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KINETIC STUDY OF THE USE OF PALM KERNEL SHELL CHARCOAL IN THE ADSORPTION OF CHROMIUM ION (Cr^{6+}) FROM INDUSTRIAL WATER EFFLUENTS

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Abstract

The widespread agricultural waste product palm kernel shell has been employed as a potentially inexpensive adsorbent to extract chromium from wastewater. KOH was investigated in the synthesis of activated carbon from palm kernel shells. Studies were conducted using variables such as carbon content, pH, temperature, agitation speed, concentration, and contact time. It was discovered that chromium adsorption efficiency is pH dependent, with 6.5 being the ideal value. Chitosan, a partially deacetylated polymer prepared under the influence of a strong alkaline solution from chitin enhanced the adsorption properties of the activated charcoal. This adsorption capacity of chromium is considered ably higher compared to the values obtained with other adsorbents (50 to 120 mg/g). After the adsorbent was saturated in the metal ions, it was regenerated with 0.1M NaOH. Maximum adsorption occurred within 5 bed volume, while complete adsorption occurred within 10 bed volumes. The use of Langmuir and Freundlich isotherm models for fitting the adsorption data is a standard practice. The study effectively demonstrates the potential of palm kernel shell charcoal as an adsorbent for chromium removal. These findings demonstrate that palm kernel shell activated carbon has the favorable characteristics needed for the adsorption of chromium ions from industrial wastewater.

Keywords: Adsorption, activated carbon, industrial effluent, Chromium ions, agitation speed.

INTRODUCTION

According to preliminary research, oil palm fruit waste can be used to create chars with high porosities and sufficient densities (Guo & Lua, 2017). The presence of certain functional groups, such as carboxylic, hydroxyl, and lactones, which have a high affinity for metal ions, is what gives palm oil shell its adsorption capabilities (Hussein, 2016). A variety of activated carbon with surface modifications has been developed in recent years to improve its metal removal capacity, add to its economic value, aid in lowering the cost of waste disposal, and most importantly, provide a potentially less expensive alternative to currently available commercial activated carbon (CAC).

According to Kortenkamp, (2014), at least twenty metals are considered toxic, and half of these are released into the environment in proportions that are hazardous to human health. Chromium has both advantageous and disadvantageous characteristics. Chromium exists in two oxidation forms, Cr^{3+} ion and Cr^{6+}ion, each of which has unique toxicities, mobility, and bioavailability. Cr^{3+} ion is necessary for human nutrition (especially in glucose metabolism).

The majority of hexavalent chemicals are hazardous, and a few of them have been linked to lung cancer. While Cr^{3+} ion is comparatively stationary, Cr^{6+}ion is a potent oxidizing agent that may be absorbed via the skin and travels easily across soils and aquatic habitats (Park & Jung, 2011). Electroplating, leather tanning, cement, dyeing, metal processing, wood preservatives, paints, and pigments are all common uses for chromium and its derivatives. According to Raji (2015), these industries generate significant amounts of harmful wastewater effluents.Cr^{6+}ion discharge limits are 0.1 mg/l for inland surface waters and 0.05 mg/l for portable water, respectively (Hussein, 2016).

There are numerous physical and chemical procedures that can be used to remove Cr^{6+}ion from wastewater, including ion exchange, ultra-filtration, and reverse osmosis (Rengaraj, 2015; Yurlova, 2012; Benito & Ruiz, 2014).

Sludge generation is a significant disadvantage of precipitation. Ion exchange is thought to be a superior alternative method for this use. However, because of the high operating costs, it is not profitable. Commercial activated carbon (CAC) can be used to remove heavy metals including cadmium (Cd) (Ramos, 2015); Nickel (Ni) (Mostafa, 2017); Chromium (Cr) (Ouki, 2016); and Copper (Cu) (Monser & Aditoum, 2012) from wastewater.

