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Utilization of Spent Coffee Grounds as Adsorbent: A Review

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Abstract. Spent coffee grounds are by-products of coffee production. Spent coffee grounds have the potential to be used as activated carbon adsorbents because they have a high carbon content and are lignocellulosic materials. The adsorption capacity of coffee ground activated carbon (CGAC) is affected by the activation method and the environmental conditions during the adsorption process. This review used databases from ScienceDirect, Springer, and Google Scholar, which were screened using keywords related to adsorption using activated carbon from spent coffee grounds. Based on the collected information, physicochemical activation produces activated carbon with a higher surface area and adsorption capacity compared to individual activation. The performance of CGAC showed a good result in adsorbing various adsorbates. The Langmuir isotherm model and the pseudo-second-order kinetic model were the best models for describing the CGAC adsorption process.

Keywords: *activation; adsorbent; adsorption; spent coffee grounds.*

1 Introduction

Coffee is the second-most consumed beverage in the world after tea [1]. Indonesia is the world's fourth-largest coffee producer, with a total coffee production of 685,980 tons in 2020 and a total consumption of 288,000 tons. Many by-products are produced throughout the coffee production process, including spent coffee grounds, coffee silverskin, and coffee husks [2]. 1 ton of coffee beans yields approximately 650 kg of coffee grounds, with 0.91 grams of coffee grounds yielded for every 1 gram of ground coffee. Furthermore, it takes up to 2 kg of coffee grounds to produce 1 kilogram of coffee drink [3].

Spent coffee grounds should not be disposed of in the environment because they can cause various negative impacts on both the ecosystem and human health. Spent coffee grounds can cause harm to the environment because, when degraded, they need a lot of oxygen and contain a variety of toxic substances such as caffeine, tannins, and phenols [4]. Spent coffee ground leachate can be mutagenic, posing a risk to human health and the environment because these

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compounds can induce DNA damage and are hazardous to aquatic organisms [5]. Therefore, spent coffee grounds must be treated or recycled so that they do not contaminate the environment. One of the ways spent coffee grounds can be used is to transform them into activated carbon, which can be utilized as an adsorbent.

Activated carbon is a porous solid produced from carbon materials through heating at high temperatures [6]. Spent coffee grounds are suitable as a raw material to produce activated carbon. This is because spent coffee grounds have a high carbon content (> 50%) [7]. There have been many studies related to the use of coffee grounds as an adsorbent to remove organic compounds (pesticides, free fatty acids), dyes (methylene blue, methyl orange, rhodamine B), and metals (lead, iron, copper).

There are several methods to activate spent coffee grounds into activated carbon. The difference in different activation methods affects the adsorption performance, isotherm model, and kinetic model. Therefore, the objective of this study was to obtain information about spent coffee grounds as activated carbon, including utilization, activation method, isotherm model, and adsorption kinetic model.

2 Methodology

The method used in writing this article is a literature review, which provides information and data about the usage of coffee grounds as an adsorbent. National and international article searches were conducted through databases such as ScienceDirect, Springer, and Google Scholar. Keywords such as "adsorption using spent coffee grounds" or "adsorption using activated carbon from spent coffee grounds" were used to conduct article searches. Furthermore, articles were filtered by limiting the publishing year from 2013 to 2023. 124 articles with titles related to the topic were obtained in the early phases. Furthermore, screening was repeated based on inclusion and exclusion criteria as well as the availability of full access to the articles, yielding a total of 25 articles. The data from the paper will be used in this literature review.

3 Result and Discussion

3.1 Composition of Spent Coffee Grounds

Spent coffee grounds vary in composition based on the growing location, the type of coffee beans, the burning conditions, the brewing method, and drying method [8] [9]. Protein, pectin, cellulose, and carbon are the primary components of coffee grounds [10]. Table 1 shows the results of the study on the chemical composition of spent coffee grounds.

