactivity needs to be improved through activation methods before use [7]. Thermal, mechanical, chemical, and compound activation are the four main methods used to enhance the reactivity of coal gangue. Among these, thermal activation is considered the

most promising approach as it destabilizes the crystal structure and increases the reactivity of coal gangue. During thermal activation, kaolinite, a major crystalline component of coal gangue, undergoes a process of dehydroxylation, resulting in the formation of amorphous metakaolin, which exhibits excellent pozzolanic properties. Metakaolin can react with calcium hydroxide derived from cement hydration. The temperature range of 500 °C to 1000 °C is known to induce changes in the mineral composition of coal gangue. At 500 °C, kaolinite transforms into metakaolin (Equation 1), and at 1000 °C, it further transforms into crystallized mullite (Equation 2) [7, 8].

$$Al_2O_3.2SiO_2.6H_2O \xrightarrow{500\ ^\circ C} Al_2O_3.2SiO_2 + 2H_2O \text{ (Metakaolin)} \quad (1)$$

$$3(Al_2O_3.2SiO_2) \xrightarrow{1000\ ^\circ C} 3Al_2O_3.2SiO_2 + 4SiO_2 \text{ (Mullite)} \quad (2)$$

The formation of mullite at 1000 °C can reduce the reactivity of coal gangue. In a study by Liu et al. [9], the reactivity of coal gangue calcined at various temperatures (500 °C, 600 °C, 700 °C, 800 °C, and 900 °C for 2 hours) was evaluated in cement mortars. The results showed that the consumption of calcium hydroxide increased with higher calcination temperatures up to 800 °C, indicating the complete transformation from kaolinite to metakaolin. However, the reactivity decreased slightly at 900 °C due to the recrystallization of the amorphous phase (metakaolin) into the crystalline phase (mullite). Another study [10] demonstrated that mortars containing coal gangue calcined at 700 °C for 2 hours exhibited high reactivity and higher compressive strengths. Chemical activation involves mixing an aluminosilicate precursor with water or a strong alkali or acid solution (such as HCl or H_3PO_4). This process enhances the surface reactivity and porosity of clay particles by disaggregation, dissolution of outer layers, and removal of mineral impurities [11]. Commonly used activators include NaOH, KOH, Na_2SO_4, water glass, Na_2CO_3, K_2CO_3, K_2SO_4, or a small amount of cement clinker [12]. It has been reported by Duchesne et al. [13] that the presence of NaOH in the solution accelerates the alkaline activation reaction. Huang et al. [14] studied the influence of NaOH content on the alkali conversion mechanism in alkali-activated mortars and found that a higher NaOH content significantly improves the stimulation capacity of the activator, promoting the transformation of free alkali into union alkali. They also noted that higher alkalinity (pH between 13.5 and 14.5) in the polymerization environment favors the formation of high-strength aluminosilicate gels. Mechanical activation through grinding is a physicochemical method that reduces the particle size and dehydroxylates the hydroxyl groups in kaolinite, enhancing the reactivity of coal gangue without the need for calcination. However, sole reliance on mechanical activation may not be sufficient for practical engineering applications [7]. Mechanical-thermal activation, achieved by grinding and subsequent calcination at temperatures between 600 °C and 700 °C for 2 hours, with a fineness value less than 45 - 63 µm or approximately 300 - 400 m^2/kg, has demonstrated significantly increased coal gangue reactivity [15]. Nevertheless, this combined activation method requires high calcination temperatures, resulting in high-energy consumption. In this study, we propose a mechanical-chemical activation binary combination, as illustrated in figure 1. This study aims to explore the intricate interplay between grinding, NaOH concentration, and the chemical, mineralogical, and thermal properties of an alkali-activated binder derived from coal gangue. For this purpose, three alkali-activated coal gangue samples were meticulously prepared and activated using water and two distinct NaOH concentrations. Following a curing period of 28 days, the samples underwent comprehensive characterization through XRD, FT-IR, and TG-DTA.

