Original Research Article

Efficiency of Sawdust from selected African indigenous wood spp. as a low-cost adsorbent for removal of copper ion from contaminated water

Abstract

Aims: This study investigates the use of sawdust from 3 hardwood species as low-cost adsorbent for the removal of copper from contaminated water.

Study Design: The experimental design used for this study was 3 x 2 x 4 factorial experiment; the different sawdust species, two baselines (treated and untreated) and four levels of pH and time as factors were combined and used for the study

Methodology: Test was carried out to investigate the effect of sawdust pre-treatment on their adsorption capacity in the removal of Copper ions from contaminated water at different pH levels; the sawdust samples were sieved through a screen size of 850µm after which a portion of each species sawdust was subjected to pre-treatment by boiling while the other portions were maintained as control samples (untreated).

Results: The results shows that adsorption capacity for both treated and untreated samples were 69.75±13.78 %, 68.60±19.48 %, 69.34±23.08 %, 74.79±17.79 %, 74.52±22.30 % and 76.90±18.21 % for *Alstonia boonei*, *Erythrophleum suaveolens* and *Ficus mucuso* respectively.

Conclusion: The contact time and pH showed no significant difference between the treated and untreated samples. Sawdusts from the selected wood species are suitable to be used as adsorbent towards the removal of copper from contaminated water.

Keywords: Adsorption, *Alstonia boonei*, Adsorbent, Copper ions, Contaminated water

1.0 INTRODUCTION

The demand for forest and forest products keeps on increasing as the forest estate is drastically reducing with increasing population and civilization all over the world. The annual demand for wood as a construction material exceeds the quantity of round log extracted from the forest reserves and forest plantations per annum (Ogunwusi & Jolaoso, 2012). The increase in the demand of forest and its products in Nigeria has led to over-exploitation of the forest estate and thereby leading to the generation of excess wood waste, as the efficiency of lumber recovery in wood processing industries ranges between 40% and 50 %, which implies that 50 – 60 % of the log volume ends up as waste usually in the form of slabs, sawdust, edgings, and shavings (Fuwape, 1998). Nigeria generates about 1.8 million tons of sawdust annually and 5.2 million tons of wood wastes (Owoyemi *et al*., 2016). Sawmills in Nigeria alone generated over 1,000,000 m^3 of wood waste while plywood mills generated about 5, 000 m^3 of waste in 2010 (Ogunwusi, 2014). It has been reported that lignocellulosic waste materials such as that from wood serves as good adsorbent since they possess some adsorption capacities and they are readily available at a low cost (Estes,1998). Although, the lingocellulosic materials with different adsorption capacities for individual metal ions have previously been reported (Sciban & Klasnja, 2004). There is only limited literature explaining the mechanism of adsorption on sawdust in stormwater and wood wastes like sawdust is abundant, inexpensive and unused resources from agricultural byproduct, the use of this material would be beneficial to the environment: polluted streams would be cleaned and a new market would be opened for the sawdust. Sawdust from wood contains cellulose, lignin, and tannins or other phenolic compounds which are active ion exchange compounds. Therefore, the increasing need to treat metal contaminated waste water prior its discharge to the environment necessitates this research especially with the use of low-cost technologies in the removal of copper from contaminated water. The use of conventional treatment processes such as chemical precipitation, ion exchange, and electrochemical removal in removing heavy metal from inorganic effluent has been studied (Eccles, 1999). Although, these processes have significant disadvantages which includes, incomplete removal, high-energy requirements, and production of toxic sludge (Eccles, 1999). Presently, numerous approaches have been studied and recommended for the development of cheaper and more effective technologies that can decrease the amount of wastewater produced and also improve the quality of the treated effluent. Adsorption has become one of the alternative treatments in recent time, although, this approach has intensified the search for low-cost adsorbents that have metal-binding capacities (Leung *et al*., 2000). The adsorbents may be of mineral, organic or biological origin, zeolites, industrial by-products, agricultural wastes, biomass, and polymeric materials (Kurniawan *et al*., 2005). Several studies have been reported. The removal of Cu (II) and Pb (II) using iron slag has been investigated and a pH range from 3.5 to 8.5 for Cu (II) and from 5.2 to 8.5 for Pb (II) was optimized (Feng *et al*., 2004). Gupta *et al*. (2003) also explored the use of bagasse fly ash, a solid waste from sugar industry, for the removal of Cd(II) and Ni(II) from synthetic solution at pH ranging from 6.0 to 6.5. In addition, sawdust treated with 1, 5-disodium hydrogen phosphate was used for adsorption of Cr(VI) at pH 2 (Uysal & Ar, 2007), although, the sorption capacity of the sawdust is dependent on a number of factors, such as absorbent dose, size of the sawdust, contact time, temperature, pH, ionic strength of the aqueous solution involved (Shukla et al., 2002; Argun et al., 2007). Also, there has been renewed interest in the removal of heavy metals from industrial effluent with the use of agricultural by-products as adsorbents. The use of agricultural by-products in bioremediation of heavy metal ions, is known as bio-sorption and also sawdust has been reported as a good sorbent used for removing mostly toxic metals from wastewaters; although, it may release potentially hazardous substances as tannins & lignin, phenolic compounds, and resin acids which results in oxygen depletion in the recipient water bodies which can be overcome by effective measurement technologies and by addition of agents such as filter aids and coagulants (Hubbe *et al*., 2016). This study therefore focused on the removal of Copper from contaminated water using sawdust from 3 hardwood species.