CAC is still a pricey substance for removing heavy metals, though the deposition of molecular species onto a surface is referred to as adsorption in light of the aforementioned. The surface on which adsorption takes place is known as the adsorbate, and the molecular species that is adsorbed on it is known as the adsorbent. This is especially fascinating because natural biopolymers can reduce the concentration of transition metal ions to parts per million, making them appealing to industry. Large-scale natural resources or specific agricultural waste may have the potential to be employed as inexpensive adsorbents since they are readily available, represent unused resources, and are environmentally acceptable (Coughlin, 2013). Oil palm is the most significant commercial cash crop in Nigeria. Vegetable waste has increased dramatically as a result of the oil palm plantations' rapid expansion. According to some reports, Malaysia currently produces roughly 30 million tons of oil palm biomass each year, which includes fruit debris, fronds, and empty fruit branches. Two million tons of fruit shell, or endocarp, is produced annually from these volumes (Chan, 2015).

According to studies, chitosan eliminates five to six times more metals than chitin. This is explained by the free amino groups contained in chitosan (Yang & Zall, 2014). Chitosan, a bio-sorbent substance, is somewhat soluble at low pH levels and presents no obstacles to the development of commercial applications. In aqueous solutions, 1t is soft and has a propensity to clump together or form a gel. Additionally, chitosan's active binding sites are crucial for process design. In order to make the metal binding sites more accessible for process applications, it is important to provide physical support. Chitosan has the highest adsorption capacity for a number of metal ions among the numerous other low-cost adsorbents found (Olin, 2013; Balley, 2014; Ilyina, 2017). The primary structural component of mollusks, insects, crustaceans, fungi, algae, and marine invertebrates like crabs and shrimp is chitin (2-acetamido-2-deoxy-D-glucose-(N-acetyl-glucan) (Chen & Chang, 2019; Ilyina, 2017).

A significant portion of the chitinaceous waste produced by the processing of shellfish, crabs, shrimp, and krill into solid waste.

Chitosan (2-acetamido-2-deoxy-β-D-glucose-(N-acetylglucosamine) is a partially deacetylated polymer of chitin and is usually prepared from chitin by deacetylation with a strong alkaline solution.

$$\text{Chitin} \xrightarrow[\text{Alkaline solution}]{\text{Deacetylation}} \text{Chitosan (A polymer)}$$

In the current study, an effort was made to get over these mass transfer restrictions by creating a bio-sorbent by coating palm oil shell charcoal with chitosan and assessing its equilibrium adsorption capabilities. Combining the beneficial qualities of natural chitosan with those of oil palm shell charcoal could result in a composite matrix with a wide range of applications and outstanding adsorption capacities.

Chromium removal using oil palm kernel shell charcoal coated with chitosan as an economically effective alternative adsorbent to the usage of commercial activated charcoal using synthetic wastewater. This justification makes the research work robust and necessary.

When compared to other heavy metal ions, chromium is more prevalent in nature and most effluent streams. Hexavalent chromium ion (Cr^{6+}), which is extremely poisonous and carcinogenic, is present in effluent streams, which is a significant environmental problem (Chakpavarty, 2017). The main contributors to Cr^{6+} ion generation in wastewater streams are the electroplating and tannery industries' effluents. The current work examines the capacity of palm kernel shell, a very inexpensive adsorbent, to remove Cr^{6+} ion from the industrial effluent of the tannery and electroplating sectors.

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The starting Cr^{6+} ion concentration used in the studies in the current investigation ranged from 50 to 500 mg/l. According to experimental findings, the palm kernel shell adsorbent has a sizable capacity for removing Cr^{6+}ion from waste water streams. For the adsorption of Cr^{6+} ion on the palm kernel shell, the influence of several parameters including pH, contact time, adsorbent quantity, and starting Cr^{6+} ion concentration was examined. The Langmuir isotherm model's highest adsorption capacity was 41.5 mg^{-1} at pH 6.5.