Table 1 Composition of Spent Coffee Grounds [11]

Chemical components	Composition (g/100 g dry material)
Cellulose (Glucose)	12.40±0.79
Hemicellulose	39.10±1.94
Arabinose	3.60±0.52
Mannose	19.07±0.85
Galactose	16.43±1.66
Xylose	-
Lignin	23.90±1.70
Fat	2.29±0.30
Protein	17.44±0.10
Nitrogen	2.79±0.10
Carbon/nitrogen (C/N ratio)	16.91±0.10
Total dietary fiber	60.46±2.19

According to Table 1, coffee grounds are a material with a high lignocellulosic component (cellulose, hemicellulose, and lignin). Materials that have a high lignocellulosic component content have great potential to be used as adsorbents [11]. Carbon is created when coffee beans are roasted or heated, making brewed coffee grounds suitable to be used as raw material to produce activated carbon [12].

3.2 Activation of Spent Coffee Grounds

Spent coffee grounds cannot be utilized directly as an adsorbent because the pore size is still too small and there are non-carbon chemicals, resulting in low adsorption capacity. Carbonization and activation are needed before using spent coffee grounds as an adsorbent. Carbonization aims to obtain pores in the carbon structure, whereas activation aims to build new pores and increase the surface area of activated carbon [6]. Furthermore, the activation improves characteristics such as contact surfaces, pore distribution, and surface chemistry on spent coffee grounds [12].

The activation process of spent coffee grounds is one of the elements that affect adsorption capacity [13], therefore choosing the proper activator is critical for the adsorption process to function optimally. There are two types of activation processes that can be used, physical activation and chemical activation. Physical activation is carried out by heating the material to be activated at very high temperatures in an inert atmosphere. If the temperature is too low during this process, the pores in the material are not entirely opened, and the organic compounds present in the material are not completely degraded and volatilized,

resulting in a reduction of pore area [14]. Meanwhile, chemical activation uses activator agents such as NaOH, KOH, HCl, ZnCl2, and H3PO4. Chemical activation can produce activated carbon with a high specific surface area (>2000 m2/g) and a high pore volume (> 1.0 cm3/g) [15]. Chemical activation is the most frequently used method for activating carbon from spent coffee grounds [12].

Chiang, et al in [15] studied the influence of activation temperature and chemical activators on the specific surface area (SSA) and pore volume of coffee ground activated carbon (CGAC). It is activated at a temperature range of 550–700 ℃ on activated carbon that has not been chemically activated. It was found that optimum heating produced the largest SSA and pore volume compared to other activation temperatures, but these results were still far below the standard for activated carbon. Generally, SSA values were >800 m2/g and total pore volume was >0.8 cm3/g. At 700°C, the porosity of CGAC decreases due to the morphological deconstruction of CGAC into ash. Activation with the addition of a chemical activator in the form of HCl is carried out at temperatures ranging from 700 to 950°C. It was found that the higher the activation temperature, the more porous the structure and SSA. After activation with HCl at 950°C, an SSA value of 2407 m2/g and a total pore volume of 1.281 cm3/g were achieved.

Jutakridsada, et al in [16] studied the effects of activator concentration and impregnation duration. The activator used was ZnCl2 in concentrations ranging from 5% to 15% by weight and impregnation times ranging from 8 to 24 hours. It was discovered that soaking coffee grounds at a concentration of 15% for 24 hours produced the highest SSA and total pore volume, with values of 821 m2/g and 0.44 cm3/g, respectively. High concentrations will trigger a stronger chemical reaction on the surface of the coffee grounds, resulting in more microporosity on the surface of the coffee grounds. Furthermore, the longer the impregnation time, the longer the activator is in contact with the coffee grounds.

Based on the summary data in Table 2, activated carbon produced through the combination of physical and chemical activation typically has a higher surface area and total pore volume compared to individual chemical or physical activation alone. This is in accordance with the statement of Tani, et al in [17], where the combined method will produce activated carbon with a larger surface area and high adsorption capacity. Combination method or physicochemical activation can provide a wide range of pore sizes, resulting in a higher overall surface area available for adsorption. Physical activation will form a porous structure, while chemical activation removes impurities in pores of activation carbon. The combination of these two processes enhances the adsorbate's accessibility to the available surface area, maximizing adsorption capacity.

