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PHOSPHORIC ACID-ACTIVATED CARBON FROM COFFEE GROUNDS AND FLY ASH FOR EFFICIENT LAUNDRY WASTEWATER ADSORPTION

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ABSTRACT

Laundry waste that is directly discharged into water bodies causes pollution due to its chemical content, such as phosphate. The adsorption method is chosen as an option to reduce the phosphate content in laundry waste due to its simple preparation and low cost. The adsorbent used is a mixture of coffee grounds and fly ash. The use of coffee grounds is based on the increasing waste from coffee shops, thus being utilized as an adsorbent. The aim of this study is to determine the effectiveness of coffee grounds and fly ash with the best combination of composition and contact time using the adsorption method. The stages begin with the preparation of coffee grounds and fly ash through carbonization and activation using H_3PO_4. The variations performed consisted of variations in the composition of coffee grounds and fly ash with a total of 1 gram in ratios of 1:4; 2:3; 1:1; 3:2; 4:1 and contact time variations of 15 minutes, 30 minutes, 45 minutes, 60 minutes, and 75 minutes. Based on the research results, the highest percentage of removal was found in the composition variation of coffee grounds and fly ash with a ratio of 1:4 for 75 minutes.

Keyword: Adsorption, Coffee Grounds, Phosphate, Fly Ash, Laundry Waste.

1. INTRODUCTION

Phosphorus is a critical nutrient that can cause eutrophication in aquatic ecosystems when present in excess [1]. Laundry wastewater, which often contains phosphate-based detergents, constitutes a significant source of phosphate pollution; indeed, laundry effluent may contain 70–80 % phosphate by mass [2]. Discharge of such phosphate-rich effluent into receiving waters can therefore trigger algal blooms and degrade water quality. The connection between detergents and phosphate-driven eutrophication is well documented, underscoring the need for effective phosphate removal from laundry wastewater.

Adsorption is widely regarded as an effective treatment for phosphate-laden wastewater due to its simplicity, low operational cost, and high removal efficiency [3]. In adsorption processes, target ions (adsorbates) transfer from the aqueous phase onto the surface of a solid adsorbent. Compared to other methods such as chemical precipitation or biological nutrient removal, adsorption offers advantages of a straightforward process and the potential for regeneration of the adsorbent for repeated use.

Coffee grounds and fly ash are attractive low-cost precursors for activated-carbon adsorbents [4]. Coffee Grounds are an abundant waste biomass (given the global scale of coffee consumption) and have been characterized as a valuable carbon-rich material for adsorbent production. They typically contain over 50 % fixed carbon, which can be converted into a porous carbon structure with high surface area [5] . Likewise, fly ash contains large amounts of silica (SiO_2) and alumina (Al_2O_3). These aluminosilicate

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components can be activated (for example, by alkaline or acidic treatment) to form porous, zeolite-like structures that enhance adsorption capacity. Valorizing coffee grounds and fly ash into adsorbents thus provides a cost-effective and sustainable way to both manage waste and remove pollutants from water.

To further enhance adsorbent performance, chemical activation is often employed. Phosphoric acid (H_3PO_4) activation is a well-established method to develop highly porous carbon adsorbents. During H_3PO_4 activation, a substantial amount of volatile matter is removed and new pores are generated in the carbon matrix [6]. The resulting activated carbon exhibits an extensive network of cavities and channels, greatly increasing its surface area and providing abundant sites for adsorption. In addition, H_3PO_4 activation introduces or preserves oxygen-containing functional groups on the carbon surface. These functional groups can increase the affinity of the carbon for phosphate ions through specific chemical interactions. Importantly, phosphoric acid is relatively low-cost and environmentally benign compared to other activating agents, making it especially attractive for large-scale production of high-quality activated carbons [7].

In this context, the present study investigates activated carbon made from a mixture of spent coffee grounds and fly ash for the removal of phosphate from laundry wastewater. The work focuses on optimizing the adsorbent composition and adsorption conditions. In particular, we examine different coffee:fly ash mass ratios and adsorption contact times with the goal is to identify the optimal formulation and operating time that maximize phosphate uptake efficiency under realistic wastewater conditions.