When Cr^{6+} ion is adsorbed by the palm kernel shell, it does so with a higher adsorption efficiency of almost 95%. The current study also looked at how interference from other ions, which are typically present in industrial effluent streams from tanneries and electroplating, affected the removal of Cr^{6+} ion.

Evaluation of chromium

Hexavalent chromium ion (Cr^{6+}) and trivalent chromium ion (Cr^{3+}) are the two most widely utilized forms of chromium and are employed in many industrial operations. Other industries that require Cr^{6+} ion include electroplating, glass, ceramics, fungicides, rubber fertilizer, tanning, mining, and metallurgy, among others. Hexavalent chromium poses a health risk due to its propensity to interact with other elements. The highly mobile Cr^{6+} ion is more dangerous than other heavy metals since it is acutely poisonous, carcinogenic, and mutagenic to living things. To prevent Cr^{6+} ion effects on the ecosystem and public health, it is therefore vital to remove it from the environment. A cost-effective alternative approach for the treatment of Cr^{6+} ion contaminated waste water is widely demanded by the industry due to more stringent environmental requirements. Therefore, there is an urgent need for a treatment technology that is easy to use, efficient, and affordable for removing Cr^{6+} ion from wastewater.

In order to create a technology with high Cr^{6+} ion retention values and a high potential for the removal of Cr^{6+} ion from wastewater streams, other materials with structural composition or chemical properties suitable for this purpose must be used. It means that the utilization of adsorption as a treatment method for the removal of Cr^{6+}ion depends critically on the solution of an adsorbent. Commercial adsorbent costs make the adsorption procedure quite expensive, which has prompted researchers to look for new ways to create low-cost materials that are effective at removing Cr^{6+} ion. It was examined how different significant parameters, including pH, time, the amount of adsorbent used, and the baseline Cr^{6+} ion concentration, affected the results.

Environmental viability

Due to the constant change in technology, which generates trash as well as industrial products, environmental concerns have grown. In major nations, the manufacturing sector has been crucial to economic growth. But if industrialization proceeds quickly, it can affect the ecology and ecosystem, produce enormous amounts of waste, and inflict harm. Due to their non-biodegradable nature and persistence in the environment, pollutants from heavy metals, such as Cadmium (Cd), Chromium (Cr), Lead (Pb), Copper (Cu), Manganese (Mn), Zinc (Zn), and Mercury (Hg), are widely discharged into wastewater from industries. These pollutants are very toxic and harmful to living organisms by lowering the reproductive ability, preventing proper growth and development, and even causing death (Alturkmanic, 2014). Ions will interfere with the transport process via the cell wall, disrupting how cells work. Concern was also expressed over Malaysia, where 27% of the rivers are healthy and 63% of them are contaminated or dead (Modak & Natarajan, 2018). These data demonstrate the necessity of treating wastewater before releasing it into rivers or other bodies of water. The effluent must therefore be treated before being released into rivers in order to solve the issue. Chemical precipitation, solvent extraction, oxidation, reduction, electrolytic extraction, dialysis/electro dialysis, cementation, dilution, adsorption, filtration, flotation, air stripping, steam stripping, flocculation, sedimentation, and soil flushing/washing chelating are just a few of the technologies that have been developed for the purification and treatment of wastewater (Caddick, 2016). It has been shown that the adsorption process is the best method for removing contaminants from wastewater. It is favored mostly because to its practicality, simplicity of use, and ease of operation. In addition to eliminating a variety of pollutants, it is often used to reduce water contamination. Since it has a large surface area and pore volume as well as being inert, activated carbon (AC) is frequently utilized as an adsorbent. Researchers are very interested in using a low-cost precursor to replace the traditional (AC). The advantages of using activated charcoal as a raw material are its high carbon content, low inorganic content, high density, and sufficient volatile content, as well as its predictable availability across the nation, potential degree of activation, and low cost (Normah, 2017). The major component of activated charcoal (AC) is carbon with a big surface area, a large pore volume, and a high porosity, which is where adsorptions occur.

