Phosphate and Nitrate Removal from Aqueous Solution by Carbonated and Uncarbonated African Nutmeg (*Monodora Myristica*) Shell

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Abstract

This study was aimed at investigating the possibility of using *Monodora myristica* shell for the absorption of phosphate and nitrate from wastewaters. Four categories of the shell (untreated raw shell, treated raw shell, untreated carbonated shell and treated carbonated shell) were used for the investigation. Three different concentrations (2.5 %, 5 % and 10 %) of the respective shells were used for the study. The results revealed that none of the different modifications of the shell had phosphate absorption capacity. All the different shell modifications showed remarkable nitrate absorption capacity from solution. The findings also revealed a saturation time of 3 h for nitrate absorption at shell concentrations of 5 % to 10 % and a saturation time of 5 h at shell concentration of 2.5 %. At the end of 3 h contact time, nitrate concentration in the presence of the untreated carbonated shell was observed to decrease from 231.47 mg/L to 63.10 mg/L and 28.57 mg/L, at shell concentrations of 5 % and 10 %, respectively. In the presence of the treated carbonated shell, after 3 h contact time, nitrate levels showed significant decreases from 231.47 mg/L to 169.99 mg/L and 56.83 mg/L, respectively. Similarly, after 3 h contact time, nitrate levels in the presence of the untreated and treated shells showed decreases from 231.47 mg/L to 167.63 mg/L and 158.81 mg/L, and from 231.47 mg/L to 56.83 mg/L and 45.59 mg/L, at shell concentrations of 5 % and 10 %, respectively. The study was able to give an insight into the potential use of the *Monodora myristica* shell as a bio adsorbent in nitrate removal from wastewaters.

Keywords: Absorbent *Monodora myristica,* nitrate, phosphate, wastewater

1. Introduction

Both surface and wastewaters are known to contain levels of phosphorus and nitrogen in various compounds, which is important for living organisms. Although the levels of concentrations of these compounds is balanced in natural conditions, when their input to waters is greater than a living organisms can assimilate, the problem of pollution occurs (Rybicki, 1997). Eutrophication of water bodies is known to promote aquatic plant growth and may lead to the proliferation of undesirable algae blooms and toxic cyanobacteria that can pose serious health hazard to humans and livestock (Henderson *et al*., 2007).

To safeguard and protect surface water bodies against the impacts of eutrophication and to maximize the health and environmental benefits associated with the use and discharge of wastewater, several legislations and guidelines, both at national and international levels have been developed (WHO, 2006). In order to meet effluent discharge standards and guidelines, wastewater treatment facilities are obliged to meet discharge consents of nutrients into the environment (Karostynska *et al*., 2012). The common methods for the removal of nutrient from wastewater are chemical precipitation, biological and adsorption. Because of the several drawbacks of chemical precipitation (increased aquatic toxicity, increased sludge production, increased filamentous growth, decreased sludge settlebility and dewatering characteristics and increased cost), its application in nutrient removal is discouraged in recent years (Imram, 2005).

Also, despite the indication that the phosphorus and nitrogen removal efficiency of the conventional biological nutrient removal process is considered to be appropriate, with possible effluent total nitrogen concentration of less than 10 mg/L, removal efficiency depends on the recirculation ratio that transfers the nitrate produced by nitrification in the aerated zone back to the anoxic zone, and is therefore limited to 75 - 90% (Galil, 2009). Although absorption studies have been most concentrated on heavy metal removal (Doyurum and Celik, 2006; Kumar, 2006; Zvinowanda *et al*., 2009), its application in the removal of phosphorus and nitrogen from wastewater has been reported (Namasivayam and Sangeetha, 2005; Jyothi *et al*., 2012).

Monodora myristica belong to the Ananaceae family and is prevalent in the Southern part of Nigeria. According to reports, almost every part of the plant has economic importance, although the most economically important part is indicated to be the seeds. The kernel is a popular condiment used as souring agent (Burubai *et al*., 2008). In Central Africa Republic, the seeds of fruits of *Monodora myristica* are used as condiment and drug in the treatment of headache and hypertension.

Although several studies have been carried out on *Monodora myristica,* the majority of them have concentrated on the chemical composition of the different parts and the evaluation of the antimicrobial of its essential oils and related studies (Agnaniet *et al*., 2004; Koodou *et al*., 2007). Based on the available literatures, no report has been found on the use of *Monodora myristica* shell for nutrient removal studies. Because, several researches have been carried out on effective and economical ways for nutrient removal from wastewater, this study was therefore aimed at investigating the possibility of using *Monodora myristica* shell for the absorption of phosphate and nitrate from wastewaters.

2. Materials and Methods

The African nutmeg (*Monodora myristica*) seeds were obtained from a traditional health practitioner at OjaJagun in Ogbomoso, Oyo State, Nigeria. The seeds were identified by the Department of Department of Pure and Applied Biology, Ladoke Akintola University of Technology, Ogbomoso, Oyo State. The seeds were dehosed to separate the shell from the main seed. The separated shells were oven-dried at 50 °C for 3 d, after which they were pulverized into fine powder using a sterilized food mill. The pulverized powder was stored in air-tight plastic containers at room temperature until use or further treatment.