2. RESEARCH METHOD

All reagents and adsorbent precursors were of analytical grade. Spent coffee grounds were collected from local coffee shops, washed with deionized water, oven-dried at 105°C for 1 h, and stored in airtight containers. Coal fly ash was obtained from e-commerce, pre-heated at 600 °C for 2 h to remove moisture and organic impurities, and then cooled to room temperature. Phosphoric acid (H_3PO_4, 85 wt %) and deionized water were used for chemical activation and solution preparation. Synthetic laundry wastewater was prepared by dissolving commercial detergent (containing sodium tripolyphosphate) in deionized water to achieve an initial phosphate concentration of approximately 20 mg P/L.

Preparation of the activated adsorbents involved two main steps. First, coffee grounds were carbonized in a muffle furnace at 600°C for 3 h, then sieved to obtain a uniform particle size (100 mesh). The char was mixed with 0.1 M H_3PO_4 at a mass ratio of 1:3 (char:acid, b/v) and left to impregnate for 48 h at ambient temperature. After impregnation, the sample was filtered, washed with deionized water until neutral pH was reached, and dried at 110 °C for 30 min. Fly ash activation was achieved solely by heating at 600 °C for 2 h to increase porosity and remove residual impurities.

Batch adsorption tests were performed in 250 mL Erlenmeyer flasks containing 50 mL of synthetic laundry wastewater and 1.0 g of adsorbent. Five coffee-to-fly ash mass ratios (1:4, 2:3, 1:1, 3:2, and 4:1) were evaluated. Each flask was agitated on an orbital shaker at 150 rpm for contact times of 15, 30, 45, 60, and 75 min at room temperature. Upon completion of each contact period, samples were filtered, and the residual phosphate concentration was measured with a UV–Vis spectrophotometer. All experiments were conducted in duplicate, and the average values were reported.

3. RESULTS AND DISCUSSION

3.1. SEM Analysis of Adsorbent Morphology

Scanning electron microscopy (SEM) of the coffee-ground carbon before and after H_3PO_4 activation on Figure 1 reveals pronounced changes in surface topology and porosity. Prior to activation, the raw char exhibits a relatively smooth exterior with few visible pores. After phosphoric-acid treatment and thermal processing, the carbon surface displays an interconnected network of cavities and channels. Such hierarchical porosity arises from the dehydration and volatilization of organic components that "etch" the carbon matrix during activation, as described by Quesada-Plata *et al.* for other H_3PO_4-activated biomasses [8]. This pore enlargement not only increases the specific surface area but also enhances mass ttransfer pathways, facilitating rapid diffusion of phosphate ions into the adsorbent interior.

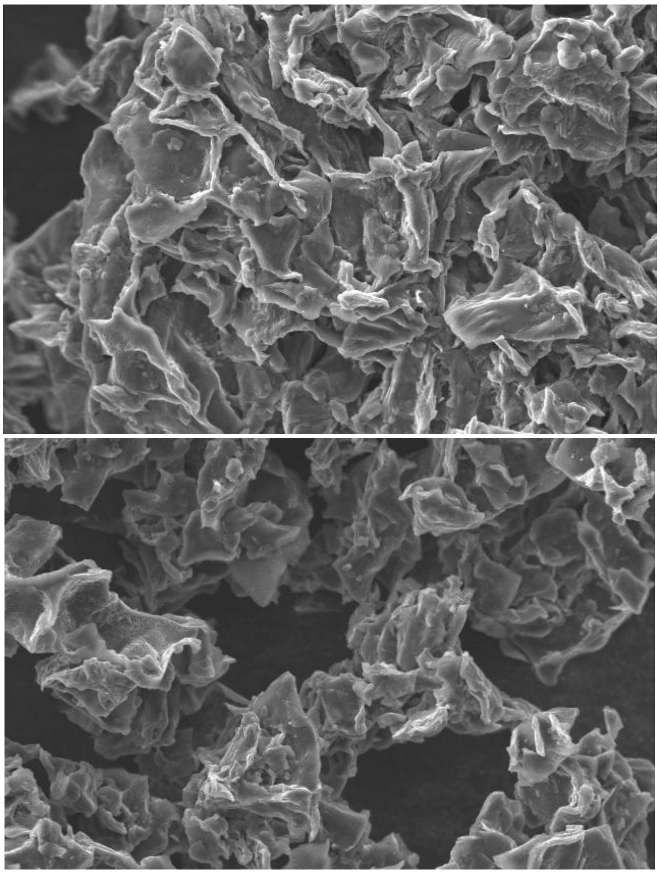

Figure 1. SEM of Coffee-Ground Carbon: (a) raw char before H_3PO_4 activation; (b) Activated Carbon after H_3PO_4 Treatment