MATERIAL AND METHODS

Oil palm kernel shell was obtained from Adapalm in Eziala-Mbawsi, Isiala-Ngwa North in Abia State, Nigeria. Preparation of char from the oil palm kernel shell was carried out as previously described (Guo & Lua, 2017). The received oil palm kernel shell was first dried at 1100C for 24 hours to reduce the moisture content. The dried shell was cut, grinded and sieved. Size fractions of 2.0-2.8 mm were used.

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In a horizontal controlled environment furnace, hydrolysis was done. Alumina bowls containing around 15g of the sample were pushed into the furnace's hot zone. Nitrogen was employed as a purge gas during the hydrolysis process at a flow rate of 150 cm^3/min. The furnace temperature was raised from ambient to 600°C at a rate of 100°C/mm, and it was maintained there for two hours. Weight before and after hydrolysis were used to calculate the amount of weight lost. The hydrolyzed sample was powdered after being crushed.

From Indorama Eleme Petrochemicals Limited, east-west Express road, Eleme, Port Harcourt, Rivers State, chitin (from crab shells) was acquired. From B.G. Technical Limited (pipeline and well services), plot 149 Trans-Amadi industrial Layout (opposite Michelin), Port Harcourt, a highly purified preparation of pectolytic enzymes from a chosen sham of Aspergillus Niger was used. The local market was used to buy the cabbage. All additional compounds were of the analytical variety. A minor adjustment was made in accordance to Couphlin, (2013) and deacetylation process. The chitin that was purchased was first blended and sieved. To prevent calcium salt demineralization, particles between 0.5 and 1.0 m in size were first soaked for an hour in a 5% hydrochloric acid solution. The decalcified chitin was then transferred to a 50% sodium hydroxide (NaOH) solution after being rinsed with distilled water. The remains was exposed to radiation in a home microwave oven (EME2662, ElecchoCux, operating frequency 2.5 GH2). The deacetylated chitin (now known as chitosan) was prepared for usage after being rinsed with distilled water and dried at 600^0C.

To 100ml of 100% oxalic acid, 50g of chitosan was gradually added while being constantly stirred. In order to facilitate mixing, the mixture was also heated to a temperature of 40 to 50^0C. The chitosan-Oxalic acid mixture solidified as a yellowish, thick gel at ambient temperature.

As mentioned by Kadirvelu (2014), oil palm shell char (OPSC) conditioning was accomplished by oxidizing it with sulfuric acid. Deionized water was used to wash the OPSC until all adhering powder and leachable impurities caused by free acid were eliminated. After 24 hours of treatment with 2% H_2SO_4 (v/v) at 110°C in an incubator, the samples were then soaked in deionized water until the solution was stable. The remaining acid was then removed by soaking the adsorbent in 2% $NaHC0_3$ (w/v) for an extended period of time.

The samples were then dried overnight in an oven at 110^0C, cooled to room temperature, and kept in a desiccator until use (hereinafter referred to as acid treated oil palm shell char- AOPSC) (Kadirvelu, 2014).

RESULTS AND DISCUSSION

Because of their naturally high densities and carbon contents, oil palm kernel shell has been effectively used to make high-quality activated carbon (Normah, 2017; Hussein, 2016; Guo & Lua, 2017). According to Guo & Lua's (2017) approach, char form oil palm kernel shell was made in this study. The apparent densities of the initial material and the char as well as their solid densities were not calculated.

Many of the documented processes for converting the chitin in crustacean shells to chitosan were time- and reagent-intensive. The deacetylation technique subject to microwave irradiation was used in this work, which utilized a rather quick and mild deacetylation approach suggested by Coughlin (2013) with some minor modifications. Polar molecules in the sample, such as water, align with the microwave's continuously shifting magnetic field during microwave irradiation. A considerably more effective chemical reaction results from microwave treatment because it increases the accessibility of the vulnerable bonds (Roy & Pectinex, 2013).

