

BIOADSORPTION OF HEAVY METAL POLLUTANTS IN ASA RIVER USING TREATED AND UNTREATED RICE HUSK.

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Abstract:

The main source of environmental degradation is the increased production of heavy metals as a result of growing industrialization, which contaminates rivers' inflow through the industries' discharges of the effluents they produce as a result of their operations.

Limiting the effects and level of these heavy metals present in the effluent on the ecosystem is essential to make the Asa River usable for other purposes and to decrease their influence on those who depend on this water directly or indirectly. This study focused on the adsorption of these heavy metals using both treated and untreated rice husk, with the mass (g) of carbonized/activated rice husks (CRH/ACRH) at 0.032, 0.066, and 0.1g at 40ml of each of the stock solutions of the metals and the effluent at various time intervals of 24, 48, and 72 hours. The adsorbent was also analyzed for Fourier-transform infrared analysis (FTIR), Scanning electron microscopy (SEM) and Energy dispersive X-ray (EDX) before adsorption process.

The adsorption process was evaluated by using Atomic Absorption Spectrophotometer and compared with the standard of WHO (2006) and USEPA(2010). Based on the results of this study, it can be concluded that the ACRH was better compared to the CRH for the removal of the metals (Cd and Cr) from the samples that were gathered because the treated or activated carbon rice husk attracted more attention than carbonized or untreated rice husk due to its comparably higher adsorption capacity, which is favored by a higher number of active binding sites.

Key words: Heavy Metals, Bioadsorption Process, Carbonized Rice husk (CRH), Activated Rice husk (ACRH),

Asa River.

Introduction

Each developing country's growth and development is significantly influenced by industrialization as reported by Gandhi *et al.*, 2013, Safauldeen, 2019 which can be attributed to the continuous discharge of organic and inorganic waste products into natural environments having negative consequences on the environment by causing serious threat to natural habitat and human health, either directly or indirectly (Petrovic, 2011, Basigilini *et al.*, 2018). In recent times, the discharge of industrial wastewaters, which contains heavy metals and hazardous wastes, is a substantial contributor and most common source to water pollution as reported by Salami, 2003, Ogendegbe and Akinbile, 2004. Because of their toxicity, persistence, propensity to accumulate in organisms, food chain amplification, and non-degradability, heavy metals are the most hazardous to human health as reported by Lenntech, 2004.

Due to their persistence presence in the environment and known possibility of having detrimental impacts on human health, heavy metals with negative effects on health pose obvious concerns (Mahipal *et al.*, 2016). Acute heavy metal intoxications may damage central nervous function, the cardiovascular and gastro intestinal (GI) systems, lungs, kidneys, liver, endocrine glands, and bones while the chronic exposure may lead to death as reported by Rajeev Kumar *et al.*, 2014.

Rivers that receive the industrial effluents are unable to provide the dilution required for the continuance of their existence as trusted and reliable water sources due to the quantity of these pollutants (Ahamaku and Animashaun, 2013). The issue has sparked widespread concern over the effects on public health associated with the deterioration of waterways as a result of these pollution from industies (Adekunle and Eniola, 2008, Kimani, 2007). According to research, poor drinking water quality is directly responsible for 80% of all diseases that take lives in third-world nations (Jeffie, 2008). Every year, waterborne infections connected to a lack of adequate drinking water or sanitation cause more than six million children to pass away (about 20,000 each day) as reported by TWAS, 2002. The deterioration of the physical and chemical properties of water is frequently gradual and difficult to discover due to the water system and the ability to adapt to the changes before there is an obvious change in the water (Zahraa *et al.*, 2012). Therefore, it has become crucial to lower the concentration of heavy metals in Asa River using an adsorption methods by use of rice husk as an adsorbent in order to prevent it from more pollution and ensure that the water is readily available for use in domestic and agricultural purposes for people living around the community.

The Asa River, which supplies the majority of the water utilized in Ilorin and its surroundings, is strategically significant to the city's residents as once reported by Eletta, 2004, Eletta and Adekola,2005, Eletta, 2007, Ogundiran and Fawole, 2014.

However, some industries situated near the River in Ilorin discharge their waste waters into this river and they pose a major risk to both human health and the aquatic ecosystem as reported by Dimple Lakherwal, 2014 (along the River's bank, to name a few, there is a prominently visible steel and wire production sector and a pharmaceutical company) and Asa Dam, located somewhere along the River's course, treating the water by using convectional techniques before releasing it to users (Dimple Lakherwal, 2014).