Four categories of the *Monodora myristica* shell were used for the study. The categories were the raw pulverized shell that was not subjected to any treatment, a raw pulverized shell that was treated with 20 % sulphuric acid, a raw pulverized shell that was carbonated but untreated and a carbonated shell that was treated with 20 % sulphuric acid. For carbonation, approximately 250 g of the pulverized shell was placed in a crucible and placed in an oven at a temperature of 250 °C for 1 h.

The treatment of the raw and carbonated portions were carried out as follows: To a 50 g of the pulverized sample (raw or carbonated) in a 250 mL beaker, 100 mL of 20 % sulphuric acid was added and allowed to stand for 1 h, with stirring every 5 min to obtain a homogenous mixture. After the expiration of the 1 h period, the contents were washed several times with deionised water and filtered through Whatman No 1 filter paper. The washing with deionised water was to ensure every trace of the acid was removed. This was confirmed by testing with a litmus paper. After filtration, the supernatant, which was the treated portion left in the filter paper, was dried in a hot air oven at 80 °C for 24 h.

In this paper, the four categories of shells used for the investigation were referred to as follows:
- The untreated raw shell
- The treated raw shell
- The untreated carbonated shell
- The treated carbonated shell

For nutrient removal studies, to a 100 mL Erlenmeyer flask, containing 40 mL of deionised water that was spiked with a known quantity of potassium nitrate and potassium dihydrogen phosphate, to serve as nitrate and phosphate sources, respectively, a known quantity of the respective categories of the *Monodora myristica* shell was added and agitated as 100 rpm and left for 5 h. At time zero, which was the initial concentration before the addition of the shell and every 1h for the next 5 h, the contents of each flask was filtered through Whatman No 1 filter paper and the filtrate analysed for nitrate and phosphate concentration, using standard procedures (APHA, 2001). Phosphate and nitrate analyses were determined using the ascorbic acid and salicylate methods, respectively. All experimental analysis and designs were done in triplicate. In addition, all reagents used were of analytical grades.

All statistical analyses were carried out using the PAST: Paleontological statistics software package for education and data analysis, as described by Hammer *et al*. (2001). Comparisons of means were determined using the One-Way Analysis of Variance (ANOVA) while test of relationship was carried out using the Pearson Correlation Index.

3. Results

As shown in Fig. 1, in the presence of the untreated carbonated shell in the aqueous solution, phosphate concentration was only observed to decrease within the first 1 h of contact, after which increases in concentration were observed with contact time. This trend was irrespective of the shell concentration used for the investigation. At the end of the 5 h contact time, phosphate concentration increased from 73.32 mg/L to 82.91 mg/L, 85.72 mg/L and 89.28 mg/L, at shell concentrations of 2.5 %, 5 % and 10 %, respectively. At the end of the 5 h contact time, the changes in concentrations were not observed to differ between the different shell concentrations ($P \leq 0.05$).
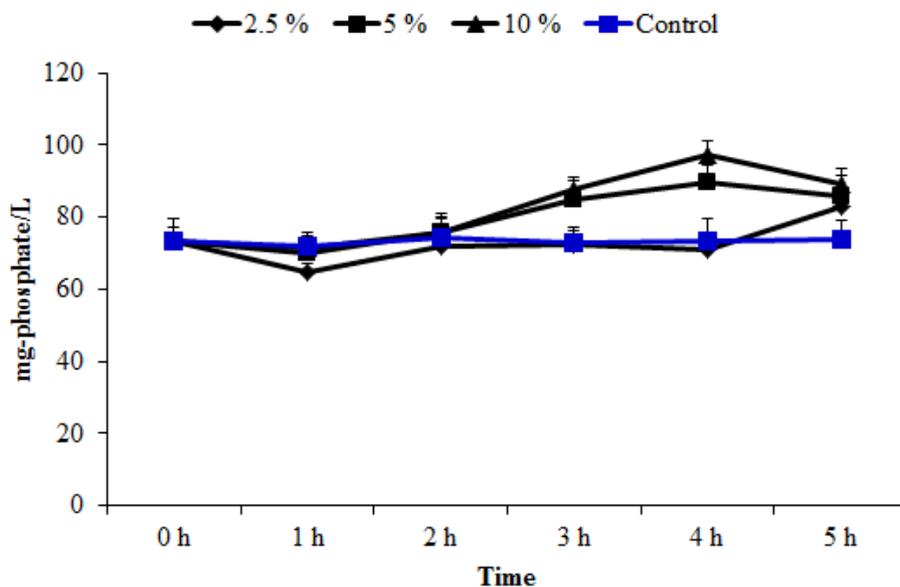
Fig. 1: Phosphate concentration in the aqueous solution when in contact with the untreated carbonated shell