2.0 MATERIALS AND METHODS

2.1 Experimental Site

The research was conducted at Sustainable laboratory services limited, km 4/5 Benin/Ilesha Expressway Akure, Forestry and Wood Technology Department Laboratory and Department of Industrial Chemistry Laboratory, Federal University of Technology, Akure (FUTA) Ondo State, Nigeria, FUTA lies between latitude 7.2971727^{o}E and longitude 5.1460648^{o}N respectively with an elevation of 359 m above the sea level. FUTA is located in Akure South Local Government Area of Ondo State in Nigeria.

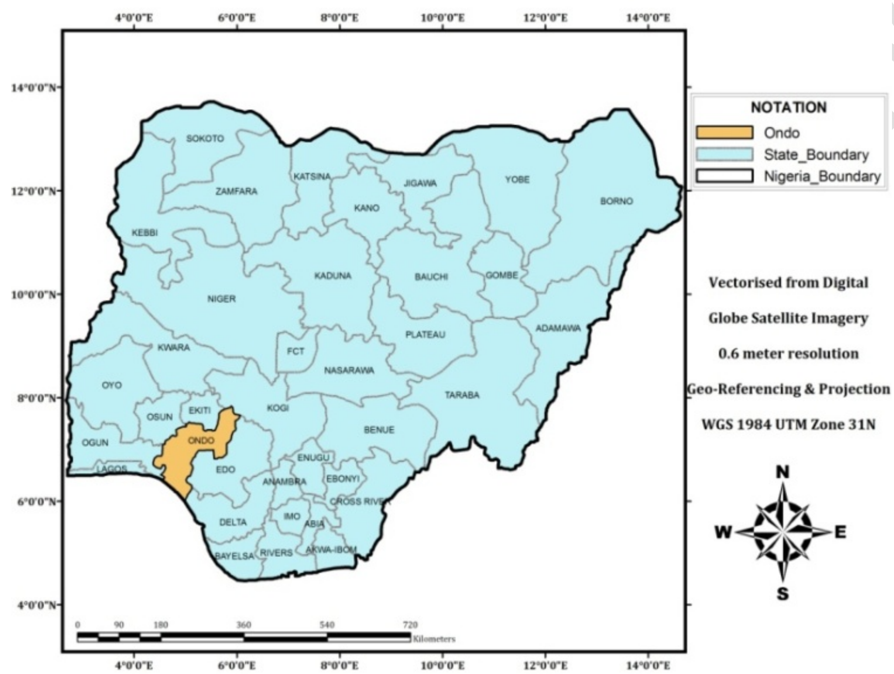

Figure 1: Map of Nigeria showing Akure and Ondo State (Study location). Adapted from Olamiju & Oyinloye (2015)