By coating it on other adsorbents like alumina, charcoal, or interacting it with other adsorbents like alginate to form rigid matrix structures of better mechanical strength, the practical issues of Chitosan solubility at low pH aqueous systems, gel forming behavior, and mass transfer limitations were overcome (Babel & Kuriawan, 2014).

In this study, chitosan was coated over oil palm shell charcoal to solve these issues, and the coating procedure produced an adsorbent that was stable in acidic conditions and was a granular composite. Due to interactions between the two materials' opposing charges, the anionic oil palm shell charcoal and the cationic chitosan produced a stable, granular composite matrix. Depending on the pH, chitosan binds to both cationic and anionic forms of Cr^{6+} ion as well as various other metals, including arsenic. At a pH of about 4, Cr^{6+} ion produces the dichromate anion.

According to Chen and Chang (2019), the chitosan's amine groups play a major role in the absorption of Cr^{6+}ions from the solution. The amine group on chitosan is protonated at low pH levels. This causes the NH_3 functional group in chitosan to engage with chromate ion ($Cr_2O_7^{2-}$), and the nature of this interaction is mostly electrostatic attraction (Olin, 2013).

Aspects that affect the adsorption of Cr^{6+}ion

The impact of several operational parameters, including adsorbent dosage, agitation speed and contact time was established. The outcome was expressed as the adsorbent's removal efficiency (E), which was defined as:

$$E (\%) = [(C_0-C_1)/C_0] \times 100$$

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Where C_0 and C_1 are the initial and final equilibrium concentration of Cr^{6+} ion solution (Mg/1), respectively. The Cr^{6+} ion concentration was determined calorimetrically according to standard methods (Clesceri, 2015).

Because it influences the solubility of the metal ions, the concentration of the counter ion on the functional groups of the adsorbent, and the degree of ionization of the adsorbent during reaction, pH is a crucial parameter for the adsorption of metal ions from aqueous solutions. The pH was changed from 1.0 to 9.0 to test how pH affected the effectiveness of the Cr^{6+} ion removal. Figure 1 demonstrates how pH 5 affects the absorption of free ionic Cr^{6+} ion.

pH removal efficiency for CCAB increased from 65% to 92%, over pH range from 1.0 to 5.0. The other two adsorbent, i.e chitosan coated oil palm shell carbon and acid treated oil palm shell charcoal also showed similar trends but with much lower removal efficiency and slight different optimum pH value (Lytle, 2018).

