

# Green-Synthesized Geopolymer from Acacia Extract and Coal Fly Ash for Sustainable Dye Degradation in Wastewater

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## ABSTRACT

The continuous discharge of dye-based industrial effluents into aquatic environments has become a serious ecological concern due to their toxicity, persistence, and adverse effects on human health and biodiversity. In the present work, a sustainable, eco-friendly geopolymer composite was synthesized via a green synthesis approach using coal fly ash and Acacia extract to efficiently remove methylene blue (MB) dye from wastewater. Coal fly ash, an industrial by-product rich in aluminosilicates, was alkali-activated with sodium hydroxide and sodium silicate solutions in order to create a geopolymer matrix. Acacia extract was used as a natural source of bioactive compounds that enhanced the surface functionality of the composite. The synthesized material was characterized using Fourier Transform Infrared Spectroscopy (FTIR), UV-Vis Spectroscopy, and Scanning Electron Microscopy (SEM) to investigate its structural, morphological, and elemental properties. FTIR analysis confirmed the formation of a geopolymer aluminosilicate network via characteristic Si-O-Si and Si-O-Al stretching vibrations, while SEM micrographs revealed a highly porous, interconnected surface morphology helpful to adsorption and photocatalytic reactions. UV-Visible spectroscopic studies showed a significant decrease in the characteristic absorption peak of methylene blue after treatment, indicating efficient dye degradation. Photocatalytic experiments demonstrated that the dye removal efficiency gradually increased with contact time, reaching about 78% after 120 min of visible-light irradiation. The pseudo-first-order kinetics of the degradation process indicate efficient interaction between the dye molecules and the active sites of the geopolymer composite.

**Keywords-** Green synthesis, Acacia extract, coal fly ash, geopolymer composite, photocatalysis, methylene blue, wastewater treatment, dye degradation, environmental remediation.

## I. INTRODUCTION

A promising approach to the development of low-carbon adsorbents is the combination of industrial by-products, such as coal fly ash, with bio-based additives [1]. These materials exploit the inherent aluminosilicate structure of fly ash to enhance surface adsorption mechanisms for the removal of persistent organic pollutants [2]. Additionally, natural phytochemicals in acacia extract are incorporated to modify the geopolymer matrix, potentially enhancing the active surface area and porosity required for efficient photocatalytic degradation of recalcitrant dyes [3]. This hybrid approach efficiently reduces the environmental

impact of synthetic dyes and valorizes the abundant industrial waste streams [4], [5]. Furthermore, these modifications can also lead to enhanced structural integrity of the adsorbent, enabling more robust operation in diverse aquatic environments [6]. By optimizing these synthesis parameters, researchers can tune the porosity and chemical surface functionality of the geopolymer to target specific industrial contaminants, such as cationic dyes and persistent textile effluents [7], [8]. The versatility of these inorganic polymers can be further extended to the immobilization of heavy metals, which co-exist with organic pollutants in untreated industrial discharge [9].

The chemical stability and mechanical resistance of these materials are due to their amorphous polymeric structures, which allow them to remain durable during long-term wastewater treatment processes [10], [11]. Moreover, the combination of fly ash-based frameworks with bio-organic modifiers has shown high adsorption capacities for phenolic compounds with higher removal efficiencies than conventional untreated materials [12]. These sustainable composite materials provide a viable solution to the global problem of wastewater management by providing a low-cost alternative to expensive commercial adsorbents such as activated carbon. This dual utility results in a significant reduction in the ecological footprint associated with waste deposition from coal-fired power plants, on the one hand, and addresses the risks posed by the mutagenic and carcinogenic properties of persistent synthetic dye effluents in aquatic ecosystems [13], [14]. The alkali activation process can be improved, and the incorporation of bio-modifiers can be optimized to enhance these composites' adsorption capacity in multi-component systems [15], [16]. Future work should address the scalability of these geopolymerization protocols at low temperatures to improve economic viability and avoid energy-intensive thermal treatment methods [17]. The combination of geopolymer-biopolymer composites may be a way to improve the simultaneous removal of multiple co-contaminants, such as heavy metal ions and complex dye mixtures [18]. The use of such innovative materials on an industrial scale will further divert industrial by-products from landfills and align with the principles of the circular economy in wastewater infrastructure [19].

