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RESEARCH ARTICLE

Development of Biomass-Based Anion Exchanger for the Removal of Trace Concentration of Phosphate from Water

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Abstract

Aluminum loaded saponified mango waste *i.e.* Al(III)–SMW adsorbent, which functions as anion exchanger, was developed by loading Al(III) onto lime treated Mango waste biomass. The characterization of adsorbent was done by an Energy Dispersive X-ray (EDX) spectroscopy and chemical analysis techniques. Elemental analysis showed an exchange of Ca(II) or K (I) from SMW with Al(III) during loading reaction *via* cation exchange mechanism. Phosphate adsorption is strongly pH-dependent and maximum adsorption occurs at pH around 7-9. The maximum uptake capacity of Al(III)–SMW for phosphate was found to be 3.28 mg/g from the Langmuir isotherm model. The residual concentration of phosphate was sharply decreased by increasing the amount of Al(III)–SMW and reached less than 0.07 mg/L (from 9.7 mg/L) with the use of only 6 g/L of Al(III)–SMW whereas higher than 7 g/L successfully removed 100% of phosphate from water. The adsorbed phosphate was successfully desorbed and the adsorbent was regenerated using dilute alkali solution. Therefore, the Al(III)–SMW adsorbent investigated in this research work is expected to be a potential material for the treatment of water polluted with a trace amount of phosphate from aqueous solution.

Keywords: Mango waste, phosphate anion, Al(III) loading, elemental analysis, adsorption capacity

Introduction

Nowadays due to the continuous growth of industrialization, industrial wastewater containing toxic substances becomes an emerging issue for the environment based researcher that also inviting health risks to humans and an environmental risk to earth [1]. The trace amount of phosphorus is essential for the growth and development of organism but an excessive amount of phosphorus caused pollution [2]. Environmental Protection Agency (EPA) sets the maximum acceptable level of phosphorus in water as 0.1 mg/L [1]. Naturally, phosphorus exists in the form of its oxoanion (e.g. phosphate) in water. Phosphate and its compounds are extensively used in chemical fertilizer, soap, detergent, cosmetics, and beverages which produce wastewater containing a high concentration of phosphate [3-5]. The discharge of such wastewater into the water bodies such as river, lakes, pond, and streams potentially increase the phosphate level in water that disturbs the balance of aquatic organisms and affect water quality mainly through depletion of the dissolved oxygen levels and finally caused eutrophication [2,6,7]. Thus it is necessary to treat the wastewater containing excessive amounts of phosphate before discharge.

Several methods such as biological treatment, precipitation, crystallization, ion exchange, adsorption, and membrane filtration are reported for the treatment of phosphate polluted water [1,3]. The precipitation of phosphate was generally done by using the salt of trivalent metal like $FeCl_3$ and $AlCl_3$ [2]. Crystallization of phosphate in the form of

HAP (hydroxyapatite, $Ca_{10}(OH)_2(PO_4)_6$) according to reaction 'a' and MAP (magnesium ammonium phosphate, $MgNH_4PO_4.6H_2O$) according to the reaction 'b' were extensively used for the removal and recovery of phosphate from high concentration [2,8].

Although precipitation and crystallization are effective for the treatment of a high concentration of phosphate, these methods are not suitable for the treatment of trace concentration. The ion exchange and membrane filtration are effective techniques but these are a little bit expensive for water treatment [2,6,9]. Thus the development of low–cost adsorbent from agricultural waste biomass looks more promising. In the previous work, authors report effective removal of fluoride anions using Al(III) loaded orange waste biomass [10]. In present work natural anion exchanger was developed by loading Al(III) onto the polymer matrix of Mango waste biomass for the removal of phosphate from water. Furthermore, adsorption of phosphate onto Al(III) loaded Mango waste biomass was carried out batch-wise at different experimental conditions such as pH, initial phosphate concentration, adsorbent dosage, etc. The main objective of this study is to prepare the adsorbent by chemical modification of Mango waste biomass and evaluate its phosphate adsorption performance in the aqueous medium.

