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REMEDIATION OF GROUNDWATER CONTAMINATION USING PLANT BASED BIOSORBENT MATERIALS

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ABSTRACT

This study reveals about the importance of using an eco-friendly approach for the removal of heavy metal ions from groundwater. The potential abilities of economically cheaper natural biosorbents (Banana and Citron peels) were studied at different intervals during the removal of heavy metal ions (Copper and Lead). The two metal ions studied were copper and lead by preparing aqueous solution and also from ground water sample. The biosorption of metals was studied by various techniques such as Atomic-absorption spectroscopy (AAS) and Fourier Transform Infra-Red Spectroscopy (FTIR). From the investigation, banana and citron peels have high ability to reduce the heavy metals in which citron peels have high potential to reduce the concentration of copper in aqueous (CuS) solution, where banana peels have more ability to reduce lead concentration in aqueous ($Pb(NO)_3)_2$) solution. The percentage of removal of Cu with citron is 87.6%, Pb with banana is 79.7%. Analysis of AAS and FTIR strongly evident that banana and citron peels plays actively in the removal of heavy metals Cu and Pb in water sample also, in which the percentage of removal of Cu with citron is 72.3% and Pb with banana is 71.6%.

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INTRODUCTION

Water as natural resource is absolutely essential to the human life and a precious resource for human civilization. The main source of water is from rain, lakes, streams, glacier, oceans and underground. The underground water is important because it is the most uniformly distributed and derived from sedimentary, igneous and metamorphic rocks of environment. The basic requirement for the availability of water is rock in the availability of porosity and permeability. The ground water makes up about 97% of the world's accessible fresh water reserve (Colonel *et al*., 2016). It has potability because it has gone through filtration through rocks. However, the rocks through which it has passed and its host rock usually have impact on its chemical composition (Offodile, 2002). The movement of groundwater along its course from recharge to discharge area always undergo some chemical and physical changes because the water comes in contact with pollutant such as heavy metals, other chemicals and organic sludge percolate from the soil and mix up with groundwater (Pacyna *et al*., 2001; Xu *et al.,* 2008; Mas Rosemal*et al*., 2010; Colonel *et al*., 2016). Urban areas are always associated with thick population and always depend on the groundwater for their day-to-day activities. Groundwater pollution and its consumption directly or indirectly act on the health problem of the population which depend on it (Mrinalini *et al*., 2015). It could cause eye irritation, nausea, vomiting, diarrhea, headache, respiratory diseases. Overconsumption and prolong consumption of pollutant groundwater will lead to kidney damage, liver damage, nervous system problems, bone problems, cancer and reproductive defects (Boulding, 1995; Wan Ngah*et al*., 2002; Gode and Pehlivan, 2006). The aim of this research work is to use plant based biosorbent material to remediate groundwater contamination caused by various environmental pollutant.

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MATERIALS AND METHODS

Materials: This study was conducted by using two plant based sorbents namely Banana peel and Citron peel on two heavy metals namely copper and lead. Copper (II), Sulphate

($CuSO_4.5H_2O$, RM 677 - Hi Media) and Lead Nitrate ($Pb(NO_3)_2$), Spectrum Analytical chemicals) was used to prepare a stock solution of 1000 ppm in deionized water respectively. 0.1 M of hydrochloric acid (HCl) and sodium hydroxide (NaOH) solutions were used to adjust the pH. The absorbance of heavy metals was measured by AAS (Model-Perkin Elmer Analyst 400) and FTIR (Bruker Alpha).

Sample collection: The ground water samples were collected from SemmankuppamCuddalore district. Fresh banana peels (Rasathali Variety) were collected from various house and fruit juice centers from the surrounding areas. In a similar manner, Citrus peels (Citron) were also collected.