In the case of nitrate removal, in the presence of the untreated carbonated shell, significant removal was observed after 5 h contact time at shell concentration of 2.5 %, decreasing from 231.47 mg/L to 59.06 mg/L. At shell concentration of 5 % and 10 %, significant removal was observed after 3 h contact time, after which there were steady increases in concentration. At the end of 3 h contact time, nitrate concentration was observed to decrease from 231.47 mg/L to 63.10 mg/L and 28.57 mg/L, for shell concentrations of 5 % and 10 %, respectively (Fig.2). When compared with the control, the concentration of nitrate at the optimum contact times with the different shell concentrations were observed to be significantly lower ($P \leq 0.05$).
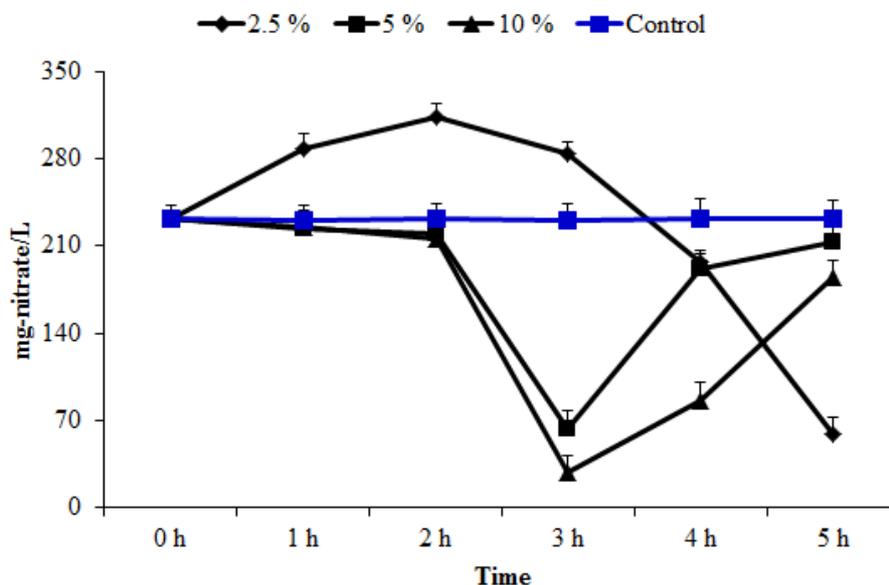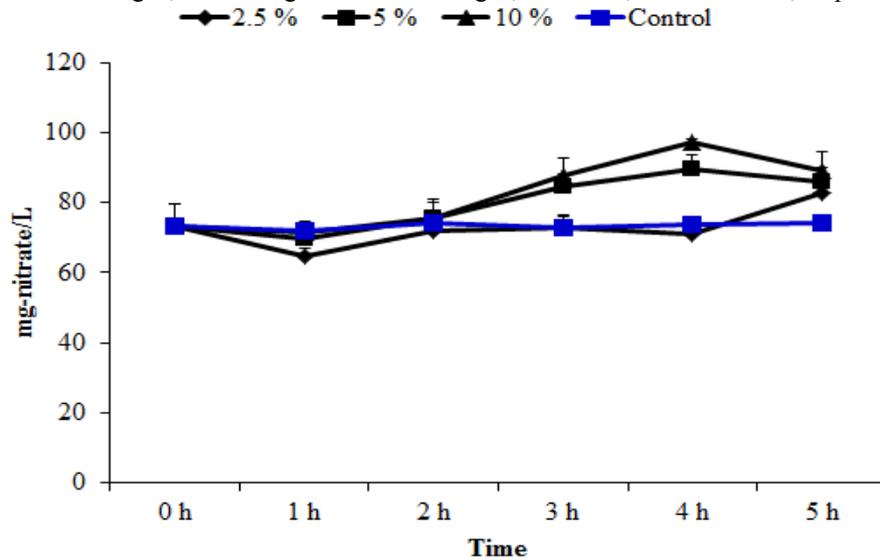

Fig. 2: Nitrate concentration in the aqueous solution when in contact with the untreated carbonated shell
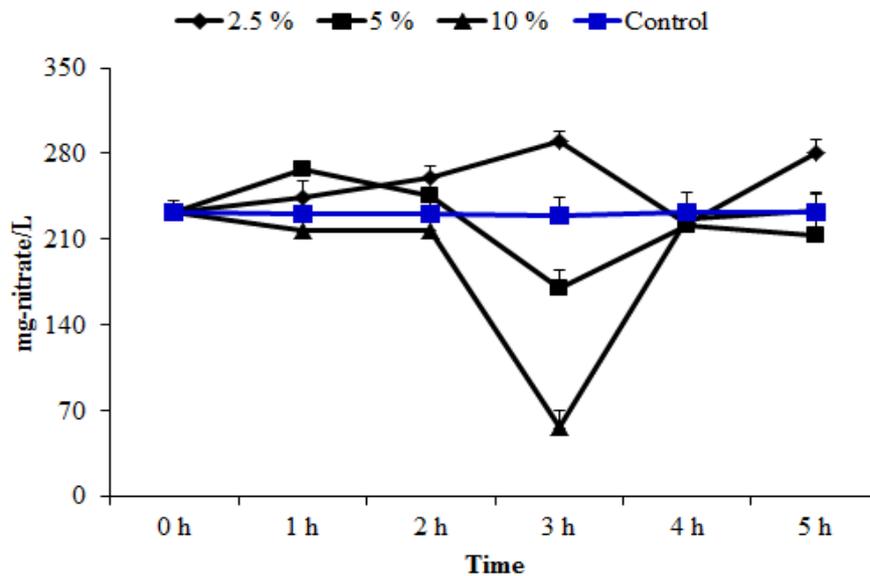
In the presence of the treated carbonated shell, phosphate concentration was not observed to follow any pattern, showing slight increases and decreases with time. This trend was irrespective of the shell concentration used for the investigation. From the initial concentration of 73.32 mg/L, phosphate level in the solution was observed to increase to 82.91 mg/L, 85.72 mg/L and 89.28mg/L, for shell concentrations of 2.5 %, 5 % and 10 %, respectively (Fig. 3). Generally, phosphate concentrations were observed to be significantly higher in presence of shell concentrations of 5 % and 10 % than that of 2.5 % ($P \leq 0.05$).