$$10\,Ca^{2+} + 2OH^- + 6\,PO_4^{3-} \rightleftharpoons Ca_{10}(OH)_2(PO_4)_6 \qquad \text{(HAP method)} \qquad \textbf{(a)}$$

$$Mg^{2+} + NH_4^+ + PO_4^{3-} + 6H_2O \rightleftharpoons MgNH_4PO_4 + 6H2O \qquad \text{(MAP method)} \qquad \textbf{(b)}$$

Materials and Methods

Chemicals and analysis

The chemicals and reagents used in this study were of pure reagent-grade or laboratory grades which were directly used without further purification. The stoke solution of phosphate (1000 mg/L) was prepared by using $Na_2HPO_4.2H_2O$ whereas Al(III) solution used for the loading reaction was prepared from its chloride salt. The pH of the solution was controlled by adding a small volume of 0.01M HCl or 0.01M NaOH and pH was measured by using pH meter. Phosphate concentration in the test solution before and after adsorption was analyzed spectrophotometrically using molybdenum blue method [11]. The elemental analysis of adsorbents was carried out using an energy dispersive X-ray spectrometer (EDX–8000).

Preparation of Al(III) loaded SMW

The waste biomass of Mango fruit employed in this study was kindly donated by the local mango juice vendor of the Kathmandu valley, Nepal. The mango waste is pectin rich biomass which contains 12-18% of pectin. For the preparation of adsorbent, the methyl ester part of Mango pectin was first converted into a carboxyl group by base hydrolysis then it was loaded with trivalent aluminum. The functional group modification and Al(III) loading onto mango waste biomass was done according to the procedure described else ware [10]. For this 100 g of mango waste was crushed with 0.5L of saturated lime water and shacked for 24 h at 30 ˚C. The methyl ester parts of mango pectin were hydrolyzed to convert into carboxyl groups by saponification reaction as shown in Scheme 1.

After this it was filtered, washed several times until neutral pH, and finally dried in an oven at 60-70 °C. The white product obtained in this way is termed as saponified mango waste and abbreviated as SMW hereafter. Loading of Al(III) onto SMW was done as follows. 5g of SMW (dry basis) was treated with 0.1M $AlCl_3$ solution and agitated for 24 h to load the Al(III) onto SMW *via* cation exchange reaction as shown also in Scheme 1. After 24 h, it was filtered washed several times with distilled water until the pH of the washing water reached around 7 and finally dried at 60–70 °C in the oven. The final product obtained in this way is known as Al(III) loaded saponified mango waste and abbreviated as Al(III)–SMW hereafter. The detailed methodology for the preparation of Al(III) loaded SMW and its phosphate adsorption is schematically shown in Figure 1.

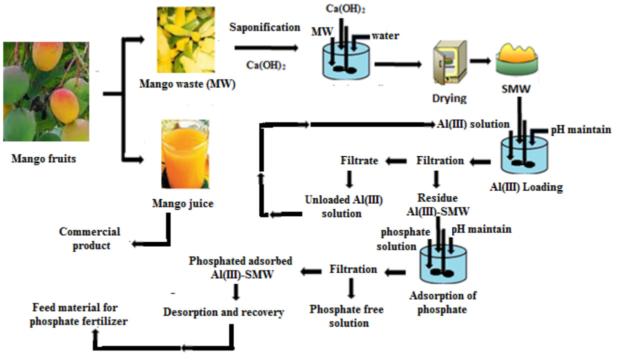
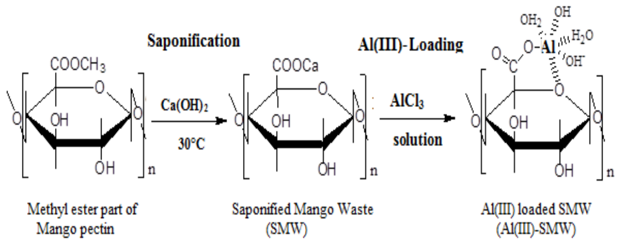


Figure 1: *Flowsheet showing the details of the adsorbent synthetic route and phosphate adsorption*

Characterization of the adsorbents

Fourier transmission infrared spectroscopy (FTIR) was used to analyze the surface functional group qualitatively and the elemental composition of adsorbent materials before and after adsorption was confirmed by energy-dispersive X-ray (EDX).