Preparation of Aqueous Solution: The solutions were self-contaminated, 1000 ppm of stock solutions of copper sulphate was prepared by adding 3930 mg of Copper (II) Sulphate inonelitre of deionized water. Likewise, 1000 ppm of stock solutions of Lead Nitrate was prepared by adding 1598 mg of Lead Nitrate added to onelitre of deionized water. From the stock solution, 5 PPM concentration of heavy metal aqueous solution was prepared, 20 ml of stock solution from 1000 PPM solution in a beaker was taken and it was made up to 250 ml with deionized water (Both for Cu and Pb respectively).

Preparation of adsorbents: Banana peels (Rasthali) were washed thoroughly, allowed to dry in shade for 7-8 days until all the moisture content was lost from it and the colour change was observed from yellow to brownish black. Citrus peels (Citron) were washed repeatedly with distilled water to remove dust and soluble impurities then dried under shade for 15-20days. Once, all these collected samples were completely shade dried, they were subjected to powdered using domestic mixer or electric mixer and then they were sieved to obtain uniform particle size. The samples were stored very carefully in air tight Ziploc bags to prevent it from moisture and contamination.

Sample Name: The before treatment banana and citron peel named as A and B, where after treatment of copper, banana and citron peel named as A_1 and B_1, after treatment of lead, banana and citron peel named as A_2 and B_2, with respective to that after treatment of copper and lead in water sample, banana and citron peel named as C_1 and C_2. Water sample subjected for AAS analysis ofCu and Pb with Banana named as C_1, Cu and Pb with Citron named as C_2.

METHODS

Experimental Setup: 5 PPM solutions were prepared (Cu and Pb), we added 100 ml of solution in 16 conical flask for Cu and Pb respectively, where each flask has 100 ml of 5 PPM solution and then pH were adjusted using 0.1 M HCl and NaOH so that flask had pH 4 to 7 respectively. The flask were labeled with respect to their pH, RPM (100 RPM and 150 RPM), then 1 g of adsorbent (banana and citron peel) was added to all the set of flask for both Cu and Pb and it was subjected to a shaker (LARK Model) with contact time of 2 and 4 hours at room temperature. The experiments were conducted in two batches, first batch was conducted for copper, second batch was conducted for lead.

First batch

Biosorption of copper using banana peel: It contains 16 set of flasks, there were samples of each– Flask 1: pH 4, RPM 100, contact time 2 hours; Flask 2: pH 4, RPM 100, contact time 4 hours; Flask 3: pH 4, RPM 150, contact time 2 hours; Flask 4: pH 4, RPM 150, contact time 4 hours; Flask 5: pH 5, RPM 100, contact time 2 hours; Flask 6: pH 5, RPM 100, contact time 4 hours; Flask 7: pH 5, RPM 150, contact time 2 hours; flask 8: pH 5, RPM 150, contact time 4 hours; Flask 9:pH 6, RPM 100, contact time 2 hours; Flask 10: pH 6, RPM 100, contact time 4 hours; Flask 11: pH 6, RPM 150, contact time 2 hours; Flask 12: pH 6, RPM 150, contact time 4 hours; Flask 13: pH 7, RPM 100, contact time 2 hours; Flask 14: pH 7, RPM 100, contact time 4 hours; Flask 15: pH 7, RPM 150, contact time 2 hours; Flask 16: pH 5, RPM 150, contact time 4 hours (Table 1).

Table 1. Treatment of Cu (CuS) with Banana Peel using AAS

pH (6-8)	Absorbance (mg)	RPM	Contact Time (CT)
6	3184.23	100	2 h
6	3175.31	100	4 h
6	3165.18	150	2 h
6	3173.77	150	4 h
7	3329.62	100	2 h
7	3310.83	100	4 h
7	3318.03	150	2 h
7	3305.54	150	4 h
8	3510.24	100	2 h
8	3524.10	100	4 h
8	3540.50	150	2 h
8	3560.10	150	4 h