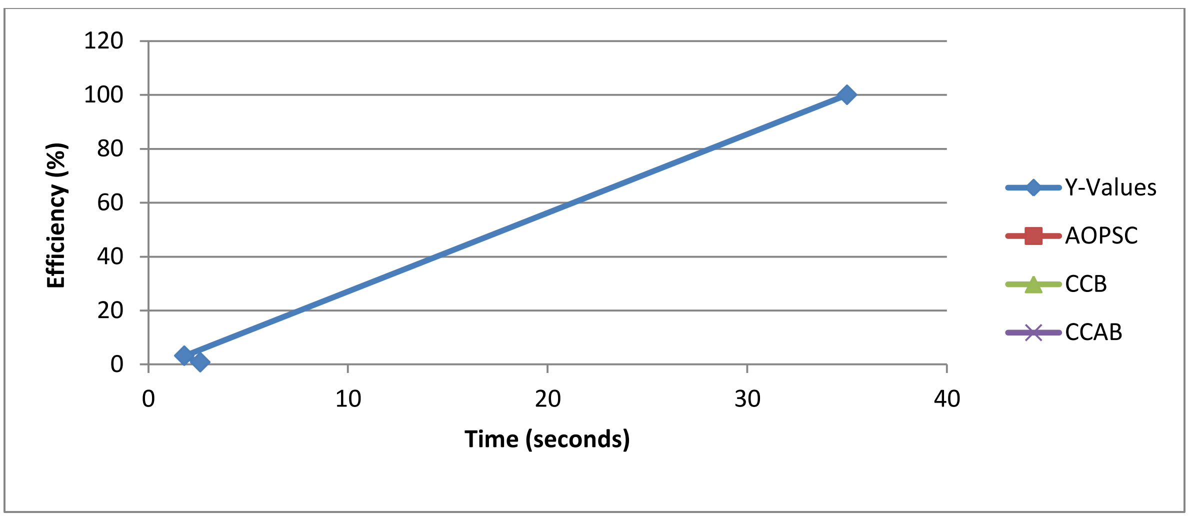

Figure 1. Effect of dose on the equilibrium removal efficiencies of chromium ions using different types of adsorbents.

Chromium ion (Cr^{6+}) concentration was 20mg/l
Agitation speed was 200rpm
Contact time was 3 hours
pH of solution was 6.5
Temperature was maintained at 25^0C

It is interesting to note that the saturated values of Cr^{6+} ion removal efficiency of the three types of adsorbents are different from one to another due to the extent of surface modification. At low pH (below 5), the amine group on chitosan is protonated to varying degree (Mostafa, 2017 & Udaybhaskar, 2011).

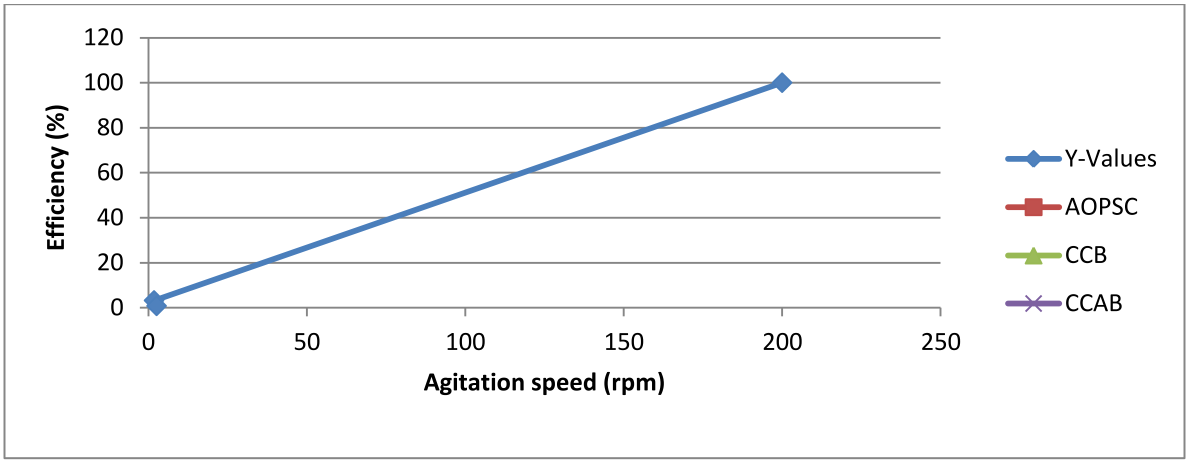

Figure 2. Effect of agitation on the equilibrium removal efficiencies of Cr^{6+} ion using different types of adsorbents.

Cr^{6+} ion concentration was 20mg/l
Contact time was 3 hours
pH of solution was 6.5
Amount of each adsorbent was 40g/l
Temperature maintained at 25^0C

This is consistent with other researchers' findings that the primary adsorption mechanism involves electrostatic interactions between anions and the protonated sites of the adsorbent as well as interactions between oxygen-free Lewis basic sites and the free electrons of the anions (Leon, 2015).