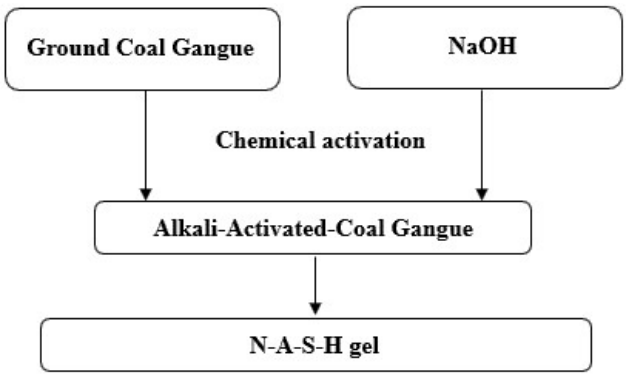

Figure 1: The schematic representation of the binary combination of mechanical and chemical activation.

Materials and Method

Preparation

Mining waste-coal gangue from Jerada city provides a significant opportunity to tap into this resource and use it to produce alkali-activated coal gangue. Table 1 shows the results of an X-ray fluorescence analysis on coal gangue from Jerada, Morocco. The mineralogical composition of the raw coal gangue was determined using FullProf software and the Rietveld method to analyze the X-ray diffractogram. The results showed that the predominant mineralogical phases of coal gangue were muscovite ($KAl_2(AlSi_3O_{10})(OH,F)_2$) with a value of 53.01%, quartz (SiO_2) with a value of 30.94%, and kaolinite ($Al_2O_3.2SiO_2.2H_2O$) with a value of 14.98% (Figure 2a and 2b). The schematic diagram of the mechanical-chemical activation binary combination is shown in figure 1. It is not possible to induce a polymerization reaction by directly activating raw coal gangue with a strong alkali activator. However, grinding the coal gangue into fine powders increases its reactivity by releasing bound water in the kaolinite phase, elim-

Table 1: Chemical composition of coal gangue.

Oxides (Materials)	Coal gangue (Weight %)
SiO_2	52.4
Al_2O_3	21.9
CaO	0.81
Fe_2O_3	4.55
SO_3	3.54
K_2O	2.24
Na_2O	1.53
MgO	1.26
P_2O_5	0.135
LIO (loss of ignition)	10.7

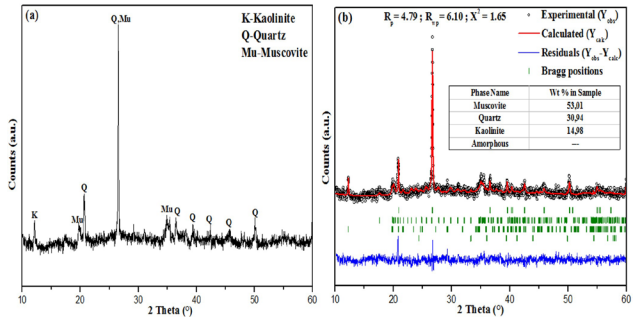

Figure 2: XRD spectra and Rietveld refinement of the raw coal gangue.

inating the need for calcination [7]. The favorable reactivity of calcined coal gangues with a fineness value of less than 45 µm was highlighted in a study on the potential use of calcined kaolinite-based wastes as cement replacements in concrete [15]. Three samples were prepared in this study to investigate the effects of grinding and NaOH concentration. Two concentrations of NaOH (5 M and 12 M) were used, as well as a distilled water-hydrated sample. The sample was prepared in three steps: first, the raw coal gangue was ground for a few minutes in a planetary mill at 600 r/min with a laboratory grinder, and the resulting particles were sieved to obtain sizes smaller than 40 m. The ground coal gangue was then placed on a 4 cm x 4 cm glass slide and soaked in a few milliliters of alkaline solution. All samples were formulated with a water/binder ratio of 0.40 and cured at 20 °C in a laboratory desiccator, which provided a controlled curing environment representative of practical conditions in various regions. Table 2 presents the sample matrix proportions. The alkaline activation reaction was stopped after 28 days of curing by wetting the activated samples with a few milliliters of ethanol solution. XRD, FT-IR, and TG-DTA were used to characterize the resulting products.