Similarly, SEM images of fly ash on Figure 2 illustrate substantial particle size reduction and surface roughening following thermal activation at 600 °C. The original fly ash particles, which typically measure several micrometers in diameter, fracture into smaller fragments with irregular shapes and microcracks. This phenomenon parallels observed. in H_3PO_4-activated diatomaceous earth, where acid treatment disrupts aluminosilicate frameworks, generating new voids and microfractures that contribute to mesoporosity [9], [10]. The increased surface roughness and micro-crack density on fly ash provide additional active sites that can interact with phosphate via ligand exchange or hydrogen bonding [11].

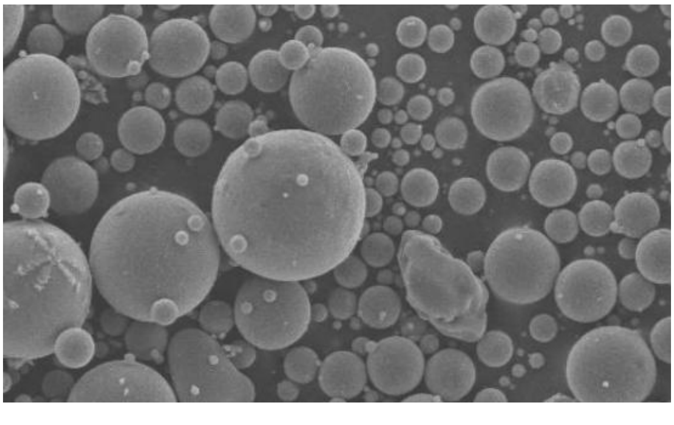

Figure 2. SEM of Fly Ash Particles

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3.2. FTIR Spectral Analysis

FTIR spectra of the coffee-ground activated carbon at Figure 3 (before and after H_3PO_4 activation) revealed key functional groups. A persistent peak near 1570 cm^{-1} was assigned to C=C stretching in aromatic rings, indicating a stable graphitic carbon backbone. More importantly, activation introduced a broad O–H band around 3420 cm^{-1} and a C–O band near 1100–1140 cm^{-1}. These correspond to hydroxyl and carbonyl groups (arising from residual cellulose), which are known to bind anions through hydrogen bonding or ligand exchange. The intensity of the C–O band diminished slightly after activation, implying partial removal of oxygenated surface groups and likely an increase in porosity.