Scheme 1: *Synthetic route of Al(III) loaded saponified Mango waste (SMW) adsorbent from Mango waste biomass*

Batch wise studies

The equilibrium adsorption experiments were conducted at 25°C±2 at a solid-liquid ratio of 2 g/L. The typical adsorption test was carried out by shacking 50 mg of the adsorbent with 25 mL of 10 mg/L phosphate solution at various pH ranging from 2 to 12 and the mixture was shacked for 24 h to attain equilibrium. Then solid-liquid separation was done by filtration and phosphate concentration in the test solution was measured. The adsorption isotherm study was carried out by varying the concentration of phosphate at a solid-liquid ratio of 2 g/L. The effect of the solid-liquid ratio was determined by varying the concentration of adsorbent dosage from 0.2 to 8 g/L using 10 mg/L of phosphate solution. The % adsorption of phosphate and phosphate adsorption potential of Al(III)–SMW were calculated according to the following equations:

$$\%A = \frac{C_i - C_e}{C_i} \times 100 \qquad (1)$$

$$q = \frac{C_i - C_e}{W} \times V \qquad (2)$$

where C_i and C_e are initial and equilibrium concentration (mg/L) of phosphate respectively. V is the volume of phosphate solution (L) and W is the weight of the adsorbent (g) used in the adsorption experiments.

Results and Discussion

Spectroscopic characterizations of adsorbent

Analysis of functional group modification by FTIR spectroscopy

Mango waste is rich in pectin that can be modified to SMW by lime water which functions as anion exchanger once it is loaded with Al(III) ions. Since SMW is also a kind of Ca(II) type of pectic acid, HMW (H^+ form of SMW) was prepared by washing SMW with 0.1 M HCl to examined the effect of loaded metal ions in the FTIR spectra. Figure 2 shows the FTIR spectra of HMW and SMW. In the case of HMW, the broad and strong band around 3350 cm^{-1} corresponds to –OH stretching vibration of alcohols, phenols, carboxylic acids. The band at around 2912 cm^{-1} is due to C-H stretching vibration of $–CH_2$ group. The absorption band at 1716 and 1629 cm^{-1} are due to the stretching vibration of COO and C=O groups respectively. The bands observed at 1025 cm^{-1} was due to C–O stretching of alcohols and carboxylic acids. The peak observed at around 1716 cm^{-1} was disappeared in SMW. This indicates that carboxylic groups have been used for Ca(II) binding in SMW.

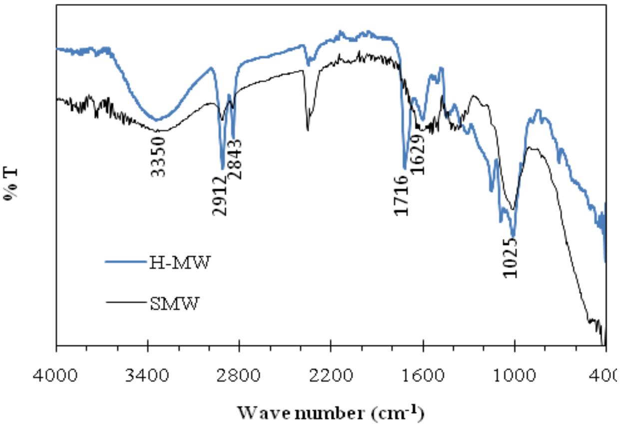


Figure 2: *FTIR spectra of HMW and SMW*

Elemental analysis using an energy dispersive X-ray (EDX) spectroscopy

Figure 3 shows the relationship between the intensity of various metal ions present in the sample of SMW before and after Al(III) loading at their respective binding energies values. In case of SMW, the metal ions at different energy values such as K (3.34 keV), Ca (3.70 and 4.02 keV), Ti (4.48keV), Mn (5.9 keV), Fe (6.4 keV and 8.06 keV), and Zn (8.64 keV) were observed. The result of this figure shows the comparatively higher value of intensity for Ca than other metals in non loaded sample indication that calcium is one of the major elements in SMW.