For nitrate levels in the presence of the treated carbonated shell at concentration of 2.5 %, no remarkable decrease in concentration was observed with contact time, rather there were steady increases and decreases with

time. At shell concentrations of 5 % and 10 %, at the end of 3 h contact time, nitrate levels showed significant decreases from 231.47 mg/L to 169.99 mg/L and from 231.47 mg/L to 56.83 mg/L, respectively before steady increases were observed. At the end of the 5 h contact time, nitrate levels at the different shell concentrations were found to be 280.78 mg/L, 212.72 mg/L and 233.65 mg/L, for 2.5 %, 5 % and 10 %, respectively (Fig. 4).
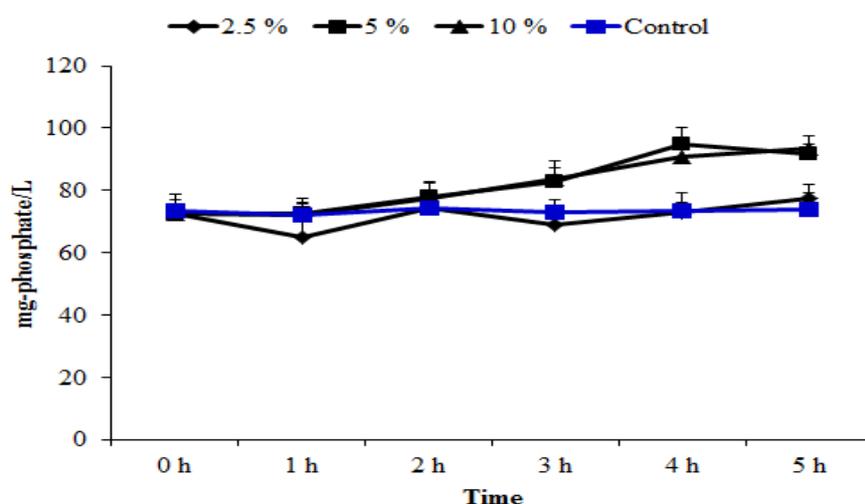

Fig. 3: Phosphate concentration in the aqueous solution in when in contact with the treated carbonated shell
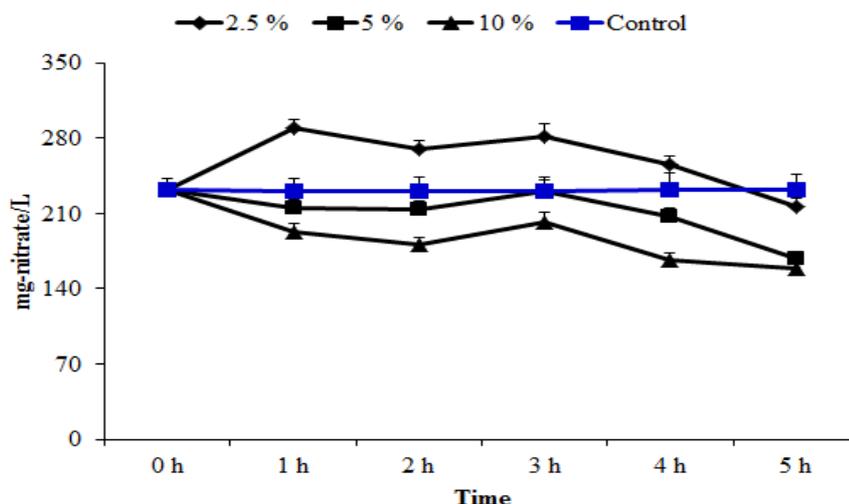

Fig. 4: Nitrate concentration in the aqueous solution in when in contact with the treated carbonated shell
As shown in Fig. 5, at shell concentration of 2.5 % of the untreated raw shell, phosphate levels in the aqueous solution was observed to show remarkable decrease after 1 h contact time, after which there was increase in concentration with contact time. At shell concentrations of 5 % and 10 %, there were steady increases in phosphate concentration with contact time. At the end of the 5 h contact time, phosphate concentrations increased from the initial level of 72.33 mg/L to 77.57 mg/L, , 91.75 mg/L and 93.48 mg/L, at shell concentrations of 2.5 %, 5 % and 10 %, respectively (Fig. 5). Although no significant decrease in phosphate concentration (when compared to the control) was observed at the different shell concentrations, at the end of the 5 h contact time, phosphate concentration at 2.5 % was observed to be significantly lower than those at shell concentration of 5 % and 10 % (($P \leq 0.05$).