2.2 Collection of Raw Materials

The materials used for the study includes Metal chemical (copper), Hot plate, Sieve, pot, volumetric flask, conical flasks, sample bottle, filter paper, pH meter, atomic absorption spectrophotometer, beakers, sawdust from *Erythrophleum suaveolens* (Eru), *Alstonia boonei* (Ahun), and *Ficus mucuso* (Obobo). Sawdust from *Erythrophleum suaveolens* (Eru), *Alstonia boonei* (Ahun), and *Ficus mucuso* (Obobo) was obtained from a sawmill at Roadblock in Akure south LGA Ondo state, Nigeria. The different wood sawdust were carefully obtained by placing a polyethylene bag on the Band-saw machine to ensure the collection of sawdust was purely from the choice species. The chemicals and other materials used for this research were procured from PASCAL scientific limited in Akure South LGA, Ondo State, Nigeria.

2.3 Material Preparations

2.3.1 Preparation of Sawdust (Adsorbent)

The wood sawdust was processed in the workshop of the Department of Forestry and Wood Technology, Federal University of Technology, Akure. The sawdust material was first screened through a sieve of screen size 850 µm, to obtain uniformly sized particles used for the adsorption tests in this study, after which the sieved sawdust from each wood species was divided into two equal halves with each weighing 50 grams bringing the total samples to six (6). One half from each divided portion (i.e. 50 grams) of each species was boiled for one (1) hour, washed with clean water and boiled for another one (1) hour to effectively remove all the extractives, while the other halves of the divided portions from each species were maintained as untreated. The six (6) samples from the three (3) wood species (treated and untreated) were dried at 105^{0}C for 2 hours using an oven to remove moisture from the sawdust before adsorption studies.

2.3.2 Inducement of Copper (Cu^{2+}) into Water Solution

2.511 grams of the copper salt were weighed on an electric weighing balance and 2 liters of distilled water using a 2 liters standard measuring flask. The copper salt was dissolved in the distilled water to obtain a 50 ppm solution of copper (Cu^{2+}). The working solutions were obtained by serial dilution of these stock solutions with distilled water to the desired concentrations.

3.0 TESTING METHODS

3.1 Batch Adsorption Studies

Batch adsorption experiments were carried out by shaking 125 cm^3 conical flasks containing 0.2 g of *Erythrophleum suaveolens*, Alstonia boonei, and *Ficus mucuso* with 20 ml of the adsorbate solution (CuSO4.4H2O) under stirring condition within a stipulated time. After the time required for the reaction process had been attained, the mixtures were filtered using Whatman filter paper, and the supernatant analyzed using atomic absorption spectrophotometer (AAS). All experiments were conducted in triplicates and their values were used in data analysis.

3.2 pH of adsorbent

In this study, the effect of pH was investigated, a 50 ppm metal solution was prepared from the initial 1, 000 ppm solution by taking 50 ml from the solution and making up to 1000 ml in a volumetric flask. Six samples were obtained and separated into six beakers. Each of the six metal solutions was adjusted to initial pH 1, 2, 3 and 4 by using 0.1 M HCl and 0.1 M NaOH (the use of NaOH is to neutralize the solution into acceptable range), 0.2g of sawdust (adsorbent) was measured in six beakers and a part added to each of the six 50 ppm metal solutions adjusted to pH range of 1-6 already in six conical flasks. Each was mechanically stirred, kept for 360 mins and then filtered. The filtrate was collected and stored in a sample bottle. The final pH of the solution was measured with pH meter, and the metal concentration in each sample was measured using an atomic absorption spectrophotometer.