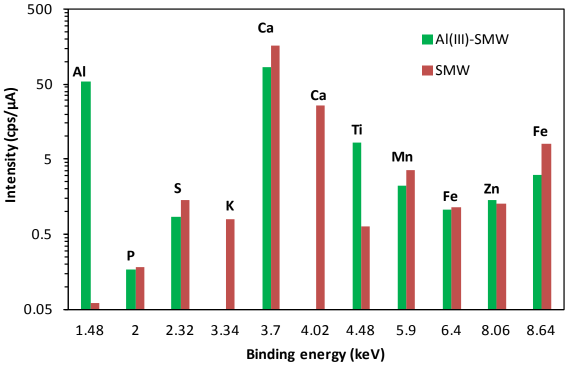
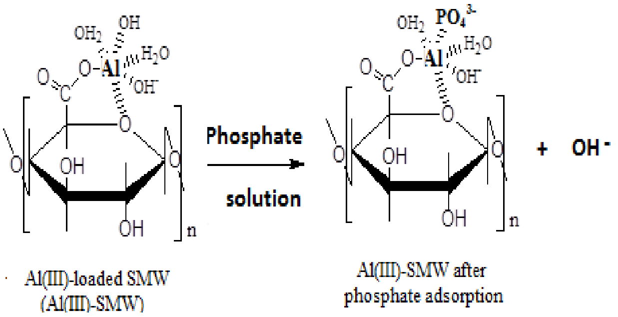


Figure 3: *EDX analysis of SMW and Al(III)–SMW*

After Al(III) loading, the K and Ca appeared at energy values 3.34 keV and 4.02 keV were completely disappeared whereas new element Al was detected with intensity 53.85 cps/µA at binding energy value of 1.48 keV. Moreover, the intensity of Fe (8.06keV) and Ca (3.70 keV) was observed to be decreased in the Al(III)–SMW. From these results it is clear that the loading of Al(III) onto SMW occurs by the substitution of Ca(II), K(I), or Fe(III) from SMW by Al(III) from aluminum chloride solution *via* cation exchange mechanism as shown in Scheme 1. Although the intensity of Ca(II) at 4.02 keV was decreased from 165.36 cps/µA to 54.28 cps/µA after Al(III) loading, complete replacement of Ca(II) from SMW by Al(III) does not occur in the present experimental condition which may be improved by repeated loading of Al (III). Similar types of loading mechanisms were also observed in the previous papers for the loading of Al (III), La(III), and Ce(IV) onto orange waste biomass [12,13].

Effect of pH and phosphate adsorption mechanism

Surface charges of the adsorbent and adsorbate speciation are very important controlling parameters in the adsorption process. Phosphate is normally existed as oxoanionic in aqueous solution. Percentage adsorption of phosphate at varying pH (2-12) using SMW before and after Al(III) loading is shown in Figure 4. It is clear from this figure that percentage adsorption of phosphate onto SMW was very low even at optimum pH (6.87% at pH 7.43) whereas it was improved significantly after Al(III) loading. In both cases, the % adsorption of phosphate increases from pH 2.01 to 7.27 and reached a maximum of around 7.83 then it was found to decrease with a further increase in equilibrium pH. The majority of phosphate is protonated at lower pH which is hardly adsorbed onto Al(III)–SMW adsorbent that ultimately decreases the phosphate adsorption percentage. The amount of hydroxyl ion increases with the rise of pH of the solution that strongly competes with phosphate anion for the adsorption site thus % adsorption of phosphate sharply decreases at higher pH. The pH of the solution after adsorption was found to be increased indicating that hydroxyl ions were released during the adsorption process according to Scheme 2. Although % adsorption of phosphate by SMW is low it also has



Scheme 2: *Inferred mechanism of phosphate adsorption using Al(III)–SMW from water*