Biosorption of copper using citron peel: It also contains 16 set of flasks, there were samples of each– Flask 1: pH 4, RPM 100, contact time 2 hours; Flask 2: pH 4, RPM 100, contact time 4 hours; Flask 3: pH 4, RPM 150, contact time 2 hours; Flask 4: pH 4, RPM 150, contact time 4 hours; Flask 5: pH 5, RPM 100, contact time 2 hours; Flask 6: pH 5, RPM 100, contact time 4 hours; Flask 7: pH 5, RPM 150, contact time 2 hours; Flask 8: pH 5, RPM 150, contact time 4 hours: Flask 9: pH 6, RPM 100, contact time 2 hours; Flask 10: pH 6, RPM 100, contact time 4 hours; Flask 11: pH 6, RPM 150, contact time 2 hours; Flask 12: pH 6, RPM 150, contact time 4 hours; Flask 13: pH 7, RPM 100, contact time 2 hours; Flask 14: pH 7, RPM 100, contact time 4 hours; Flask 15: pH 7, RPM 150, contact time 2 hours; Flask 16: pH 5, RPM 150, contact time 4 hours (Table 2).

Table 2. Treatment of Cu (CuS) with Citron Peel using AAS

pH (6-8)	Absorbance (mg)	RPM	Contact Time (CT)
6	3430.48	100	2 h
6	3436.92	100	4 h
6	3432.84	150	2 h
6	3444.74	150	4 h
7	3337.23	100	2 h
7	3360.51	100	4 h
7	3359.69	150	2 h
7	3340.15	150	4 h
8	3362.10	100	2 h
8	3370.10	100	4 h
8	3375.40	150	2 h
8	3380.10	150	4 h

Second Batch

Biosorption of lead using banana peel: It contains 16 set of flask, there were samples of each – Flask1: pH 4, RPM 100, contact time 2 hours; Flask 2: pH 4, RPM 100, contact time 4 hours; Flask 3: pH 4, RPM 150, contact time 2 hours; Flask 4: pH 4, RPM 150, contact time 4 hours; Flask 5: pH 5, RPM

100, contact time 2 hours; Flask 6: pH 5, RPM 100, contact time 4 hours; Flask 7: pH 5, RPM 150, contact time 2 hours; Flask 8: pH 5, RPM 150, contact time 4 hours; Flask 9: pH 6, RPM 100, contact time 2 hours; Flask 10: pH 6, RPM 100, contact time 4 hours; Flask 11: pH 6, RPM 150, contact time 2 hours; Flask 12: pH 6, RPM 150, contact time 4 hours; Flask 13: pH 7, RPM 100, contact time 2 hours; Flask 14: pH 7, RPM 100, contact time 4 hours; Flask 15: pH 7, RPM 150, contact time 2 hours; Flask 16: pH 5, RPM 150, contact time 4 hours (Table 3).

Table 3. Treatment of Pb ($Pb(NO_3)_2$) with Banana Peel using AAS

pH (6-8)	Absorbance (mg)	RPM	Contact Time (CT)
6	1073.38	100	2 h
6	1085.25	100	4 h
6	1078.00	150	2 h
6	1089.45	150	4 h
7	1268.40	100	2 h
7	1263.18	100	4 h
7	1255.64	150	2 h
7	1275.00	150	4 h
8	1280.10	100	2 h
8	1282.15	100	4 h
8	1284.10	150	2 h
8	1285.70	150	4 h