3.3 Contact Time

The 50 ppm metal solution was prepared from the initial 1,000 ppm solution; each of the 50 ppm solutions was separated into ten different beakers to study the contact time for 30, 60, 90 and 120. The pH of the solutions in each of the beakers was adjusted to 4 for copper. 0.2 g of the sawdust (adsorbent) was added to each solution and they were agitated using an orbital shaker at 118 rpm and subsequently, the solutions were filtered after each time interval and samples were subjected to analysis.

3.4 DATA ANALYSIS

The experimental design used for this study was 3 x 2 x 4 factorial experiment; the different sawdust species, two baselines (treated and untreated) and four levels of pH and time as factors were combined. The data obtained was analyzed using Analysis of variance (ANOVA) and descriptive statistics was used to show the results. Graphical representation was carried out using Microsoft Excel sheet.

4.0 RESULTS AND DISCUSSION

Table 1 shows the mean separation results for the adsorption capacity (% bound metal ion) of treated and untreated *Alstonia boonei*, *Erythrophleum suaveolens*, and *Ficus mucuso* sawdust at different pH levels in the removal of Copper ion from contaminated water observed in this study. It can be seen that *ficus mucuso* had the highest average adsorption capacity value of 75.71±19.95 % followed by *Erythrophleum suaveolens* which had the average value of 72.07±20.34 %, while *Alstonia boonei* had the least average adsorption capacity value 69.17±16.51 % from the three species investigated (Table 1). For the *Alstonia boonei* (AHUN) species, the treated sawdust samples had an average adsorption capacity (% bound metal ion) value of 69.75±13.78 % with its highest mean Adsorption capacity value (90.20±0.20 %) observed for sawdust samples at pH 4, followed by pH 3 (70.00±0.00 %), and pH 1 (65.00±0.40 %) while pH 2 had the least Adsorption capacity value of 53.78±0.43 %.

However, with respect to the untreated sawdust samples of *Alstonia boonei* (AHUN), an average Adsorption capacity value of 68.60±19.48 % was observed, with its highest Adsorption capacity value (94.40±0.00 %) observed for the sawdust samples at pH level 4, followed by those of pH 3 (77.00±0.20 %), and pH 1 (57.00±3.20 %), while the least Adsorption capacity value of 46.00±2.00 % was observed for untreated sawdust samples at pH 2.

Table 1: Adsorption capacity (% bound metal ion) of treated and untreated sawdust from *Alstonia boonei*, *Erythrophleum suaveolens,* and *Ficus mucuso* sawdust at different pH levels in

SPECIES	**TREATMENT**	**pH LEVEL**	**ADSORBTION CAPACITY (%)**	**SPECIES AVERAGE**

the removal of Copper from contaminated water.

		pH 1	65.00±0.40[c]	
		pH 2	53.78±0.43[d]	
	TREATED	pH 3	70.00±0.00[b]	
		pH 4	90.20±0.20[a]	
		Average	**69.75±13.78[j]**	
***Alstonia boonei* (AHUN)**				
		pH 1	57.00±3.20[c]	**69.17±16.51[c]**
		pH 2	46.00±2.00[d]	
	Nil	pH 3	77.00±0.20[b]	
		pH 4	94.40±0.00[a]	
		Average	**68.60±19.48[j]**	
		pH 1	53.00±0.20[c]	
		pH 2	43.00±1.00[d]	
	TREATED	pH 3	84.32±0.36[b]	
		pH 4	97.04±0.02[a]	
		Average	**69.34±23.08[k]**	
***Erythrophleum suaveolens* (ERU)**				
		pH 1	54.00±0.20[d]	**72.07±20.34[b]**
		pH 2	62.00±0.60[c]	
	UNTREATED	pH 3	91.00±0.02[b]	
		pH 4	92.16±0.04[a]	
		Average	**74.79±17.79[j]**	
		pH 1	56.80±0.80[c]	
		pH 2	50.00±4.00[d]	
	TREATED	pH 3	94.00±0.08[b]	
		pH 4	97.28±0.00[a]	
		Average	**74.52±22.30[j]**	
Ficus mucuso		pH 1	61.00±0.00[c]	**75.71±19.95[a]**