Ultimately, the use of these inorganic amorphous aluminosilicates represents a step towards scalable, low-cost remediation strategies that can mitigate the environmental persistence of complex industrial effluents [20], [21]. In addition, the valorization of industrial residues such as fly ash aligns with the global drive to convert waste into functional materials, thereby alleviating the serious ecological burden of fly ash disposal [22], [23]. Moreover, the technical and economic hurdles to upscaling these laboratory-synthesized composites remain critical to achieving significant environmental and industrial synergy [24].

## II. MATERIALS AND METHODS

### *Preparation of Acacia bark extract*

Acacia bark was collected from the Yamuna Biodiversity Park, Delhi Development Authority, Delhi. Washed and dried at room temperature until crispy, powdered finely, sieved to 125–250  $\mu\text{m}$ , and washed several times with distilled water. Then about 30 g of acacia bark powder was boiled in a 250 ml beaker with the addition of 5 ml (5 N) HCl to activate the adsorbents and to remove the dark green colour and soluble

impurities. The acacia bark powder was filtered and boiled 2–3 times with distilled water again for complete removal of the colour and dried at room temperature for 48 h. The moist activated acacia bark was placed in the oven at a temperature of about 60–70°C for 6–8 h to remove the moisture completely, and the dry activated acacia bark powder was stored in an airtight vessel [25].

Geopolymer composites based on acacia bark powder can be prepared with coal fly ash as the main aluminosilicate precursor and an alkaline activator of sodium hydroxide (NaOH) and sodium silicate ( $\text{Na}_2\text{SiO}_3$ ). Acacia bark powder is used in most of the experimental studies as a partial replacement of fly ash at the rate of 5–10% by weight because higher percentages may reduce the compressive strength due to the organic nature of the biomass material.[26] The alkaline activator is prepared by dissolving NaOH pellets in distilled water to obtain a solution concentration of around 10–12 M. The NaOH solution is left to cool down for 24 hours before mixing with the sodium silicate solution. A commonly reported effective proportion of activator for fly ash geopolymers is [27]

$$\frac{\text{Na}_2\text{SiO}_3}{\text{NaOH}} = 2.5$$

This ratio improves the geopolymerization, the compressive strength, and the bonding properties of the matrix. In the first step, fly ash and acacia bark powder are dry-mixed properly to ensure homogeneity for the preparation of geopolymers. Then, the alkaline activator is added slowly, with constant stirring, to form a homogeneous paste. Fresh geopolymer paste is poured into molds and compacted by vibration or tamping to remove entrapped air. The specimens are cured either at ambient temperature or under heat-curing conditions after casting. Heat curing is usually done at 60–70°C for 24 h, which accelerates geopolymerization and gives higher early compressive strength. The dry materials were mixed for about three minutes. The dry materials included fly ash and aggregates. Then the prepared alkaline activator and superplasticizer are added and mixed for a further 4–5 min until a homogeneous, glossy mixture is obtained.[28]