Characterization

The phase structures of the samples were checked by XRD. Diffractograms were recorded on a Shimadzu 6100 diffractometer using a Cu (Kα) signal at 40 kV and 25 mA, with step size of 0.04° over the range 10 to 60° (2θ) at a scan speed of 2.4°/min. The diffraction peaks of crystalline phases were identified using the International Centre for Diffraction Data (ICDD) database. The weight percentages of the detected phases were calculated by applying the Rietveld Method using the FullProf software package [16]. Structural characterizations of the different functional groups were performed using a Jasco FT/IR 4600 FT-IR fitted with a reflectance accessory total attenuated type Pro One, in the frequency region of 4000 to 400 cm^{-1}. TG-DTA were performed on a Setaram instrument (LabsysEvo 1F). A sample of approximately 10.5 mg was carried out between 50 °C and 1000 °C at a heating rate of 10 °C/min under a dried-air atmosphere.

Results and Discussion

X-ray diffraction and Rietveld quantitative phase analysis

Figure 2 shows the XRD patterns of uncalcined coal gangue. The X-ray diffractogram was also analyzed with FullProf software using the Rietveld method to determine the possible mineralogical composition of the raw coal gangue. The CIF files used for the Rietveld refinement of each phase were extracted from the Crystallography Open Database. The diffraction peaks were modeled using the Pseudo-Voigt function. Scale factors, instrumental parameters, background coefficients, lattice constants, profile half-width (u, v, w), and preferential orientations have been refined. The good quality of the structural refinement can be confirmed by the negligible differences between the experimental patterns and theoretical data (the intensity of the residual plots (blue curves) close to zero) and the low deviations of the reliability factors (R_p < 10, R_{wp} < 10, and X^2 < 4). The results of the analyzed X-ray diffractogram with FullProf software using the Rietveld method to determine the possible mineralogical composition of raw coal gangue showed that the major mineralogical phases of coal gangue were muscovite ($KAl_2(AlSi_3O_{10})(OH,F)_2$) with a value of 53.01%, 30.94% of quartz (SiO_2), and 14.98% of kaolinite ($Al_2O_3.2SiO_2.2H_2O$) (Figure 2b). Because of their stable chemical structures, it is widely assumed that the presence of inert muscovite, kaolinite, and quartz in these percentages is the cause of coal gangue's low activity [16].

Figure 3 shows the XRD diffractograms of the synthesized samples after a 28-day curing period at room temperature. The results show that as the NaOH concentration increases, the intensity of crystal phase peaks, specifically muscovite and quartz decrease noticeably. Furthermore, the kaolinite peak is completely absent, indicating that the internal hydroxyl structure has been eliminated. There are also notable new diffractions corresponding to the sodium silicate phase $Na_6Si_8O_{19}$ and the sodium aluminosilicate phase. These phases are critical in the formation of an alkaline medium as well as geological polymerization [17]. With increasing NaOH concentrations, the intensity of these diffractions increases. Gao et al. [18] investigated the effect of NaOH concentration on SiO_3^{2-} leaching in their study. According to the findings of the study [18], higher NaOH concentrations promote the formation of aluminosilicate

Table 2: Designed proportions of sample matrices.