(OBOBO)		pH 2	58.00±0.20[d]
		pH 3	93.80±0.00[b]
	UNTREATED	pH 4	94.80±0.00[a]
		Average	**76.90±18.21[j]**

a, b c, d was used to represent pH according to their adsorption capacity while j represents the average.

4.1 Effect of Treatment and pH on the adsorption capacity

The ANOVA results at (α = 0.05) shows the Adsorption capacity (% bound metal ion) of both treated and untreated sawdust from *Alstonia boonei*, *Erythrophleum suaveolens*, and *Ficus mucuso* at different pH levels in the removal of Copper from contaminated water. The result shows that there was significant difference (P <.05) in the average % bound metal ion values of sawdust samples obtained from the three wood species with $F_{(2,48)}$ = 187.755 and P = 0.000, also there was significant difference (P <0. 5) in the mean % bound metal ion values of sawdust samples obtained from the three wood species subjected to the different treatments (treated and untreated) *with* $F_{(1, 48)}$ = 65.153 and P =0.000, as well as in the mean % bound metal ion values of sawdust samples from the three wood species at different pH levels (i.e. 1, 2, 3 and 4) with $F_{(3, 48)}$ = 5510.698 and P= 0.000. Furthermore, there were significant interactions (P < .05) between all the factors in this experiment with respect to percentage bound metal ion values of sawdust obtained from the three wood species as reported in this study. The mean separation results for the adsorption capacity (% bound metal ion) of treated and untreated *Alstonia boonei*, *Erythrophleum suaveolens*, and *Ficus mucuso* sawdust at different pH levels in the removal of Copper from contaminated water was observed in this study. The pH is a figure that expresses the acidity or alkalinity of a solution on a logarithmic scale on which 7 is neutral, lower values are more acid while higher values are more alkaline. It is a significant parameter whose influence must be investigated during adsorption of metal ions from contaminated water because it affects the solubility of metal ions, concentration of counter ions on the functional groups of the sawdust (adsorbent), the surface charge of the sawdust (adsorbent) and the degree of ionization of the adsorbate (Cu^{2+}) during reaction (Iftikhar *et al*., 2009; Sheng *et al*., 2004). Also studies have shown that with respect to the removal of Cu (II) at lower pH values, most of the carboxylic groups are not dissociated and cannot bind the metal ions in the solution;

as the increase in biosorption is due to dissociation of carboxylic groups at higher pH values and this could be explained by the increase in density of the negative charge on cell surface, causing proton removal on the cell binding sites, thereby increasing its biosorption capacity (Iftikhar *et al*., 2009; Cordero *et al*., 2004; Antunes *et al*., 2003). Consequently, the pH of 4 was taken as optimal pH value for further adsorption experiments, upon which further investigations were carried out since its respective hydroxides start precipitating from solutions at higher pH values, making true sorption studies impossible (Iftikhar *et al*., 2009). The adsorption capacities at different pH was shown (Figure 2).