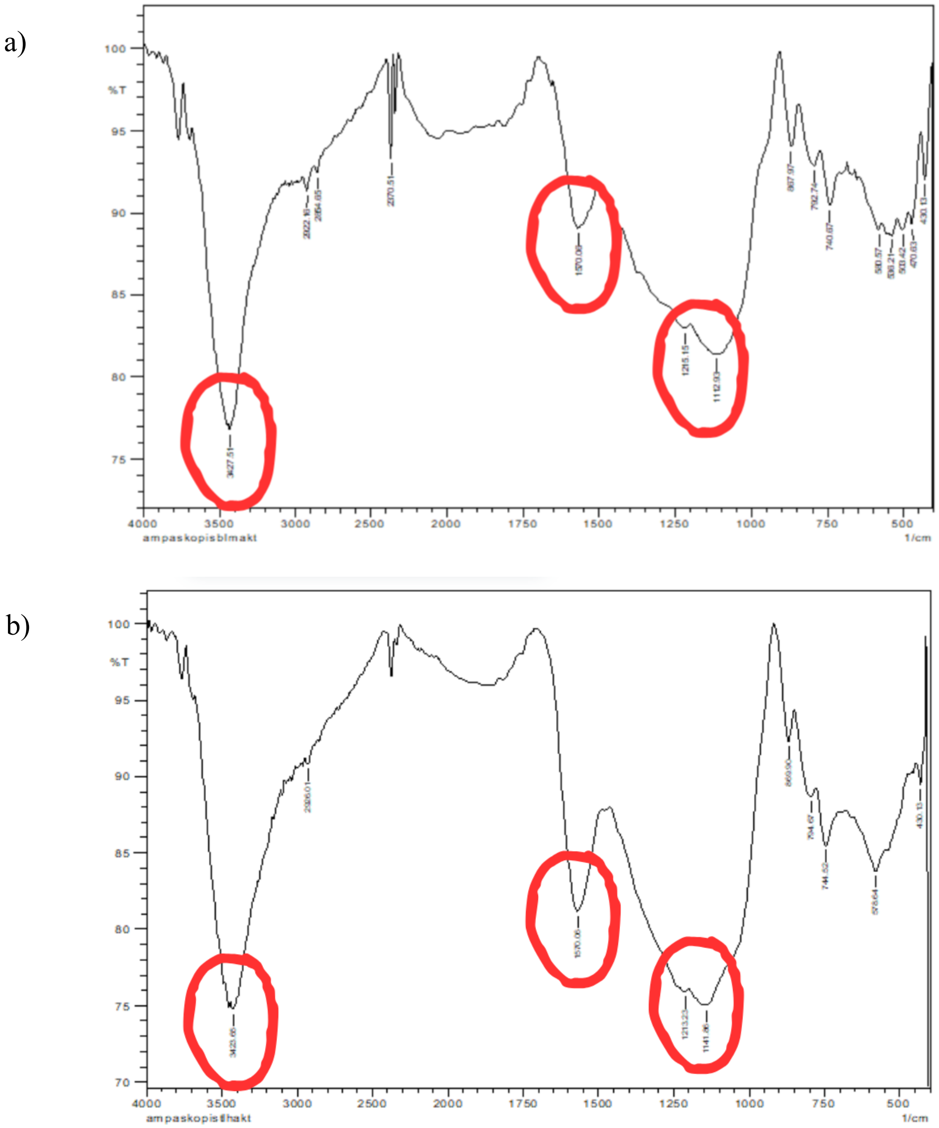

Figure 3. FTIR spectrum of Coffee-Ground Carbon: (a) raw char before H_3PO_4 activation; (b) Activated Carbon after H_3PO_4 Treatment

For the fly ash component, the FTIR spectrum at Figure 4 showed characteristic silicate features: a strong asymmetric Si–O–Si stretch around 1100 cm^{-1} and a Si–O bending mode near 460 cm^{-1}. These confirm the presence of silica and alumina species. Silanol (Si–OH) groups on fly ash can attract phosphate via hydrogen bonding or electrostatic attraction. Table 1 lists the main FTIR peaks and their assignments.

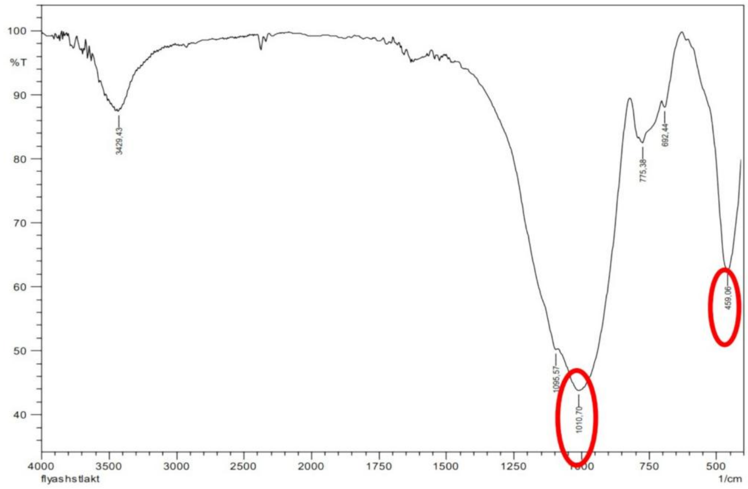

Figure 4. FTIR Spectrum of Fly Ash

Table 1. FTIR Peak Assignments for Coffee-Ground Carbon and Fly Ash

Sample	Wavenumber (cm^{-1})	Fuctional Group
Coffee carbon (before)	3427.5	O-H
	1570.1	C=C
	1112.9	C-O
Coffee carbon (after)	3423.7	O-H
	1570.1	C=C
	1141.9	C-O
Fly ash	1010.7	Si-O-Si
	459.1	Si-O

The activated carbon derived from H_3PO_4-activation of spent coffee grounds bears abundant oxygen-containing surface groups [12]. These O–H and C–O functionalities can interact with phosphate anions via hydrogen bonding and inner-sphere ligand exchange. In particular, surface hydroxyls can form hydrogen bonds with P–O oxygens of phosphate, and deprotonated –OH (or carbonyl) sites can coordinate phosphate (ligand exchange) [13]. Silanol groups can participate in phosphate binding by ligand exchange. Phosphate oxyanions can displace the proton of Si-OH and forming covalent Si-O-P bonds. Thus, the Si-O or Si-O-Si framework in fly ash provides additional binding modes as inner-sphere complexation for phosphate.