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Fig. 5: Phosphate concentration in the aqueous solution when in contact with the untreated raw shell
In the case of nitrate levels in the aqueous solution, at the end of the 5 h contact time, concentrations were observed to decrease from 231.47 mg/L to 216. 94 mg/L, 167.63 mg/L and 158.81 mg/L, at shell concentrations of 2.5 %, 5 % and 10 %, respectively (Fig. 6). At the end of the 5 h contact time, the concentration of nitrate at 5 % and 10 % shell concentrations were observed to be significantly lower than that at shell concentration of 2.5 %. When compared to the control, after the 5 h contact period with the shell, when compared with the control, decrease in nitrate levels was only observed at shell concentrations of 5% and 10 % (P≤ 0.05).

Fig. 6: Nitrate concentration in the aqueous solution when in contact with the untreated raw shell
In the treated raw shell, except for 1 h contact time, at 2.5 % shell concentration when there was a slight decrease, phosphate concentration was observed to consistently increase with contact time at the different shell concentrations. After the 5 h contact period, phosphate levels increased from 73.32 mg/L to 88.94 mg/L, at 2.5 % shell concentration, from 73.32 mg/L to 95.26 mg/L, at 5 % shell concentration and from 73.32 mg/L to 101.09 mg/L, at shell concentration of 10 % (Fig. 7). When compared with the control, phosphate levels in the aqueous solution after the 5 h contact time were observed to be significantly higher at shell concentration of 5 % and 10 % (P≤ 0.05).
In the treated raw shell, no remarkable decrease in nitrate levels was observed with time at shell concentration of 2.5 %. At shell concentrations of 5 % and 10 %, significant reductions in concentrations were observed after 3 h contact time, before increases were observed. After 3 h contact period, nitrate levels decreased from 231.47 mg/L to 56.83 mg/L and 45.59 mg/L, respectively (Fig. 8). After the 5 h contact period, when compared with the control, nitrate levels were observed to be significantly lower at shell concentrations of 5 % and 10 %, respectively ((P≤ 0.05).
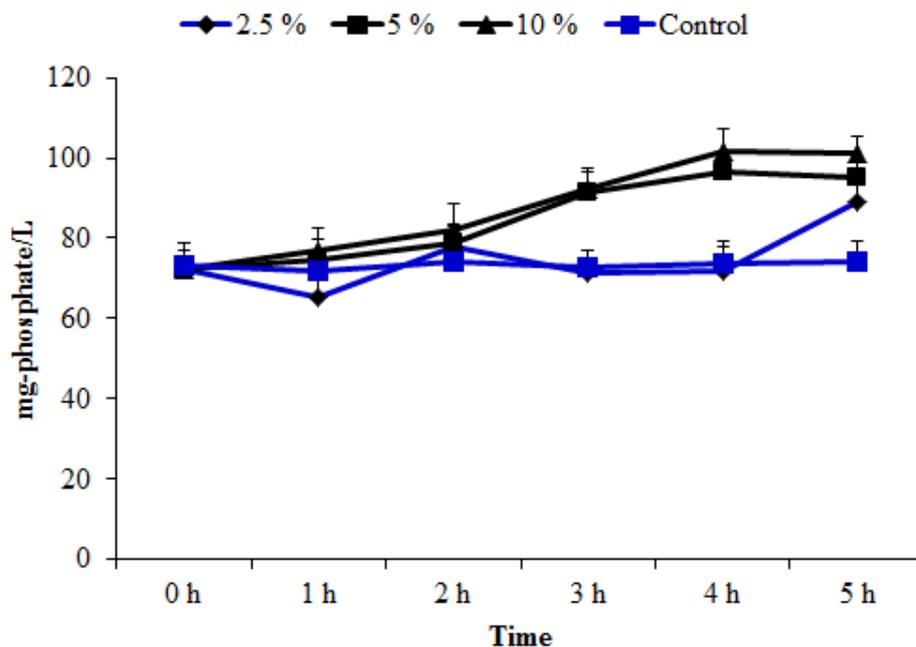
Fig. 7: Phosphate concentration in the aqueous solution when in contact with the treated raw shell
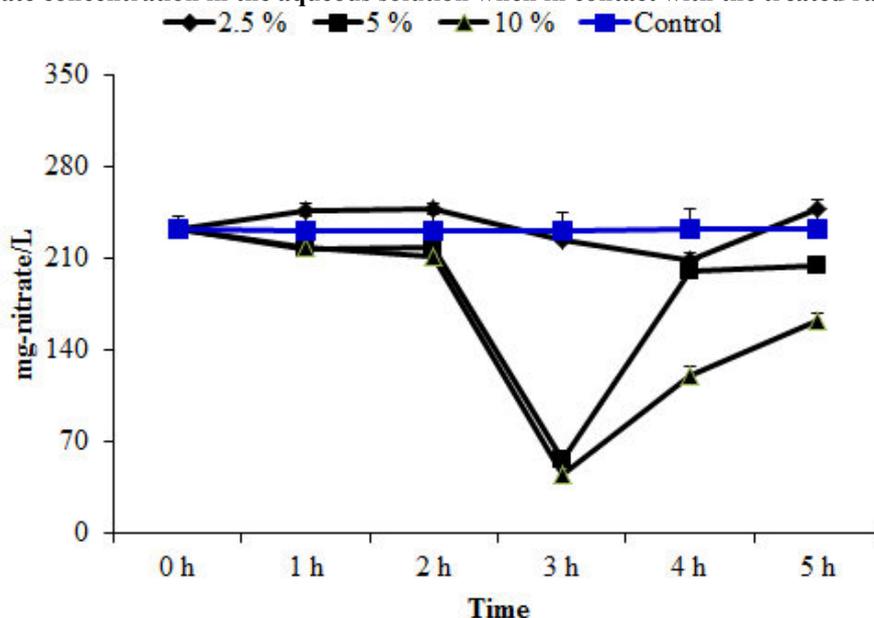