Table 2 Summary of several activation methods of spent coffee grounds

Activator	Surface area (m^2/g)	Total pore volume (cm^3/g)	Adsorbate	Reference
N_2, CO_2, 950°C HCl	2407	1,281	Methyl orange	[15]
NaOH	-	-	Methylene blue	[18]
Thermostatic chamber 60°C	451	0,27	Tetracycline	[19]
N_2, 500°C, H_3PO_4 (50 wt%)	2118	0.182	Landfill leachate	[20]
$ZnCl_2$ (15 wt%), HCl (5 wt%)	821	0,44	Cu (II)	[16]
H_3PO_4	435,98	0,295	Phenol	[14]
NaOH, HCl	-	-	Rhodamine B	[21]
KOH	-	-	Aniline yellow dye	[22]
N_2, 800°C KOH-urea	1665,45	0,59	Methylene blue	[23]

3.3 Utilization of Spent Coffee Grounds

The adsorption capacity is strongly influenced by the chemical structure of the adsorbent surface, which affects surface characteristics such as polarity, acidity, and hydrophobicity [24]. Furthermore, the properties of activated carbon, type of activator, pH, temperature, and ionic strength all affect adsorption capacity [13]. According to Kusuma, et al in [25], the procedure of extracting oil from coffee grounds affects the quality and quantity of activated carbon generated. The oil extraction technique can improve absorption, decrease ash and volatile matter content, and boost the carbon content of activated carbon generated from coffee grounds.

Sukmawati and Sunarto in [26] did research on the effect of adsorbent dosage and contact time on the removal of free fatty acids (FFA) and peroxide in used cooking oil. The adsorbent dose varied by 1, 2, 3 grams, while the contact time was varied by 1, 2, 3 hours. According to the result of this study, the highest removal rate of FFA and peroxide occurred at the 3 gram adsorbent dose and 3 hours of contact time, with removal rates of 50.1113% and 49.9749%, respectively. The more adsorbent, the larger the surface area of the adsorbent that can adsorb free fatty acids and peroxide molecules. The adsorption process will

run rapidly at the initial contact time because there are still many available sites on the adsorbent, but as time passes, the adsorption rate will drop because the sites on the accessible adsorbent will start to decrease.

Lafi, et al in [27] studied the effects of temperature on the adsorption of toluidine blue (TB) and crystal violet (CV). Temperature was discovered to influence the amount of adsorbate adsorbed. The dye removal rate was 96% at 20°C, while when the adsorption temperature was 50°C, the dye removal rate dropped to 91%. This shows that dye removal is more effective at low temperatures or that the adsorption process is exothermic.

Rattanapan, et al in [28] studied the effect of pH on the methyl orange adsorption process using CGAC. The optimum pH results can be described using pHzpc, which is the pH value when the adsorbent's surface charge is zero. pHzpc is calculated by graphing the beginning pH against the final pH and obtaining the point of intersection as the pHzpc value [29]. The pHzpc value of CGAC is 8, which means that when the pH is less than 8, the surface of the CGAC is positively charged, and when the pH is greater than 8, the surface is negatively charged. After that, the influence of pH on the adsorption process was studied by adjusting pH 3–12 for 120 minutes with time control environments, an adsorbent dose of 0.1 g, a temperature of 30°C, and an initial concentration of 300 mg/L methyl orange. The optimal pH environment for methyl orange adsorption was pH 3. This condition is described by the pHzpc CGAC value, which states that at pH 3, the surface will be positively charged, allowing it to bind with methyl orange ions. According to the data, the adsorption capability of CGAC decreases as pH increases. It's possible due to weak electrostatic interactions.