In addition, with respect to the untreated sawdust samples of *Alstonia boonei* (AHUN), an average Adsorption capacity value of 68.60±19.48 % was observed, with its highest Adsorption capacity value (94.40±0.00 %) observed for the sawdust samples at pH level 4, followed by those of pH 3 (77.00±0.20 %), and pH 1 (57.00±3.20 %), while the least Adsorption capacity value of 46.00±2.00 % was observed for untreated sawdust samples at pH 2 which is in accordance with previous studies (Sciban *et al*, 2006). However, it can be seen that for the treated sawdust samples from *Erythrophleum suaveolens* (Eru), an average Adsorption capacity (% bound metal ion) value of 69.34±23.08 % was observed, with its highest adsorption capacity value (97.04±0.02 %) observed for sawdust samples at pH 4, it was followed by pH 3 which had an adsorption capacity of 84.32±0.36 %, and pH 1 which had Adsorption capacity of 53.00±0.20 %, pH 2 had the least adsorption capacity of 43.00±1.00 % as observed in this study. It has shown in previous studies (Sciban et al., 2006) that sawdust of deciduous soft wood (poplar) and coniferous softwood (fir), are capable for adsorption of copper and zinc ions from water, although, the adsorption capacities of these wood sawdusts are almost the same for both metal ions which is due to the difference in chemical composition and anatomical structure which is also in line with the result of this study as sawdust from the selected species are suitable for use as adsorbent for the removal of copper from contaminated water. The results were summarized in Table 1.