Fig. 8: Nitrate concentration in the aqueous solution when in contact with the treated raw shell
FTIR absorption studies were carried out on the samples to determine the functional groups that are present in each of them. The spectra are shown in Figures 9-12. A broad and intense, but medium absorption band range of 3215-3376 cm^{-1} was observed for the presence of primary amines while the medium band of 3266 cm-1 was an indication of the presence of an intermolecular bonded hydroxyl stretching vibrations. An alkenyl c-H stretching vibration was observed at 3073 cm^{-1}. The weak and medium absorption bands observed at 2926 and 2855 cm^{-1} shows the presence of alkyl c-H stretching vibrations. A C=O stretching vibration at 1713 was a medium absorption showing the presence of a ketone. An aromatic stretching vibration appeared at 1636 and 1456 cm^{-1}. A study band, which appeared at 1508 cm-1 is an indication of the presence of N-H binding for an amino group. A C-H binding vibration was observed at 1375 cm^{-1} while the C-O stretching vibration for esters, ethers or alcohols appeared at 1319 cm^{-1}. The absorption bands for C-N stretching vibrations for aryl ammis appeared as medium bands at 1244 and 1161 cm^{-1}. The weak absorption bands at 1055 and 1036 cm^{-1} might be due to the presence of C-O stretching vibration for secondary alcohols while the strong band of 896 cm^{-1} indicated that there is a 1,2,3-substituted C-H binding vibration.

With the treated raw sample, new absorption bands were observed at 1107, 1373, 1740 and 3009 cm^{-1} while the bands at 1319and 3073 cm^{-1} were absent. The medium band at 1107 cm^{-1} indicate the presence of C-O stretching vibration while the strong band at 1373 cm-1 shows the presence of an alkyl C-H binding vibration. A strong carbonyl C=O is seen at 1740 cm^{-1}. The new absorption bands indicate that oxidation might have taken place after treatment of the sample with the H_2SO_4. This could mean that more reactive centres were introduced on the sample surface (Figs. 9 and 10).
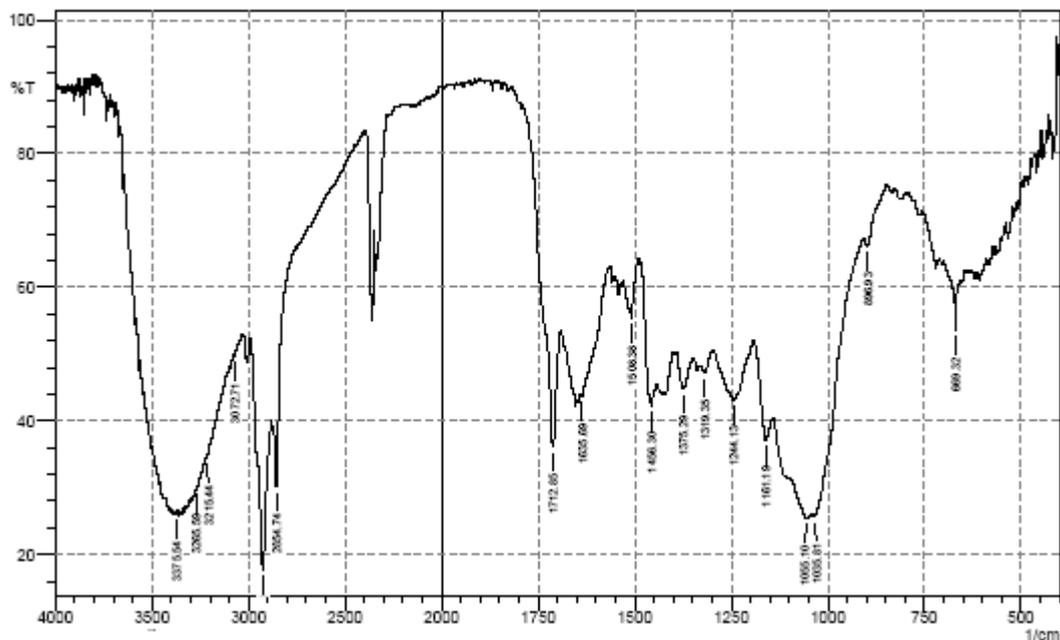