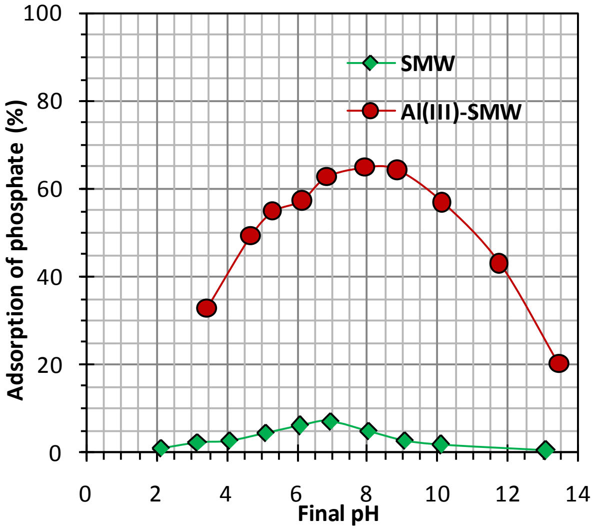


Figure 4: *Effect of pH for the adsorption of phosphate using SMW and Al(III)–SMW from water*

some affinity towards phosphate anion which is due to the electrostatic interaction between positively charge Ca(II) in SMW adsorbent and negatively charged phosphate anion as shown in reaction ‘c’.

$$\textbf{Polymer} \equiv \textbf{Ca}^{\delta+} + \delta^{-}\mathrm{PO}_4^{3-} \rightarrow \textbf{Polymer} \equiv \textbf{Ca}^{\delta+} .... \delta^{-}\mathrm{PO}_4^{3-} \qquad \textbf{(c)}$$

Mechanism of phosphate adsorption using Al(III)–SMW adsorbent can be described as follows. During Al(III) loading onto SMW, neutralization of all the positive charge of Al(III) by the big polymer of Mango pectic acid is impossible due to steric hindrance so that one or two positive charge of Al (III) is neutralized by mango pectic acid and rest are neutralized by anionic species such as hydroxyl ion in aqueous solution. These hydroxyl ions were inferred to be exchanged with phosphate anion according to the reaction shown in Scheme 2 which is further evidenced from the observation of raising equilibrium pH after phosphate adsorption. Similar types of adsorption behavior for phosphate were also observed by Biswas *et al.* 2008 for the adsorption of phosphate using Zr(IV) loaded SOW and Paudyal *et.al.* 2012 for the adsorption of fluoride using rare earth metal loaded SOJR adsorbents [14,15].

Adsorption isotherm of phosphate

To investigate the phosphate adsorption capacity of investigated Al(III)–SMW adsorbent, the experiment was carried out by varying the concentration of phosphate anion at 2 g/L and the result is presented in Figure 5. As seen from this figure that the adsorption of phosphate was found to increase sharply at a lower value of equilibrium phosphate concentration whereas it was found to adsorb slowly at higher concentrations suggesting the formation of monolayer phosphate on the surface of Al(III)–SMW adsorbent. For the evaluation of the best fit model for the phosphateadsorption onto this adsorbent, the experimental data were analyzed by using well known Langmuir and Freundlich isotherm models. Langmuir isotherm that describes the homogeneity in the surface adsorption sites is expressed according to equation 3 in the linear form as [16].

$$\frac{C_e}{Q_e} = \frac{1}{Q_{\max} b} + \frac{C_e}{Q_{\max}} \qquad \textbf{(3)}$$

where, C_e (mg/L) and Q_e (mg/g) are equilibrium concentration and amount of adsorption, respectively, while Q_{max} is the maximum loading capacity and b is the adsorption equilibrium constant related to the energy of adsorption. The maximum adsorption capacity (Q_{max}) and adsorption equilibrium constant (b) were determined from the slope and intercept of the linear plot of C_e/ Q_e *versus* C_e (Figure 6a). These values are listed in Table 1. Similarly, Freundlich isotherm that describes the adsorption on heterogeneous adsorption sites can be expressed mathematically by equation 4 in its linear form (Freundlich 1906) as [17].

$$\log Q_e = \log K_F + (1/n) \log C_e \qquad \textbf{(4)}$$

where K_F and n are Freundlich constants related to the adsorption capacity and adsorption intensity. The value of K_F and n were evaluated from the intercept and slope of the log Q_e *versus log C_e plot* as shown in Figure 6b**.** The evaluated values of Langmuir parameters are also listed in Table 1. The result of this table shows that the correlation regression coefficient obtained from the Langmuir isotherm model (R^2 = 0.99) is comparatively higher than the value obtained from the Freundlich isotherm model (which is only R^2 = 0.92). This strongly suggests that the experimental data were fitted better with Langmuir isotherm than the Freundlich isotherm model. Maximum adsorption capacity and adsorption equilibrium constant were evaluated to be 3.28 mg/g and 0.12 L/mg respectively using the Langmuir isotherm model. Furthermore, to investigate the suitability of an adsorption process, a dimensionless separation factor (R_L) was calculated using Langmuir equilibrium constant according to equation 5 expressed as [18].