Samples	Raw materials	Solid addition/Wt.%	L/S ratio	Pre-activated way
Coal Gangue-water (CG-W)	Coal Gangue	2	0.4	Griding; Hydraulic reactivity (H_2O)
Coal Gangue-NaOH 5 M (CG-5M)	Coal Gangue	2	0.4	Griding; Alkaline reactivity (NaOH 5 M)
Coal Gangue-NaOH 12 M (CG-12M)	Coal Gangue	2	0.4	Griding; Alkaline reactivity (NaOH 12 M)

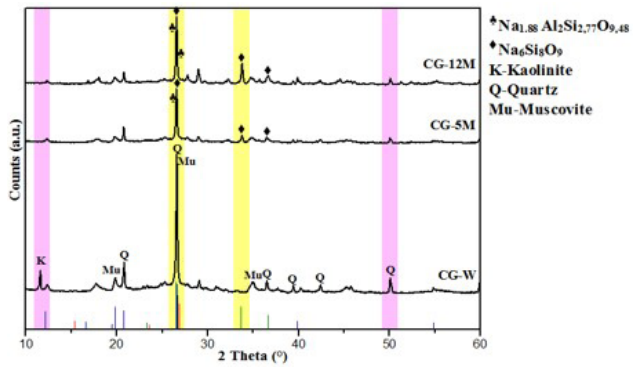

Figure 3: XRD patterns of the alkali-activated coal gangue samples after 28 days of curing.

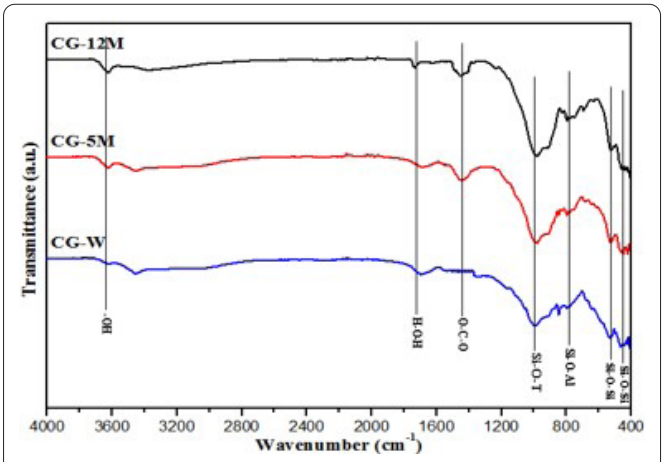

Figure 4: FT-IR spectra of the alkali-activated coal gangue samples after 28 days of curing.

gel. When the NaOH concentration is less than 25%, the reaction between alkali and active SiO_2 produces a low modulus, water-soluble $Na_2O.nSiO_2$, as shown in equation 3. When the concentration of NaOH exceeds 25%, the excess alkali reacts with active Al_2O_3 to produce $NaAlO_2$, which then reacts with $Na_2O.nSiO_2$ to form an insoluble sodium aluminosilicate gel, as shown in equation 4.

$$2NaOH + nSiO \longrightarrow Na_2O.nSiO_2 + 3H_2O \quad (3)$$

$$Na_2O.nSiO_2 + 2NaAlO_2 + 2H_2O \longrightarrow \underbrace{Na_2O.Al_2O_3.nSiO_2.H_2O}_{\text{Sodium aluminosilicate (N-A-S-H)}} + 2NaOH \quad (4)$$

The identification of specific peaks in the XRD analysis in our study validates this hypothesis. A distinct peak at 26.62° indicates the presence of the sodium aluminosilicate ($Na_{1.88}Al_2Si_{2.77}O_{9.48}$) phase (ICDD card No. 048-0730), while a prominent peak at 33.82° indicates the presence of the sodium silicate ($Na_6Si_8O_{19}$) (ICDD card No. 049-0162). Huang et al. [14] examined the XRD patterns of solidified paste samples in their study. The results showed that quartz crystalline phases and aluminosilicate gel phases coexisted. The weak characteristic peaks associated with the gel phases, on the other hand, indicated insufficient production. The XRD diagrams showed more significant changes when the samples were activated with a higher concentration of NaOH. The intensities of the crystalline peaks were reduced even further when compared to samples activated with lower NaOH concentrations, indicating a greater degree of mineral phase dissolution and transformation. Furthermore, the XRD patterns showed broader and weaker peaks, indicating a more amorphous structure. These findings suggest that increasing the NaOH concentration promotes enhanced crystalline phase dissolution and transformation, resulting in the formation of more amorphous alkali-activated products. The increased NaOH concentration increases the alkalinity of the activating solution, allowing minerals to dissolve and amorphous aluminosilicate gels to form.