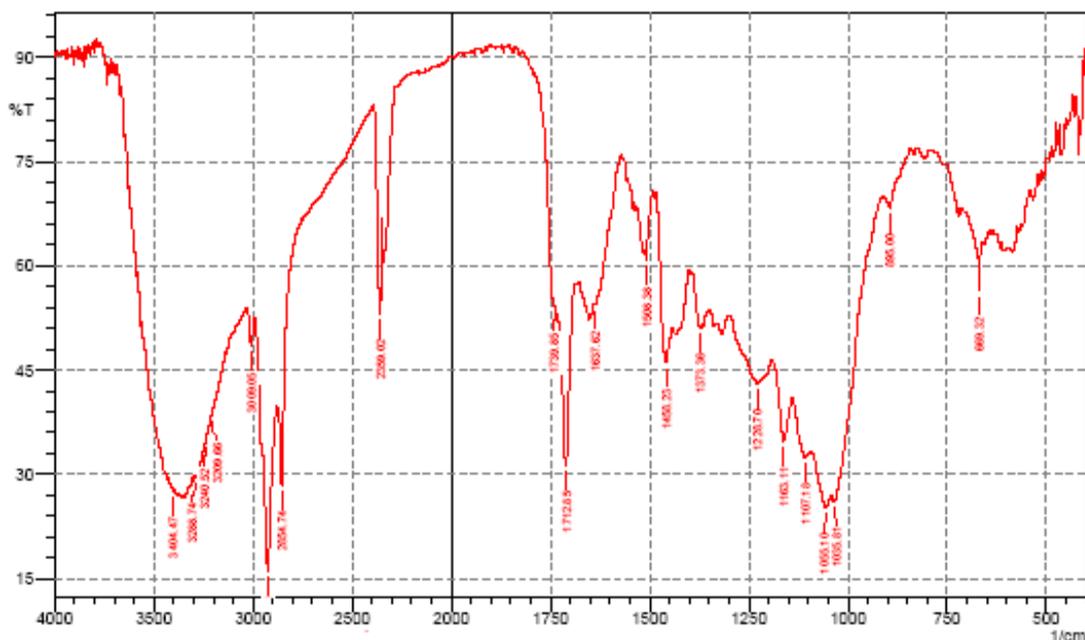**Fig. 9: FTIR of the untreated raw shell**

Fig. 10: FTIR of the treated raw shell

 With the carbonated untreated sample, the absorptions for amine and ketone were absent while new absorption band at 1117 cm^{-1} for C-O stretching vibration was observed to be present (Fig. 11). In the case of the carbonated sample, new bands were observed at 1109 and 1734 cm^{-1}, indicating that C=C and a carbonyl group were introduced on the surface of the sample (Fig. 12)
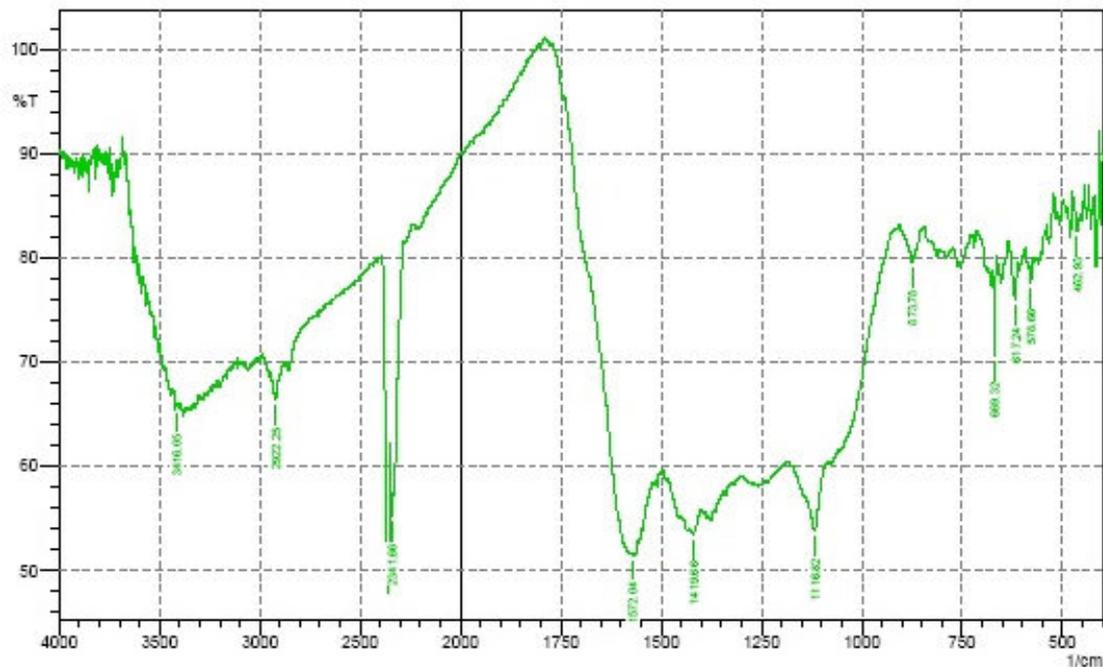
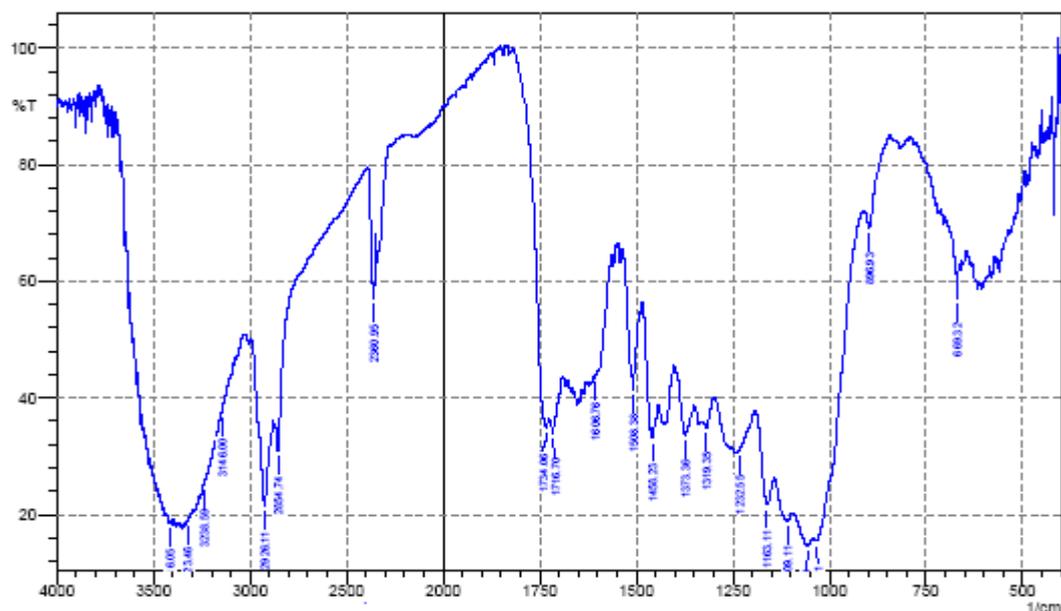**Fig. 11: FTIR of the untreated carbonated shell**