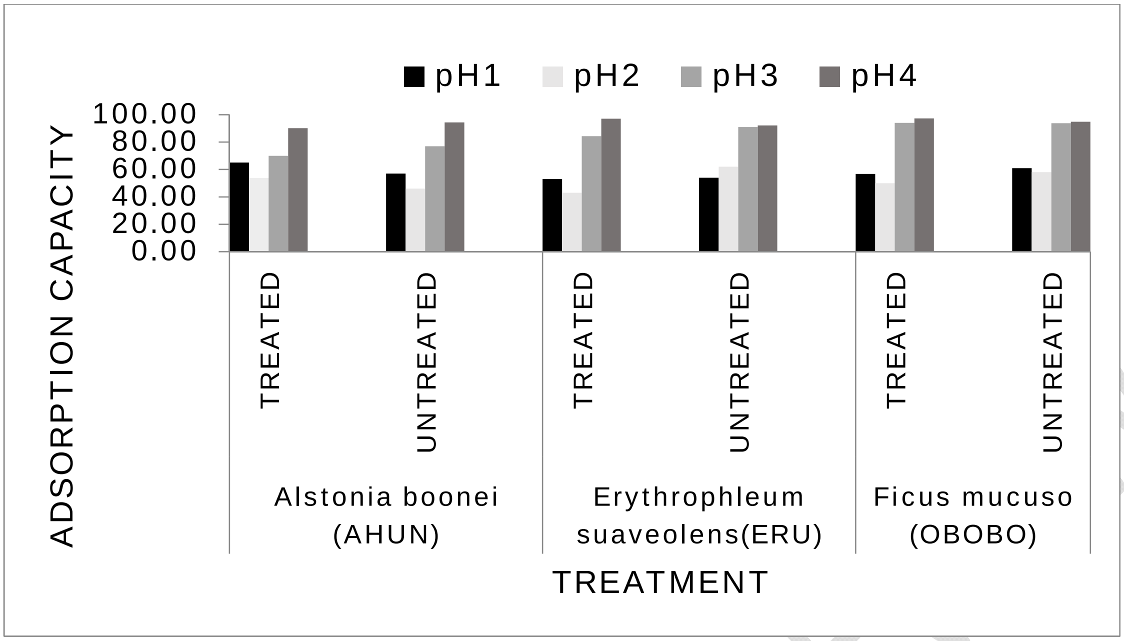

Figure 2: Adsorption capacities at different pH

4.2 Effect of Treatment and Contact Time at Optimal pH

The ANOVA results at (α = 0.05) shows the Adsorption capacity (% bound metal ion) of treated and untreated *Alstonia boonei*, *Erythrophleum suaveolens*, and *Ficus mucuso* sawdust at different contact time in the removal of Copper from contaminated water. The result shows that there was significant difference ($P < .05$) in the average % bound metal ion values of sawdust samples obtained from the three wood species with $F_{(2,48)}$ = 14341.048 and P = 0.000, also, there was a significant difference ($P < .05$) in the mean % bound metal ion values of sawdust samples obtained from the three wood species subjected to the different treatments (treated and untreated) *with* $F_{(1, 48)}$ = 4588.736 and P =0.000, as well as in the mean % bound metal ion values of sawdust samples from the three wood species at different contact time (i.e. 30, 60, 90 and 120) with $F_{(3, 48)}$ = 4022.673 and P= 0.000. Furthermore, significant interactions were observed between all the factors ($P<0.05$) in this experiment with respect to % bound metal ion values of sawdust obtained from the three wood species as reported. The mean separation results for Adsorption capacity (% bound metal ion) of *Alstonia boonei*, *Erythrophleum suaveolens*, and *Ficus mucuso* sawdust at different contact time in the removal of Copper from contaminated water obtained from the different treatments was observed in this study. It was also observed that *Erythrophleum suaveolens* had the highest average adsorption capacity value of 94.55±2.75 % followed by *Ficus mucuso* (OBOBO) which had the average adsorption capacity value of 93.17±2.73 %, while *Alstonia boonei* had the least average adsorption capacity value 89.65±5.98% from the three species studied. The treated *Alstonia boonei* (AHUN) species sawdust samples also had an average adsorption capacity (% bound metal ion) value of 87.30±7.53 % with its highest Adsorption capacity value (98.23±0.07 %) observed for the wood sawdust samples at 90 minutes contact time, this was followed by Adsorption capacity value 87.00±0.40 % which was observed at 120 minutes, and followed by adsorption capacity of 85.96±0.00 % which was obtained at 60 minutes, the least adsorption capacity value (77.99±0.19 %) was observed at 30 minutes These results are also consistent with past studies. Furthermore, the result shows that untreated *Ficus mucuso* (OBOBO) species sawdust samples had a mean time adsorption capacity value of 93.30±2.31, with its highest adsorption capacity value at 90 minutes observed to be 95.58±0.02 %. At 60 minutes, the Adsorption capacity value is 95.58±0.02 %, and then 30 minutes had Adsorption capacity value of 92.68±0.02 %. The least Adsorption capacity value (89.96±0.16 %) was observed at 120 minutes as observed in this study.

4.4 Effect of Contact Time

A suitable adsorbent for water treatment must not only have a high capacity but also a fast adsorption rate. Contact time is one of the most critical factors that must be considered in a batch adsorption process. The effect of contact time in the biosorption of Cu (II) at optimum pH as investigated in this study at different treatments levels revealed the time dependency of Cu (II) adsorption by the treated and untreated sawdust from the three wood species. However, the effect of contact time did not seem to follow any definite pattern and it also varied with both the species and treatments. The effect of contact time was correlated for the adsorption of Cu^{2+} ions at regular time intervals ranging from 30 minutes to 120 minutes. It was revealed that the percentage removal of metal ions by the different species sawdust samples increased quickly at 30 minutes (Figure 3). The initial rapid rate of adsorption may be due to the availability of the vacant active surfaces of the adsorbents for the adsorbate species in the solution and the slow pore diffusion of the ions (Karthikeyan *et al*., 2005; El-Ashtouky *et al*., 2008). The adsorption was fast within the first 30 minutes, slowing down between 30 and 60 minutes, adsorption and desorption occurred simultaneously from 30 to 120 minutes. The different treatment for sawdust had different adsorption capacities at different times which could be attributed to the unavailability of reaction sites which decreases with time. The high rate of metal removal in the beginning was due to the larger surface area being available for the adsorption of the metals. The process slowed down with time because the active sites were used up and the remaining vacant sites were difficult to be occupied by metal ions due to repulsive forces between the solute molecules on the solid and bulk phases. The amount of metal ion adsorbed at equilibrium time reflects the maximum adsorption capacity of the adsorbent under these particular conditions. The effect of treatment was not significant at the optimum pH (4) for the three species.