Table 3 Application of CGAC as an adsorbent

Adsorbate	Adsorption condition	Adsorption capacity (mg/g)	Removal rate	Reference
Methylene blue (MB)	Dosage 0,1 g, initial concentration 500 mg/L, 400 rpm, 30 minute	142.8	-	[18]
Tetracycline	Dosage 0,6 g/L, pH 6,5, 25°C, 450 rpm	123.46	-	[19]
Landfill leachate (COD)	Dosage 20 g/L, pH 4, 20°C, 125 rpm, 3 hour	40	>90%	[20]
Ag+	Dosage 0,1 g, initial concentration 50 mg/L, pH 6, 1 hour	49,543	92,4%	[30]
Cooper	Dosage 1 g, initial concentration 10 mg/l, 300	2,46	85%	[31]

	rpm, 3 hours			
Aniline yellow dye	Dosage 0,6 g, initial concentration 35 ppm, 2,5 hours	2,58	88,72 ± 1,88%	[22]
Rhodamine B	Dosage 0,2 g, pH 7, 45 minutes	4,43	-	[32]
Methyl orange	Dosage 0,05 g, initial concentration 300 mg/L, pH 3, 90 minutes, 30℃	658	-	[28]
Ammonia	Dosage 2 g, 300 rpm, 30 minute	-	84,64%	[33]
FFA			50,1113%	
Peroxide number	Dosage 3 g, 3 hours	-	49,9749%	[26]

3.4 Adsorption Isotherm Models

The adsorption isotherm described is related to the equilibrium of the adsorbent at a constant temperature. The isotherm model is affected by the adsorbate, adsorbent, fluid physical characteristics, ionic strength, and temperature used during the adsorption process [34]. It is critical to understand the adsorption mechanism and the adsorption isotherm model in order to determine the interaction between the adsorbate and the adsorbent surface [12]. There are various adsorption isotherm models such as Freundlich, Langmuir, Temkin, Gibbs, Redlich-Peterson, Dubinin-Radushkevich, and others. The Langmuir and Freundlich isotherms are the most frequently used models in determining adsorption equilibrium [35].

In the Langmuir isotherm model, it is assumed that the adsorption process is homogeneous and monolayer without any interactions between adjacent adsorbed molecules when occupying a site [36]. Meanwhile, the Freundlich isotherm model shows that the adsorption process is reversible and not ideal, with the adsorbent's surface being heterogeneous and containing multiple types of active sites [37]. According to Table 4, most of the research findings indicate that the

Langmuir isotherm model is best suited for characterizing the adsorption process utilizing CGAC. This shows that the CGAC adsorption process takes place in a monolayer on the adsorbent's surface. However, apart from using the Langmuir isotherm model, the research conducted by Jutakridsada, et al in [16], Rattanapan, et al in [28], and Jr, et al in [22] explained that the adsorption process carried out is suitable for using the Freundlich isotherm model.

In a study conducted by Ovando-Medina, et al in [21], adsorption of rhodamine B using CGAC adsorbents is suitable for using the Redlich-Peterson isotherm model. The Redlich-Peterson isotherm model is a three-parameter isotherm model. This model combines the Freundlich and Langmuir equations, resulting

in a mixed adsorption mechanism and non-ideal monolayer adsorption that may be applied to homogeneous or heterogeneous systems [38].

Cuccarese, et al in [18] found that variations in pH might modify the isotherm model, which is useful for explaining the ongoing adsorption process. The Langmuir model is appropriate at pH 6, while the Temkin isotherm is appropriate at pH 7.5. The Temkin isotherm model assumes a multilayer adsorption process, with the heat of adsorption on all molecules in the layer decreasing linearly as surface coverage rises [37] [39]. The change in the isotherm model from Temkin to Langmuir as the pH decreases due to an increase in H+ ions at pH 6 leads the adsorption process to only occur in a monolayer because electrostatic interactions decrease, and the surface becomes more homogenous.