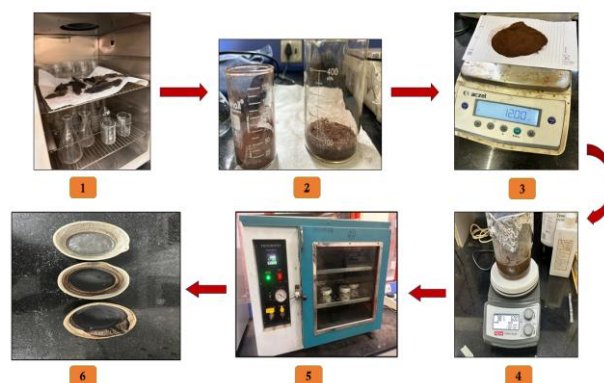


Table 1: Contents of the composition for the preparation of geopolymers

| Acacia bark(g) | Fly ash (gram) | Na <sub>2</sub> SiO <sub>3</sub> solution (gram) | NaOH solution (gram) | Total activator (gram) | Binding activator (gram) |
|----------------|----------------|--------------------------------------------------|----------------------|------------------------|--------------------------|
| 0              | 10             | 28.6                                             | 11.4                 | 40                     | 50                       |
| 0.5            | 9.5            | 28.6                                             | 11.4                 | 40                     | 50                       |
| 1              | 9              | 28.6                                             | 11.4                 | 40                     | 50                       |

III. RESULTS AND DISCUSSION

Characterization

The photocatalytic efficiency of green synthesized geopolymer composites from Acacia extract and coal fly ash was investigated by using methylene blue (MB) dye as a model organic pollutant under visible light irradiation. FTIR analysis revealed the successful geopolymerization of coal fly ash and incorporation of bioactive functional groups from the Acacia extract. The presence of a characteristic band around 995-1085 cm<sup>-1</sup>, due to the Si-O-Si/Si-O-Al stretching vibrations, confirms the formation of a stable aluminosilicate geopolymer network. The broad bands at 3420–3445 cm<sup>-1</sup> and 1648–1630 cm<sup>-1</sup> are attributed to O–H stretching and H–O–H bending vibrations, respectively, indicating the presence of hydroxyl groups, which can

provide active sites in the photocatalytic reactions [34-35].

Scanning Electron Microscopy (SEM) images further support the enhanced photocatalytic performance of the geopolymer composite. The raw fly ash mainly consists of smooth, spherical particles typical of aluminosilicate glass microspheres. After alkali activation and geopolymerization, the microstructure changes into a highly porous interconnected network with irregular surface morphology. SEM images at higher magnification indicate the existence of numerous pores and channels to enhance the surface area for dye adsorption and photocatalytic reactions. The porous structure helps in the mass transfer of dye molecules and the interaction between pollutants and reactive species generated during irradiation (Fig. 1) [36-37].

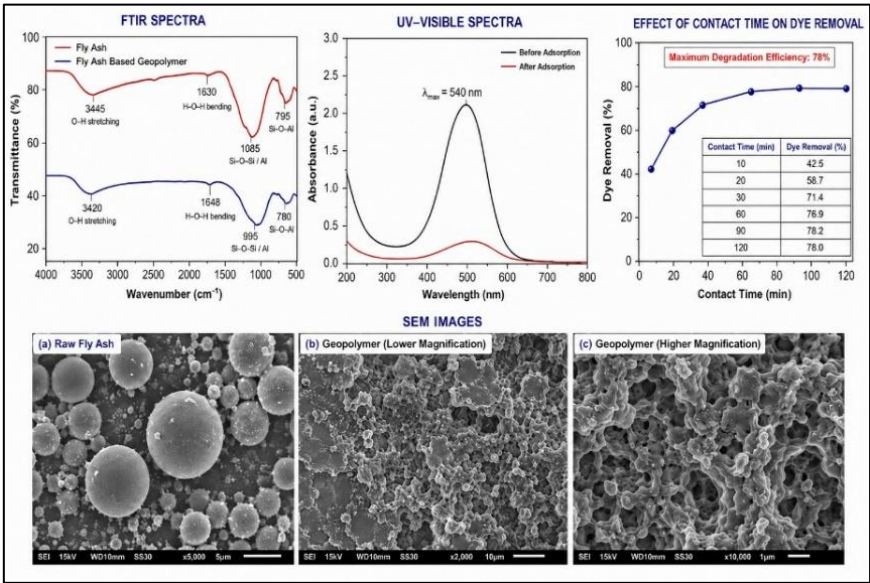


Fig. 1. Different Characterisation of AB powder and CFA-derived geopolymer

Photocatalytic Degradation Data

| Irradiation Time (min) | Absorbance at 540 nm | Degradation Efficiency (%) |
|------------------------|----------------------|----------------------------|
| 0                      | 2.10                 | 0.0                        |
| 10                     | 1.21                 | 42.5                       |
| 20                     | 0.87                 | 58.7                       |
| 40                     | 0.60                 | 71.4                       |
| 60                     | 0.49                 | 76.9                       |
| 90                     | 0.46                 | 78.2                       |
| 120                    | 0.46                 | 78.0                       |