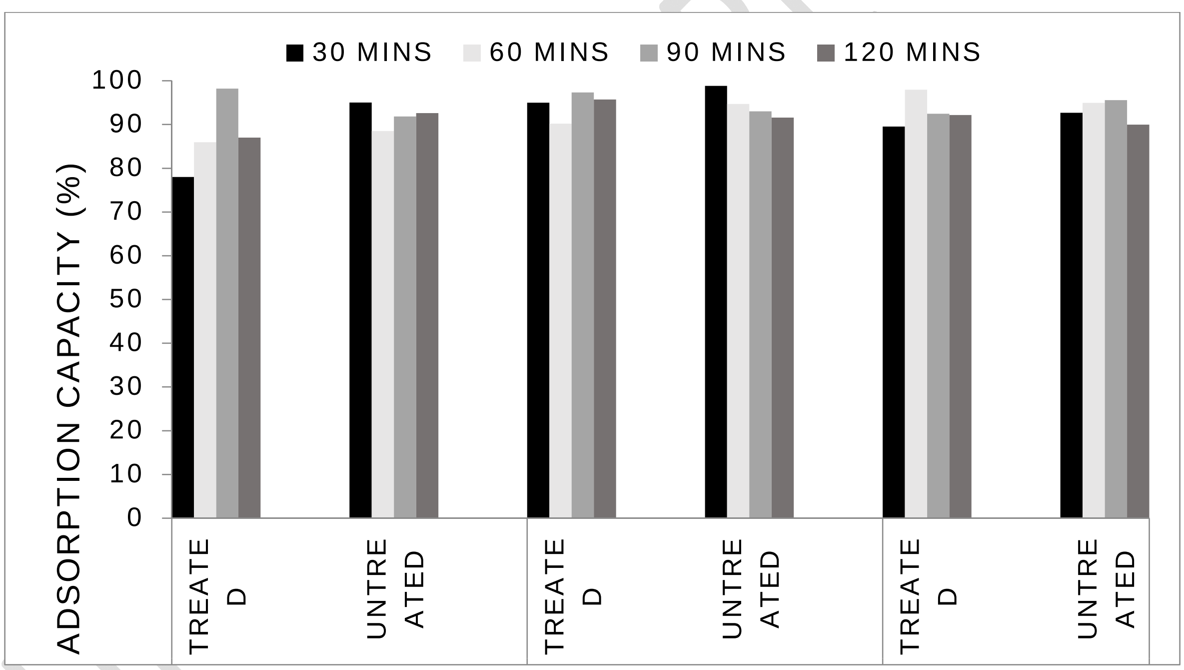

Figure 3: Adsorption capacities of the treatments on sawdust at different contact times

5.0 CONCLUSION

This study affirms the suitability of wood-waste for the development of low-cost, low energy and affordable technologies for effective and efficient removal of copper from contaminated water. Although, large volumes of sawdust are generated in sawmills, a portion of the sawdust can be used as adsorbent in removal of heavy metals from contaminated water. Effective wood-waste utilization is very crucial in wood processing industries. As previously reported, the adsorption of heavy metals by sawdust, the ion exchange and hydrogen bounding has been established as the principal sawdust heavy metal adsorption mechanism. The sawdust used for the removal of Cu (II) in this study had optimum adsorption of 97.28 % for pH 4 and 98.84 % removal for contact time at 30 minutes. Therefore, their high adsorption capacity per unit cost makes this material a promising and economical alternative. It was observed that Cu (II) is best adsorbed at pH of 4 using sawdust from the selected wood species used in the study. Also, it was observed that there was no significant difference in the adsorption capacity between treated and untreated wood sawdust from the three wood species, although, the untreated wood sawdust performed better than the treated samples in some cases. The implication of this is that there might be no need for treatment since it would require more energy consumption thereby increasing cost.

6.0 References

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