By adjusting the agitation speed from 0 to 200 rpm (without shaking) and holding the ideal pH and adsorbent dose constant, it was possible to determine the impact of agitation speed on the removal effectiveness of Cr^{6+}ion. Figure 3 illustrates how the efficacy of Cr^{6+}ion removal generally improved with agitation speed. When the agitation speed was increased from 50 to 100 rpm, the CCAB adsorbent's ability to remove Cr^{6+}ion rose from 70% to 90%. The findings revealed a significant differential deviation of 0.1. These results can be attributed to the fact that agitation speed improvement enhances Cr^{6+}ion transport to the adsorbent surface.

This also suggests that a shaking velocity between 100 and 200 rpm is adequate to guarantee that all of the surface binding sites are made accessible for Cr^{6+}ion absorption. The impact of external film and diffusion on the rate of adsorption can hence be assumed to be minimal. For ease of use, a 150 rpm agitation speed was chosen as the best speed for all adsorbents.

The findings also show that removal effectiveness increased as contact time before equilibrium was attained. The ideal values for other parameters, including adsorbent dosage, solution pH, and agitation speed, were maintained while the temperature was held at 250°C.

It can be shown that increasing the contact time from 30 to 150 mm caused the CCAB's Cr^{6+}ion removal effectiveness to rise from 60% to 90%. The ideal contact period between CCAB and CCB as adsorptive materials and AOPSC was 300mm. Consequently, the chitosan-coated beads need less contact time. The binding capacity was greatly boosted, and the process moved along quickly, thanks to the increased availability of different functional groups on the surface of chitosan, which are necessary for interaction with anions and cations. This result is important, as equilibrium time is one of the important parameters for an economical waste water treatment system.

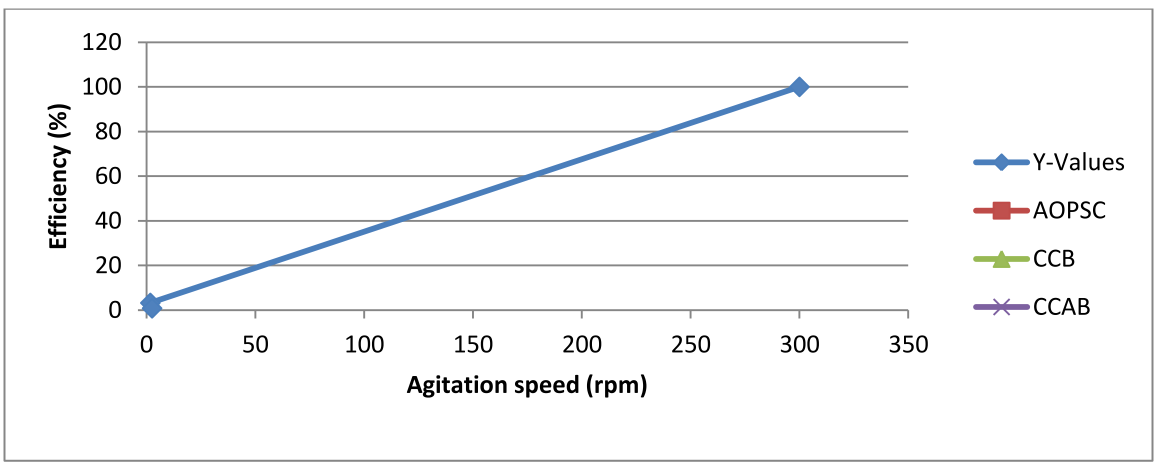

Figure 3. Effect of constant time on removal of chromium ions using different adsorbents.