The photocatalytic activity of the acacia extract-based geopolymer and coal fly ash green synthesis was studied for the degradation of methylene blue (MB) dye under visible-light irradiation. The efficacy of the photocatalysis was examined using UV-Visible spectroscopy by monitoring the change in the characteristic absorption peak of methylene blue at 540 nm. The synthesized geopolymer showed a porous aluminosilicate structure, containing bioactive phytochemicals of acacia extract, which enhanced the dye molecules adsorption and facilitated photocatalytic reactions on the catalyst surface [38-39].

The UV-Visible absorption spectra showed a gradual decrease in the intensity of the methylene blue absorption peak with increasing irradiation time, indicating the progressive degradation of dye molecules. The untreated dye solution showed a strong absorption maximum at 540 nm, corresponding to the chromophoric structure of methylene blue initially. The absorbance intensity decreased significantly after treatment with the geopolymer photocatalyst, indicating degradation of the dye molecules. After 120 min of irradiation, the absorption peak almost disappeared, which confirms extensive degradation and decolorization of the dye solution [40].

The photocatalytic degradation efficiency was gradually improved with the irradiation time. The dye removal efficiencies after 10, 20, 40, and 60 min were 42.5, 58.7, 71.4, and 76.9%, respectively. Further irradiation led to 78 % degradation at 90 min, while the maximum degradation efficiency was reached at 120 min, which was 78 %. The decrease in UV-Visible absorbance was directly proportional to the decrease in dye concentration, confirming the efficiency of the geopolymer composite in eliminating organic pollutants from aqueous media [40-41].

The observed photocatalytic activity can be ascribed to the generation of electron-hole pairs under light irradiation. These charge carriers react with adsorbed water molecules and dissolved oxygen to form highly reactive hydroxyl radicals ( $\bullet\text{OH}$ ) and superoxide radicals ( $\text{O}_2\bullet^-$ ). The successive oxidation and mineralization of methylene blue by reactive oxygen species on the aromatic ring structure and chromophoric groups produces harmless products such as carbon dioxide and water. Moreover, the porous microstructure of the geopolymer increases the diffusion of dye molecules and the accessibility of active reaction sites [42].

The degradation kinetics (degradation efficiency) followed a pseudo-first-order model according to the Langmuir-Hinshelwood equation:

$$\ln\left(\frac{C_0}{C_t}\right) = kt$$

where  $C_0$  and  $C_t$  represent the initial and residual dye concentrations, respectively, and  $k$  is the

apparent rate constant. The linear relationship between  $\ln(C_0/C_t)$  and irradiation time confirms that the photocatalytic degradation process is predominantly governed by surface-mediated reactions.

The UV-Visible absorption studies and degradation efficiency measurements collectively suggest that the fly ash geopolymer derived from Acacia exhibits strong photocatalytic activity toward methylene blue degradation. The significant decrease in absorbance at 540 nm and removal of approximately 78% of dye in 120 min suggest that this eco-friendly geopolymer can serve as a sustainable photocatalyst for wastewater treatment and environmental remediation.

#### IV. CONCLUSION

This study successfully synthesized a green geopolymer from Acacia extract and coal fly ash for sustainable application in wastewater treatment. Characterization studies showed the formation of a stable aluminosilicate network with a porous structure conducive to photocatalytic activity. UV-Visible analysis revealed the efficient degradation of methylene blue dye with nearly 78% removal within 120 min under visible-light irradiation. The results indicate that the fly ash geopolymer derived from acacia can be used as a low-cost, eco-friendly, and sustainable material in the removal of dyes in wastewater remediation and environmental protection.

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