3.3. Phosphate Adsorption Performance

Batch adsorption experiments demonstrated that phosphate removal improved with longer contact time for all adsorbent mixtures. As shown in Figure 3, the lowest equilibrium phosphate concentration (≈5.82 mg/L) was achieved with the 1:4 coffee:fly ash mixture at 75 minutes, whereas the highest residual phosphate occurred with the 4:1 mixture at only 15. This behavior indicates that the adsorption process requires sufficient time to approach equilibrium, consistent with other studies reporting significant improvements in removal at extended contact times [14]. In addition, mixtures containing a higher fraction of fly ash consistently outperformed those richer in coffee carbon. In our tests the 1:4 mixture (20% coffee carbon, 80% fly ash) yielded the greatest reduction, suggesting that the fly ash enhances overall uptake. This synergistic effect agrees with literature reports that combining coffee carbon with fly ash increases porosity and adsorption sites, thereby boosting removal efficiency [15].

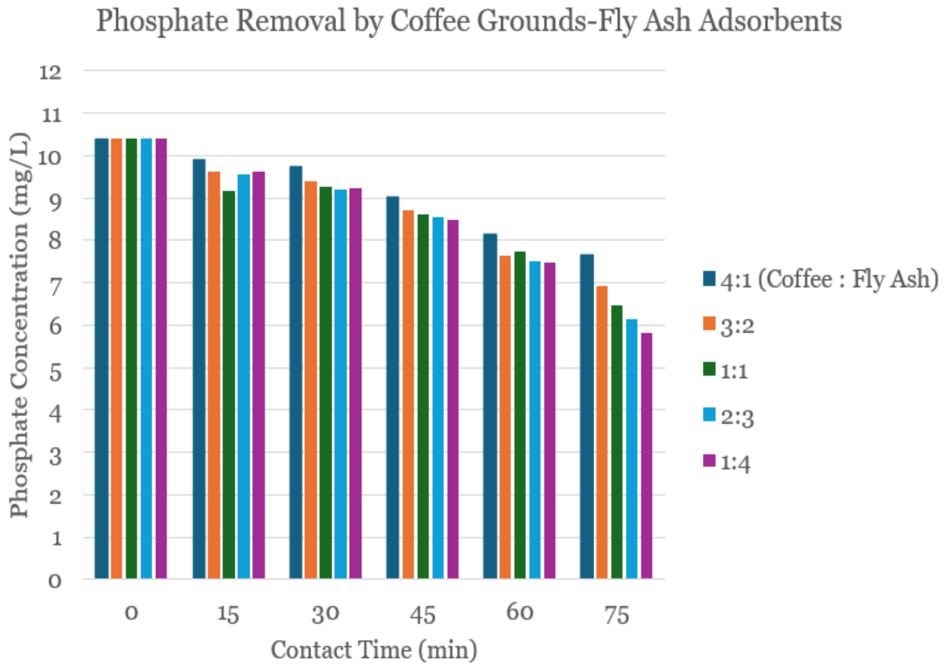

Figure 5. Phosphate Concentration in Synthetic Laundry Effluent versus Contact Time at Varying Coffee Grounds to Fly Ash Mass Ratios

4. CONCLUSION

The phosphoric acid activated carbon composite prepared from spent coffee grounds and fly ash at a 1:4 mass ratio with 75 minutes contact time achieved a 44% phosphate removal, lowering concentration from 10.4 mg/L phosphate to 5.82 mg/L phosphate and moving substantially toward the Indonesian discharge limit of 2 mg/L phosphate as stipulated in Ministerial Regulation No.5/2014 on Wastewater Quality Standards. While full compliance was not attained, the marked reduction confirms the composite's efficacy and underscores the need for further treatment.

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