Fourier transform infrared spectroscopy

The FT-IR spectra of alkali-activated coal gangue samples after 28 days of curing are shown in figure 4 in the frequency range 400 to 4000 cm^{-1}. The spectra revealed that all samples have a prominent and intense band between 800 and 1200 cm^{-1}, which corresponds to the Si-O-T asymmetric stretching vibration (where T stands for Al or Si). This distinct band supports geological polymerization [19] and indicates the formation of an aluminosilicate gel (N-A-S-H) [20], which correlates with XRD results. The main spectral bands of the FT-IR spectra of the samples CG-W, CG-5M, and CG-12M, respectively, are 11001.83 cm^{-1}, 1997.98 cm^{-1}, and 984.48 cm^{-1}. In other words, as the NaOH concentration increases, the absorption peak shifts to lower wavenumbers, indicating that the Si-O bands extend, and the bond angle decreases [21]. Any decrease in frequency in this region indicates more tetrahedral aluminum incorporation in the geological polymerization process [22, 23]. The spectral band at 790 cm^{-1} indicates the stretching vibration of Si-O-Si or Si-O-Al. The 450 cm^{-1} and 480 cm^{-1} absorption bands are attributed to Si-O-Si bending vibration modes in the plane of the coal gangue SiO_4 tetrahedra, and the intensity of these bands decreases as NaOH concentration increases, indicating the formation of aluminosilicate gels. García-Lodeiro et al. [24] investigated the chemical changes in an alkali-activated material using FT-IR analysis with varying concentrations of NaOH in a research study. When the binder was activated with NaOH solutions less than 5 M, the FT-IR spectra revealed distinct absorption peaks corresponding to different functional groups. The intensity of peaks associated with Si-O-Si bonds decreased as the NaOH concentration increased, indicating the formation of aluminosilicate gels. Additionally, the emergence of new peaks in the 1200 - 800 cm^{-1} range indicated the presence of newly formed phases. Furthermore, when a higher NaOH concentration of 12 M was used, the intensity of peaks associated with aluminosilicate gels increased, indicating a more extensive reaction and the formation of more gels. FT-IR analysis also revealed an absorption band at 1477 cm^{-1}, which is attributed to the stretching vibrations of the carbonate group. This absorption peak indicates the presence of carbonation in the alkali-activated coal gangue binder during handling. As atmospheric CO_2 enters the alkali-activated sample, it reacts with unreacted sodium, resulting in the formation of the $NaCO_3.H_2O$ compound [25, 26]. The stretching vibrations of the H-O-H bond at 3610 cm^{-1} and the bending vibrations of the -OH bond at 1650 cm^{-1} were also observed. These peaks indicate the amount of chemically bound water present in the alkaline-activated carbon gangue samples. A higher peak intensity indicates a greater amount of chemically bound water within the binder [14].