$$R_L = \frac{1}{1+bC_i} \qquad \textbf{(5)}$$

where b and C_i are the Langmuir equilibrium constant and initial phosphate concentration, respectively. The value of R_L may be zero, one, or greater than one showing irreversible, linear, and unfavorable nature of adsorption reaction, respectively. From the experimental result for all the concentration tested it was found that the value of R_L is between zero and one *i.e* $0 < R_L < 1$, which suggests that the adsorption of phosphate onto Al(III)–SMW is favorable.

Table 1: *Langmuir and Freundlich parameters for the adsorption of phosphate using Al(III)–SMW*

Adsorbent	Langmuir isotherm model			Freundlich isotherm model		
	q_m (mg/g)	b (L/mg)	R^2	K_F (mg/g)	n	R^2
Al (III)–SMW	3.28	0.12	0.99	1.21	2.57	0.92

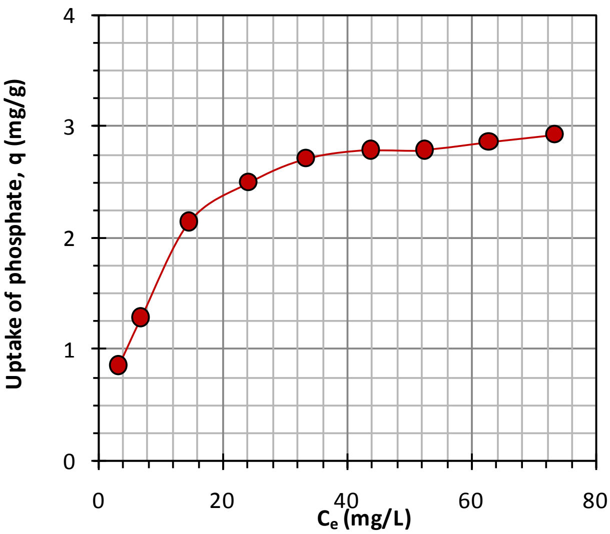


Figure 5: *Experimental adsorption isotherm plot of phosphate adsorption using Al(III)–SMW*

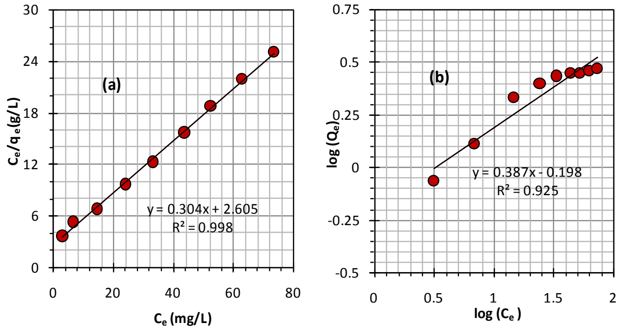


Figure 6: *Modeling of experimental data for the adsorption of phosphate onto the Al(III)–SMW using (a) Langmuir isotherm model and (b) Freundlich isotherm model*

Effect of adsorbent amount

The amount of adsorbent plays a very important role in the effective removal of phosphate from water. Figure 7 shows the relationship between the concentrations of phosphate ions remains after adsorption using Al(III)–SMW at the different solid-liquid ratio. As can be clear from the result of this figure that remaining concentration of phosphate after adsorption was found to be decreased from 9.70 mg/L to 5.28 mg/L by adding 1.2 g/L of Al(III)–SMW whereas phosphate concentration can be successfully lowered down to the WHO standard or Nepalese drinking water standard (5 mg/L) by using only 1.6 g/L of Al(III)–SMW adsorbent [19,20]. Besides, complete or 100% removal of phosphate from water can be successfully achieved at Al(III)–SMW dose higher than 7 g/L. The reason for increasing phosphate removal efficiency with the increase of Al (III)-SMW amount can be reasonably attributed due to the increase of several active adsorption sites for phosphate at a higher dosage. Therefore, the Al(III)–SMW adsorbent investigated in this study can be an excellent and potential material for the treatment of effluents containing trace concentration of phosphate ions from water.