Convectional treatment methods of wastewater such as ion exchange, adsorption, reverse osmosis, chemical precipitation and electrochemical treatment are used for the removal of heavy metals, organic and inorganic contaminants (Akpor and Muchie, 2010, Singh *et al.*, 2012, Mohiyaden *et al.*, 2016). However, high energy requirement, carbon emission, excess sludge discharge and high maintenance cost poses serious challenge to these convectional methods of wastewater treatment (Hauwa and Gasim,2020, Kumar *et al.*, 2013). Therefore, sustainable management of clean water and aquatic ecosystem requires eco-friendly and low cost adsorption method (Anand *et al.*, 2017).

Adsorption which is the most efficient of these methods because the others have inherent flaws such producing a lot of sludge, being inefficient, requiring delicate operating conditions, and costing a lot of money to dispose of. According to reports, adsorption has become a heavy metal removal technology that is effective, reasonably priced, and admirably easy to use (Geethamani *et al.*, 2014).

Commercial adsorbents and bio-sorbents, both of which have a high removal capability, are used to remove heavy metals from wastewater. Since the adsorption of heavy metal ions onto activated carbon is a widely used but expensive technique, it is necessary to identify low-cost alternatives that can adhere to the surface of heavy metals in order to remove the metals to be employed as adsorbents. Agricultural waste, aquatic plants, natural materials, etc. are examples of such adsorbents. A variety of agricultural waste products, sea weeds, and other microbial biomass have allegedly been used as bio-adsorbents to remove heavy metals (Sudha and Abraham, 2003).

Rice husks can be utilized as an adsorbent to lessen environmental pollution and make the adsorbed water useful for domestic and agricultural uses because they are agricultural wastes that are thrown into the environment in significant amounts. The purpose of the study was to ascertain how the heavy metal concentrations and some physical and chemical properties of River Asa varied. It also aimed to use rice husk as a bioadsorbent which is of low cost to treat the water rather than convectional methods of using expensive chemicals in order to lower the concentration of heavy metals to a useful level that would serve as a portable water supply for the entire community.

MATERIALS AND METHODS

Study site

The study was carried out on the Asa River in the Nigerian city of Ilorin, Kwara State, where water samples were collected, evaluated, and agricultural wastes were collected from Oko town in Oyo State. Upper, Middle, and Lower Streams generated the samples that were used. Both carbonized and activated carbonized rice husk were used in the adsorption processes.

Sampling Procedure and Sample Analysis

Three sampling stations, each at a different distance from the source of effluent discharged from the industries located along the river, were used to collect water samples in triplicate. As shown in plate 1, rice husks were gathered from Oko Town in Oyo State, washed repeatedly with de-ionized water to remove all dirt in its original particle size and being filtered and dried at 100°C. The cleaned and dried rice husk was oven dried at 500°C for 3 hours without any other further treatment to produce what is called Carbonized Rice Husks (CRH) as shown in plate 2 according to Nhapi *et al.*,2011. For two hours at 20°C, 100g of carbonized rice husks were soaked in 0.6 M of citric acid. Under aerobic conditions, acid husk slurry was dried overnight at 50°C, and the dried husks were then heated to 120°C. Repeated washing with distilled water (200ml/g) were performed on the reacted product. The cleaned rice husk was then dried in an oven at 100°C for a whole night to produce what is called activated carbonized rice husks (ACRH).

The test solutions was prepared by diluting of stock solution containing 1000mg/l of Cu, Cd, Pb, Zn and Fe to the desired concentrations. Each test solution's pH was brought to the desired level by being diluted with HCL and concentrated with NaOH solution before adding the adsorbent. A pH meter, type Hanna 211, was used for all pH readings.

A specified amount of rice husk was added to each 500 ml beaker containing a heavy metals solution of Pb, Cd, Cu, and Zn at a pH of 7±0.2. Rice husks that had been carbonized or activated had a mass (g) of 0.032, 0.066, and 0.1. Once equilibrium had been reached, the flasks were shaken continuously for 250 rpm. The research was based on the assumption that the applied shaking speed allows all of the surface area to interact with heavy metal ions. To be indicative of environmental circumstances, the investigation was conducted at a constant temperature of 25 °C. The average value was used for, after each experiment was carried out in duplicate. The bottles were capped and maintained closed during the experiment to prevent pH fluctuations caused by the exchange of gases.