Cr^{6+}ion concentration was 20mg/l
pH of solution was 6.5
Amount of each adsorbent was 40g/l
Agitation speed was 200rpm
Temperature as maintained at 25^{0}C

To examine the relationship between adsorbent and aqueous concentration at equilibrium, adsorption isotherm models are widely employed for fitting the data of which the Langmuir and Freundlich equations are most widely used. The Langmuir model assumes that the uptake of metal ions occurs on a homogenous surface by

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monolayer adsorption without any interaction between adsorbed ions. To get the equilibrium, mass in each sample was kept constant. Three hours of equilibrium periods for adsorption experiments were used to ensure equilibrium condition. The Langmuir model takes the form:

$$Qe = \frac{K_L C_e}{1+a_L C_e}$$

Where Qe (mg/g) is the amount of metal ions adsorbed onto the unit mass of the adsorbent to form a complete monolayer on the surface.

K_L is the Langmuir equilibrium constant which is related to the affinity of binding sites.

Ce is the solution phase metal ion concentration of the Langmuir constant (Alturkmanic, 2014).

Using Langmuir isotherm, the equilibrium data yielded the ultimate adsorption capacity value for the chitosan coated AOPSC on per gram basis of chitosan as 154mg/g.

This adsorption capacity for chromium is considered ably higher compared to the values obtained with other adsorbents (50 to 120 mg/g) (Rodriguez-Reinoso, 2018). After the adsorbent was saturated in the metal ions, it was regenerated with 0.1M NaOH. Maximum adsorption occurred within 5 bed volume, while complete adsorption occurred within 10 bed volumes.

CONCLUSION

It was explored whether waste materials like palm kernel shells could be used to remove Cr^{6+} ion from industrial waste water streams. Comparing palm kernel shell to other inexpensive, commercially available adsorbents, it was discovered that palm kernel shell was a better adsorbent for the removal of Cr^{6+} ion. At pH 6.5, the greatest proportion of Cr^{6+}ion elimination was accomplished. The surface of the adsorbent was positively charged at low pH, and $HCrO_4^{2-}$was the predominant form of Cr^{6+} ion.

The kinetic experiments revealed that the rate of Cr^{6+}ion adsorption was higher during the first 200 mm and then decreased in the latter stages of adsorption. At 105mm, Cr^{6+} ion adsorption on the palm kernel shell reached equilibrium. The proportion of Cr^{6+} ion removed increases with an increase in adsorbent quantity, and the ability of palm kernel shell to adsorb Cr^{6+} ion diminishes as more unsaturated adsorption sites become available.

As the initial Cr^{6+} ion concentration increases, the % removal drops and the adsorption capacity rises. With a variety of isotherm models, including the Langmuir and Freundlich isotherm models, the equilibrium adsorption data were satisfactorily fitted.

The Langmuir model provided the best match for the equilibrium data, confirming that chromium ion Cr^{6+} was adsorbed in a monolayer on the palm kernel shell. Langmuir isotherm models had a maximum adsorption capacity of 154mg/l, which was a respectable amount compared to other adsorbents.

The second-order kinetic model was used to explain the kinetics of Cr^{6+}ion adsorption employing palm kernel shell as an adsorbent for a range of starting Cr^{6+}ion concentrations. It was discovered that other ions including Fe^{2+}, Pb^{2+}, Na, and SO_4^{2-} in aqueous solution had a substantial impact on Cr^{6+} ion adsorption. The palm kernel shell can be used to remediate Cr^{6+} ion rich industrial waste water streams from the tannery and electroplating processes. Acid and base treatment regenerates the saturated adsorption. For the elimination of Cr^{6+} ion, an adsorption efficiency of more than 95% was achieved.

RECOMMENDATIONS

Due to its severe toxicity to people, aquatic life, and the environment in general, only chromium was researched throughout this investigation. Additionally, it was discovered that the inexpensive adsorbent (palm kernel shells) worked well for removing chromium ion from industrial wastewater, but it was not tested on other heavy metals.

In order to remove other heavy metals from industrial waste water, such as nickel (Ni), lead (Pb), cobalt (Co), vanadium (V), manganese (Mn), etc., a thorough examination using the adsorbent mentioned above is advised.

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