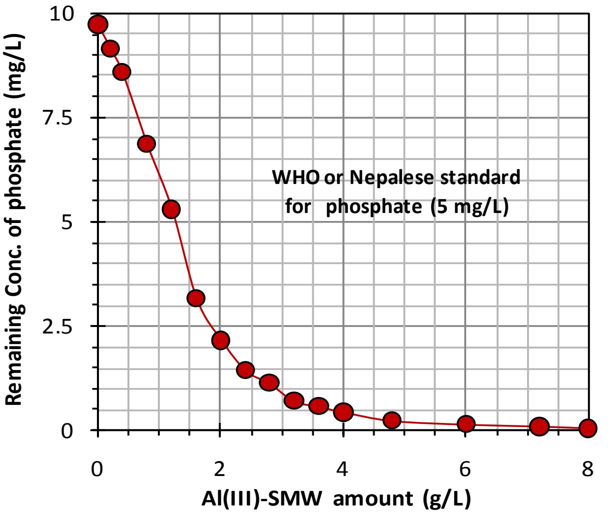


Figure 7: *Effect of Al(III)–SMW amount for the adsorptive removal of phosphate from water*

Desorption of loaded phosphate

The desorption of phosphate from Al(III)–SMW is important to determine the extent of its recovery and the regeneration of the adsorbents for repeated use. As shown in Figure 4, adsorption of phosphate on Al(III)-SMW is observed negligible at pH higher than 10, dilute alkaline solutions are the candidate for desorption of phosphate. Thus, the optimization of NaOH concentration was carried out by using different concentrations of NaOH (from 0.01 to 2 M) as shown in Figure 8. From the result, 1 M NaOH solution was found to be the optimum concentration to effectively desorb phosphate from the loaded

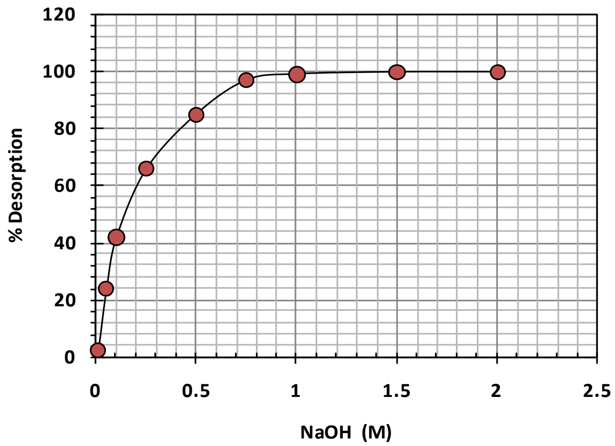


Figure 8: *Desorption of phosphate from phosphate loaded Al(III)–SMW using varying concentrations of NaOH solution.*

adsorbent. The desorption of phosphate is inferred to take place by the replacement of adsorbed phosphate by hydroxyl ions provided from NaOH solution as shown in reaction d.

Polymer≡Al–PO_4^{3-} + OH^- → **Polymer≡Al**–OH^- + PO_4^{3-} **(d)**

Conclusions

The adsorbent for phosphate ion removal was prepared by loading Al(III) onto the waste biomass of Mango fruit. Adsorption of phosphate onto Al (III) loaded Mango was successfully controlled by changing the pH and optimum adsorption occurs at pH around 7.5. The maximum adsorption capacity for phosphate on to this adsorbent was found to be 3 mg/g which is a satisfactory value from that have been reported in the literature. Phosphate concentration can be successfully lower down to the WHO standard or Nepalese government standard of phosphate for drinking water (5 mg/L) by using Al(III)–SMW higher than 1.6 g/L and 100% removal can be achieved by using more than 7 g/L. Phosphate loaded on Al(III) – SMW can be effectively desorbed by 1 M NaOH solution. Thus, the Al(III) loaded Mango waste adsorbent investigated in this study can be expected to be an excellent adsorbent for the treatment of water contaminated with a trace amount of phosphate from aqueous medium.

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