3.5 Adsorption Kinetic Models

Adsorption kinetics illustrates the absorption of a substance (adsorbate) by an adsorbent over time. The adsorption rate can indicate the characteristics of the absorption ability of the adsorbent on the adsorbate [40]. The adsorption process is divided into three stages: mass transfer of the adsorbate from the bulk solution to the adsorbent's external surface; diffusion of the adsorbate into the pore or adsorbent site; and reaction between the adsorbate and the adsorbent's surface so that the adsorbate adheres to the adsorbent's surface [41]. Several kinetic models, including pseudo-first order, pseudo-second order, Weber-Morris, and others, were used to test the adsorption process.

The pseudo-first-order kinetic model assumes that adsorption occurs only at local sites and does not involve interactions between adsorbed ions; the adsorption energy is independent of surface coverage; the maximum adsorption corresponds to a monolayer on the adsorbent surface; and the adsorption process follows the first-order rate equation. For the pseudo-second order (PSO), the assumptions are the same as for the pseudo-first order, with the exception that PSO follows the second-order rate equation [42]. According to the PSO kinetics model, the adsorption process that occurs is chemical adsorption, in which covalent bonds are established between the adsorbate molecules and the atoms on the adsorbent's surface [43]. Based on Table 4, the pseudo-second order kinetic model is the most suitable model for adsorption data using CGAC.

Table 4 Isotherm and kinetic models of adsorption using CGAC.

Adsorbate	Model isotherm	Kinetika Adsorpsi	Referensi
Methylene blue (MB)	Langmuir	Pseudo-second order	[15]
Methyl orange (MO)	Langmuir	Pseudo-second order	
Methylene blue (MB)	Temkin (pH 7,5) Langmuir (pH 6)	Pseudo-second order	[18]

Tetracycline	Langmuir	Pseudo-second order	[19]
Landfill leachate	Langmuir	-	[20]
Lactide acid	Langmuir	-	[12]
Ag^{+}	Langmuir	Pseudo-second order	[30]
Cu(II)	Freundlich	-	[16]
Timbal	Langmuir	-	[31]
Phenol	-	Weber-morris	[14]
Rhodamine B	Redlich-peterson	-	[21]
Aniline yellow dye	Freundlich	Pseudo-first order	[22]
Methyl orange	Freundlich	-	[28]
Methylene blue	Langmuir	Pseudo-second order	[23]

3.6 Regeneration of CGAC

The adsorbent can absorb the adsorbate on its own. When an adsorbent's capacity is surpassed, it becomes saturated, and the adsorption process stops since the surface of the adsorbent on all sites has absorbed the adsorbate. As a result, when the adsorbent becomes saturated, it must be replaced or regenerated. Regeneration can be carried out by the desorption process, which involves contacting the adsorbent with the desorption agent solution. The desorption agents used can be acidic, alkaline, or neutral [44].

Sukhbaatar, et al in [23] studied CGAC regeneration/recovery. The initial procedure was to centrifuge the MB adsorption sample, wash it with water, and dry it at 105°C for 6 hours. The samples were then agitated for 2 hours in a 70% ethanol solution before being centrifuged, dried, and annealed at 800°C. It has been discovered that CGAC can be retrieved up to five times. The adsorption capacity of the adsorbent after five times of recovery is 96.1% of the initial adsorption capacity.

4 Conclusion

According to studies, activated carbon generated from coffee grounds can be utilized as an adsorbent to adsorb various adsorbents. Various activation methods can be used to optimize the surface area and pore volume of activated carbon from coffee grounds (CGAC). It was found that physicochemical activation produces activated carbon with a higher surface area and adsorption capacity compared to individual activation. The adsorption capacity of CGAC varies with the type of activator as well as adsorption parameters such as pH, temperature, agitation speed, initial concentration, and contact time. It is critical to conduct a test first to determine the optimum conditions for the adsorption process. According to the results of multiple studies, the Langmuir isotherm model and

the pseudo-second order kinetic model are the best models for describing the CGAC adsorption process.

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