The carbonized rice husk samples were subjected to scanning electron microscopy (SEM) analysis utilizing a JEOL JSM-6390model scanning electron microscope paired with an energy dispersive X-ray (EDX) analyzer to examine the surface morphology and elemental composition of the initial materials. SEM provides a high resolution, enabling greater magnification of materials having tiny openings. Diffuse Fourier-transform infrared analysis (FTIR) was used to qualitatively identify the functional groups in the carbonaceous components of the adsorbates in frequency range between 4000 to 400 cm-1 spectrometer (Thermo Nicolet, NEXUS, USA) at room temperature according to Mohamed and Amaal, 2017, Olakunle et al., 2017, Bello et al., 2017.

RESULTS AND DISCUSSION

Water samples from US, MS and LS were analyzed and were collected in triplicate.

Analysis on the adsorbent

The FTIR absorption spectrum of carbonized rice husk showed distinct strong peak at 3437.00 with functional group N-H stretching vibrations in amines and amides, a distinct strong peak at 2919.00 - 2848.19 with the functional group C-H stretching vibrations having CH₃, CH₂, CH generally in the region was observed, other peaks were shown at 2356.62, 1712.00, 1604.00, 1444.00, 1373.14, the peak at 1034.00 refers to the stretching vibrations of the O-H-O bond, where there is characteristics band between silica and oxygen as also reported by Simon et al., 2019 and Pallavi et al., 2012.

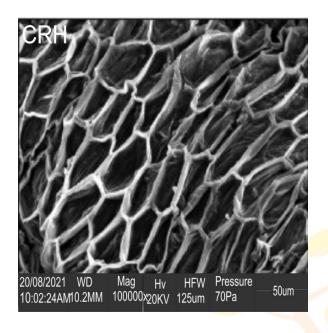
The SEM micrographs showed that many residual pores are distributed within the sample of carbonized rice husk, indicating that it is a highly porous material with large internal specific surface area. The rice husk might have become broken up during thermal decomposition of organic matter, thus obtaining highly porous structure. The EDX micrograph shows that Silicon has the highest percentage of 74.35% followed by Oxygen which has 5.40% and other elements. Naeem and Batool, 2021 revealed that Rice husk has binding sites for the bioremediation of Chromium. The presence of silicon in high percentage is responsible for the reduction and removal of chromium. Adsorption process

Carbonized rice husk and activated rice husk as a bio-sorbent was efficient for the removal of heavy metals in the collected water samples at varying concentration (0.032g, 0.066g and 0.100g) and contact time (24hrs, 48hrs and 72hrs). The highest adsorption efficiency of carbonized rice husk in the removal of Iron was obtained in US at 0.1g (24hrs) with 0.397mg/l, US at 0.066g (48hrs), US at 0.032g (72hrs) of 0.411mg/l and 0.0321mg/l respectively as shown in Table 1 and in activated rice husk it was obtained in US at all contact time and varying concentrations. In sample LS, there was an increase in concentration of Copper after it was treated with carbonized bio-sorbent (Table 2) and reduces in all varying concentrations and time contact in activated bio- sorbent, the adsorption efficiency of the bio-sorbent was fluctuating in all the concentration and contact time (Table 3) for both. As shown in Table 4. there was a complete removal of chromium metal ions in the water samples treated with activated carbonized rice husk with exceptions in US at 24hrs in which the concentration of chromium was 0.0702mg/l and 0.107mg/l for 0.066g and 0.100g of the bio-sorbent respectively. Table 5 shows a total reduction in the concentration of Cadmium at varying concentration and contact time for both activated and carbonized rice husk.