Thermal behavior

Figure 5 depicts the TG-DTA results of the various samples of alkali-activated coal gangue synthesized. All the specimens exhibited similar characteristic trends up to 1000 °C. The TG curves are classified into three steps. The evaporation of the water adsorbed on the surface and the water inside the pores of the alkali-activated binder N-A-S-H resulted in significant weight loss in each alkali-activated coal gangue sample from room temperature to 300 °C. Weight loss for CG-5M and CG-12M is similar but significantly greater than CG-W below 300 °C. The release of condensed/polymerized water in the Si-OH and Al-OH groups results in additional weight loss between 300 and 600 °C. The third temperature range, 600 - 900 °C, is attributed to the combustion of unburned carbon in raw materials and the decomposition of carbonate species (Na_2CO_3 and $CaCO_3$), which confirms the XRD and FT-IR results. The mass loss of the samples tends to gradually stabilize after 900 °C. Three distinct features were observed in the CG-12M curve, as shown by the DTA curve. The endothermic peak at 103 and 300 °C was attributed to evaporation of adsorbed and interstitial water in the alkali-activated binder structure, and it was consistent with TG-DTA analysis. The unburnt carbon in the raw material is responsible for the large exothermic peak observed in the temperature range of 400 to 600 °C. Other studies have found similar results [27-30]. Cheng et al. [29] investigated the thermal behavior of coal gangue geopolymers using TG-DTA analysis in a research study. The geopolymers were made with different sodium hydroxide (NH) moduli of 8 M, 12 M, and 16 M, and their thermal properties were tested from 40 °C to 600 °C. The primary weight loss of coal gangue geopolymers occurred between 60 and 200°C, which was attributed to the dehydration of N-A-S-H gels, which represented hydrated aluminosilicates. Interestingly, when the NaOH modulus was 8 M, the weight loss was minimized, while the differences in weight loss between the 12 M and 16 M moduli were negligible.

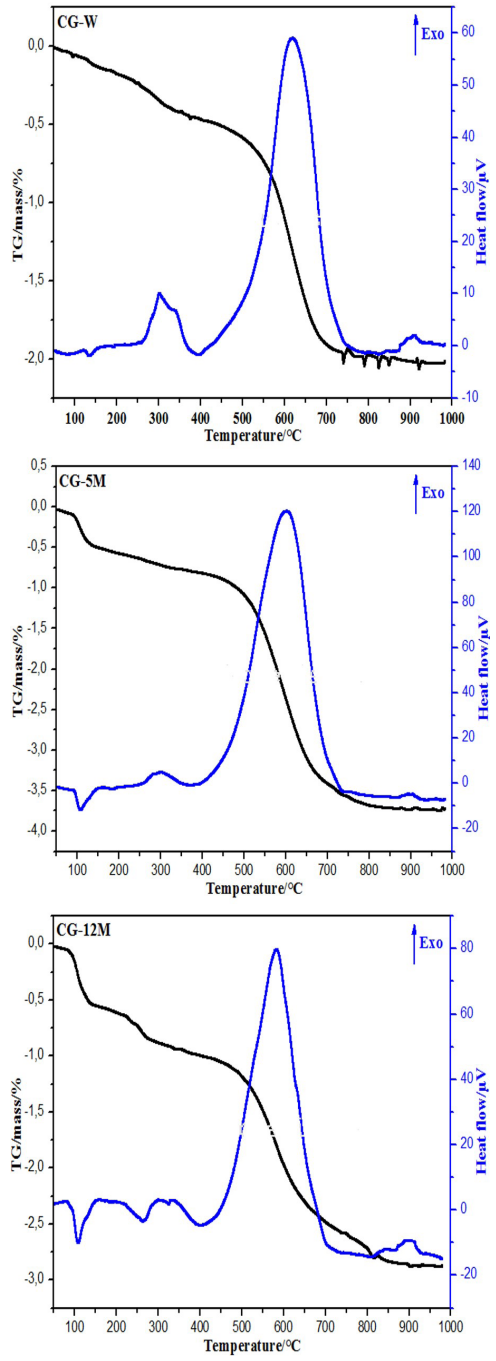

Figure 5: TG-DTA diagram of the alkali-activated coal gangue samples after 28 days of curing from room temperature to 1000 °C.

Conclusion

This study explored the use of coal gangue, a waste by-product from coal mining, as a raw material for alkali-activated binders' production. The alkali-activated coal gangue samples were subjected to different activation conditions using NaOH solutions of different concentrations (5 M and 12 M). XRD, FTIR, and TG-DTA analyses confirmed the successful transformation of coal gangue into amorphous aluminosilicate phases. Higher NaOH concentrations led to a greater reduction in quartz peaks, increased geopolymeric gel formation, and improved thermal stability. The study provides valuable insights for optimizing the activation process and highlights the potential of using coal gangue as an environmentally sustainable alternative for binder production.

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Conflict of Interest

None.

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