Fig. 12: FTIR of the treated carbonated shell

4. Discussion

In the present study, after 3 h contact time, there was a drastic decrease in removal rate of the nutrients. This was most evident in nitrate removal, with the trend irrespective of the form of shell that was used for the study. In a study Surchi (2011), when investigating the possibility of agricultural wastes as low cost adsorbents for lead removal, the removal rate of the metal was observed to gradually decrease with increase in contact time. It is hypothesised that during absorption, removal rate is higher when sorption sites are vacant after which when sites become reduced, there is reduced uptake (Mousavi *et al*., 2010; Surchi, 2011).

The quicker and higher nutrient absorption time attained at higher concentrations of the shells may have been due to an increase in the amount of available binding sites by the competing nutrient ions. Although the effect of initial nutrient concentration on absorption rate was not investigated in the study, it is reported that during absorption that more ions are left unabsorbed in solutions at higher concentration levels (Krsihnan and Anuirudhan, 2003; Surchi, 2011).

It could be deduced that at higher absorbent concentration, there is the likelihood of having a larger number of active sites for absorption. The trend observed in this study is in conformity with the observation of earlier investigators (Namasivayam and Sangeetha, 2005; Opeolu and Fatoki, 2012). In the study carried out by Namasivayam and Sangeetha, (2005) on the recovery of nitrate from wastewater by $ZnCl_2$ activated carbon from coconut coir pith, when the effect of absorbent dose in nitrate uptake was investigated, percentage removal was observed to increase with an increase in absorbent dose. Similarly, in a study on the dynamics of zinc sorption from aqueous matrices using plantain peel biomass Opeolu and Fatoki, (2012), adsorption was reported to show an increase with increase in adsorbent dosage. An increase in absorption with increasing adsorbent dosage is an indication for the provision for greater surface area available for adsorption. This may be due to increased active sites on the biomass. It is indicated that at higher adsorbent weight, there is the possibility for the presence of numerous unsaturated active sites for binding (Opeolu and Fatoki, 2012).

Also revealed in this study is the likelihood that the saturation time of 3 h for nitrate removal at absorbent concentration of 5 % and 10 %, after which no further nitrate absorptions take place, instead there were releases. This trend was evident in the different shell treatments used. At absorbent concentration of 2.5 %, significant nutrient removal was observed only after 5 h contact time. In the case of phosphate removal, no significant removal was observed in the presence of the different shell treatments. It was however observed that in cases were slight removals took place, the saturation point was found to be 1 h, after which there were increases in concentrations with increase in contact time.

Previous studies conducted on the effect of contact time for heavy metal with different initial concentrations and adsorbent doses revealed that a 5 h contact time was sufficient to attain equilibration (Krishnan and Anuirudhgan, 2003; Zvinowanda et al., 2009) . Although some studies (Krishnan and Anuirudhgan, 2003) have indicated that the saturation time of an absorbent is totally independent of the initial concentration, a contrary trend was observed in this study. The present study observed a shorter saturation time at higher absorbent concentration.

5. Conclusion

Although numerous adsorbents have been reported in literature, none of such studies have indicated the possible use of *Monodora myristica* shell, which is an agricultural waste for the removal of phosphate and nitrate from wastewater. This study, which used different modifications of the shell for investigation showed that the raw and carbonated forms of the shells were the most effective for nitrate removal from aqueous solution. None of the different modifications of the shell showed the potential for phosphate removal from aqueous solution. Also indicated in this study is a saturation time of 3 h for nitrate removal at shell concentrations of between 5 % to 10 % and a saturation time of 5 h at shell concentration of 2.5 %.

Although further studies still need to be carried out to ascertain the optimum conditions for the removal of nutrients from water by the *Monodora myristica* shell, the present investigation has given an insight into the potential use of the plant shell as a bio adsorbent in nitrate removal study from wastewater treatment systems.

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