Plate 1a: The plate showing rice husk 1b: the carbonized rice husk



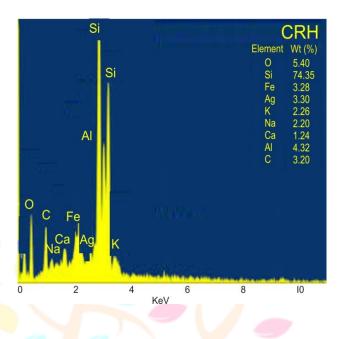


Plate 2a: SEM of Carbonized Rice Husk

Plate 2b: EDX of carbonized rice husk

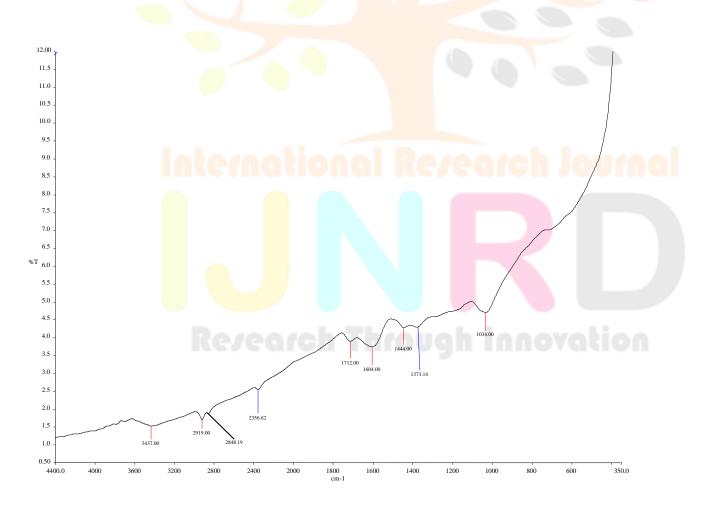


Figure 1: FT-IR result of Carbonized Rice Husk

	CR	КН			AC	RH	STD
IRON (Fe)	LS	MS	US	LS	MS	US	WHO, USEPA
							(2006) (2010)
Initial conc. (mg/l)	35.660	10.764	2.001	35.660	10.764	2.001	0.300
0.032g (24hrs)	7.603	9.146	0.421	10.146	8.156	1.483	
0.066g (24hrs)	7.952	9.258	0.521	9.523	8.100	1.662	
0.1g (24hrs)	1.254	7.745	0 <mark>.39</mark> 7	5.2 <mark>1</mark> 5	6.417	1.545	
0.032g (48hrs)	1.605	6 <mark>.4</mark> 82	0.440	6. <mark>0</mark> 12	6.101	1.694	
0.066g(48hrs)	6.711	8.001	0.411	<mark>8.</mark> 482	6.217	1.401	
0.1g (48hrs)	7.254	7.534	0.466	7.501	4.615	1.545	
0.032g (7 <mark>2hrs</mark>)	9.903	8.041	0.321	7.543	5.143	1.476	
0.066g (72hrs)	5.056	<mark>9.6</mark> 87	0.463	<mark>5</mark> .132	7.012	1.397	
0.1g (72hrs)	8.259	7.765	0. <mark>392</mark>	<mark>4</mark> .143	6.177	1.456	

Table 1: Concentration of Iron present in the water samples treated with carbonized rice husk (CRH) and activated carbonized rice husk (ACRH).

Table 2: Concentration of Copper present in the water samples treated with carbonized rice husk and activated carbonized rice husk.

	CRH	ACRH	STD	
COPPER (Cu)	LS MS US	LS MS	US WHO, USEPA (2006) (2010)	
Initial conc. (mg/l)	0.015 0.062 0.057	0.015 0.062	0.057 0.01	
0.032g (24hrs)	0.066 0.051 0.056	0.015 0.051	0.056	
0.066g (24hrs)	0.109 0.046 0.067	0.005 0.045	0.040	
0.1g (24hrs)	0.079 0.037 0.476	0.004 0.033	0.034	
0.032g (48hrs)	0.049 0.05 0.037	0.008 0.033	0.034	
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0.066g(48hrs)	0.060	0.060	0.018	0.005	0.025 0.026
0.1g (48hrs)	0.051	0.054	0.041	0.003	0.030 0.024
0.032g (72hrs)	0.046	0.060	0.036	0.008	0.013 0.020
0.066g (72hrs)	0.049	0.070	0.039	0.003	0.004 0.012
0.1g (72hrs)	0.043	0.051	0.031	0.005	0.002 0.000

Table 3: Concentration of Zinc present in the water samples treated with carbonized rice husk and activated carbonized rice husk

	CRH	0	0		ACRH		STD
ZINC (Zn)	LS	MS	US	LS	MS U	JS	WHO (2006
Initial conc. (mg/l)	2.852	1.172	2.123	2.852	1.172	2.123	0.03
0.032g (24hrs)	0.476	0.788	0.2 <mark>33</mark>	1. <mark>35</mark> 1	0.812	0.310	
0.066g (24hrs)	0.459	0.797	0.265	1.214	0.714	0.264	
0.1g (24hrs)	0.543	0.689	0.294	1.320	0.801	0.2 <mark>4</mark> 1	
0.032g (48hrs)	0.550	0.576	0.158	0.512	0.512	0.211	
0.066g(48hrs)	0.411	0.843	0.549	0.415	0.413	0.117	
0.1g (48hrs)	0.444	0.647	0.179	0.301	0.402	0.104	
0.032g (72hrs)	0.475	0.899	0.169	0.112	2 0.214	0.105	
0.066g (72hr <mark>s)</mark>	0.501	<mark>0.949</mark>	0.266	0.21	3 0.112	0.071	
0.1g (72hrs)	0.511	<mark>0.844</mark>	0.171	0.003	3 0.010	0.002	

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Table 4: Concentration of Chromium present in the water samples treated with carbonized rice husk and activated carbon rice husk

	CRH		ACRH	[STD
CHROMIUM (Cr)	LS MS	US	LS M	IS US	USEPA 2010
Initial conc. (mg/l)	0.084 0.27	0.240	0.084 0.2	278 0.240	0.10
0.032g (24hrs)	0.000 0.00	0.000 0.000	0.000 0.0	000 0.000	
0.066g (24hrs)	0.000 0.00	00 0.072	0.000 0.	.000 0.000	
0.1g (24hrs)	0.000 0.00	0 0.107	0.000 0.	.000 0.000	
0.032g (48hrs)	0.00 0.00	000.0 00	0.0 <mark>0</mark> 0 0	0.000 0.000	
0.066g(48hrs)	0.00 0.00	000.0 00	0. <mark>0</mark> 00 0	0.000 0.000	
0.1g (48hrs)	0.000 0.00	000.0	0.000 0	0.000 0.00 <mark>0</mark>	
0.032g (72hrs)	0.00 0.00	000.0 00	<mark>0</mark> .000 0	0.000 0.000	
0.066g (72hrs)	0.000 0.00	000.000	0.000 0	0.000 0.000	
0.1g (72hrs)	0.00 0.00	000.0 00	0.0 <mark>00 0</mark>	0.000 0.000	

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Table 5: Concentration of Cadmium present in the water samples treated with carbonized rice husk and activated carbonized rice husk.

CI	RH		ACR	Н			STD
CADMIUM (Cd)	LS	MS	US	LS	MS	US	WHO
Initial conc. (mg/l)	0.020	0.022	0.021	0.020	0.022	0.021	0.003
0.032g (24hrs)	0.000	0.000	0.000	0.000	0.000	0.000	
0.066g (24hrs)	0.000	0.000	0.000	0.000	0.000	0.000	
0.1g (24hrs)	0.000	0.000	0.000	0.000	0.000	0.000	
0.032g (48hrs)	0.000	0.000	0.000	0.000	0.000	0.000	
0.066g(48hrs)	0.000	0.0 <mark>0</mark> 0	0.000	0.000	0.000	0.000	
0.1g (48hrs)	0.000	0.000	0.000	0.000	0.000	0.000	
0.032g (72hrs)	0.000	0.000	0.000	<mark>0.0</mark> 00	0.000	0.000	
0.066g (72hrs)	0.000	0.000	0.000	0.000	0.000	0.000	
0.1g (72hrs)	0.000	0.000	0.000	0.00	0 0.000	0.000	

DISCUSSION

Carbonized Rice Husk was efficient in the reduction of cadmium metal ions in all collected samples. At varying contact time (24hrs, 48hrs, 72hrs) and bio-sorbent dose (0.032g, 0.066g, 0.100g), the bio-sorbent was able to adsorb cadmium metal ions completely hence facilitating its complete removal. The total adsorption of cadmium could be due to the abundance of silica element in the rice husk. Samadder and Snehlata (2014) made use of both raw and processed rice husk for the remediation of heavy metals and their result showed that those heavy metals were greatly reduced due to the numerous binding site on rice husk.

From this research it was deduced that the ACRH was more effective than the CRH for the removal of the metals (Cd and Cr) from the samples collected because the treated or activated carbon rice husk attracted more attention than carbonized rice husk due to comparatively higher adsorption capacity favoured by higher number of active binding sites which improved the ion exchange properties and also activate and motivate the enhancement of

functional groups after the chemical treatment and can also be recommended for a large scale industrial applications after field work as reported by Samadder and Snehlata, 2014.

In conclusion, most heavy metals are carcinogenic in nature and can cause severe problem for humans and the aquatic ecosystem when consumed directly or indirectly. Thus, for the removal of these heavy metals from waters and rivers, carbonized rice husk has proven to be efficient and activated carbonized rice husk was more efficient. The presence of high percentage of silicon in rice husk has proof the responsibility of removing cadmium completely in all water samples used.

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