Biosorption of lead using citron peel: It also contains 16 set of flask, there were samples of each – Flask 1: pH 4, RPM 100, contact time 2 hours;Flask 2: pH 4, RPM 100, contact time 4 hours;Flask 3: pH 4, RPM 150, contact time 2 hours;Flask 4: pH 4, RPM 150, contact time 4 hours;Flask 5: pH 5, RPM 100, contact time 2 hours;Flask 6: pH 5, RPM 100, contact time 4 hours;Flask 7: pH 5, RPM 150, contact time 2 hours;Flask 8: pH 5, RPM .150, contact time 4 hours;Flask 9: pH 6, RPM 100, contact time 2 hours;Flask 10: pH 6, RPM 100, contact time 4 hours;Flask 11: pH 6, RPM 150, contact time 2 hours;Flask 12: pH 6, RPM 150, contact time 4 hours;Flask 13: pH 7, RPM 100, contact time 2 hours;Flask 14: pH 7, RPM 100, contact time 4 hours;Flask 15: pH 7, RPM 150, contact time 2 hours; Flask 16: pH 5, RPM 150, contact time 4 hours (Table 4).

Table 4. Treatment of Pb ($Pb(NO_3)_2$) with Citron Peel using AAS

pH (6-8)	Absorbance (mg)	RPM	Contact Time (CT)
6	978.78	100	2 h
6	972.40	100	4 h
6	977.52	150	2 h
6	986.00	150	4 h
7	859.29	100	2 h
7	845.88	100	4 h
7	867.45	150	2 h
7	856.61	150	4 h
8	850.20	100	2 h
8	850.10	100	4 h
8	846.70	150	2 h
8	842.15	150	4 h

RESULTS AND DISCUSSION

Effect of pH on Cu and Pb adsorption: In the lower pH values, the absorbance was less mas compared to that of higher pH values (6 and 7) because at the lower values, the adsorption could have been compromised due to competitive interactions between the heavy metal ions and the hydrogen ions. Banana and citron peels were showing good results at high pH, which may be due to the various compounds and acids present. The overall best results were obtained at pH 6 and 7. The maximum adsorption of Cu and Pb using banana peel was attained at pH 7 and in Cu and Pb the maximum adsorption using citron peel was attained at pH 6.

Effect of RPM on Cu and Pb adsorption: RPM or agitation is very important to increase the efficiency of adsorbance and it helps to quicken the process. The best adsorbance was found at both 100 and 150 RPM. The maximum adsorbance of Cu using banana peel was found at 100 RPM, in Cu the maximum adsorbance using citron peel was found at 150 RPM, and in Pb using banana and citron peel the maximum adsorbance was found at 150 RPM.

Effect of contact time on Cu and Pb adsorption: Good and efficient results were obtained at contact time of both 2 and 4 hours. The efficient adsorption of Cu using banana peel was obtained at 2 hours, in Cu the efficient adsorption using citron peel was obtained at 4 hours, and in Pb using banana and citron peel the efficient adsorption was obtained at 4 hours.

Effect of removal of Cu and Pb: The removal of metal ion or adsorption of Cu is very efficient in citron peel, the total percentage of removal of Cu in banana is 84.72% while the percentage of removal of Cu in citron peel is 87.65%.The adsorption of Pb is very effective in banana peel, the total percentage of removal of Pb in citron is 61.7%, whereas percentage of removal of Pb in banana is 79.78% (Table 5).

Table 5. Aqueous solution analysis by AAS

Sample Name	Initial Concentration (mg/l)	Final Concentration (mg/l	Adsorption (mg/l)
A_1 (Cu)+Banana	3930	600.38	3329.62
B_1 (Cu)+Citron	3930	485.26	3444.74
A_2 (Pb)+Banana	1598	323.00	1275.00
B_2 (Pb)+Citron	1598	612.00	986.00

The total amount of metal ions in mg for Cu and Pb was measured by AAS, with respect to that percentage of removal of metal ion was calculated.

$$\text{Percentage of Removal} = \frac{\text{Initial Concentration} - \text{Final Concentration}}{\text{Initial Concentration}} \times 100$$

Table 6. Physico-chemical Parameters of water sample

Parameters	Desirable Limit	Permissible Limit
pH	6.5	8.5
Electrical Conductivity (mg/l)	500	1500
Total Hardness (mg/l	300	600
Turbidity (NTU)	5	10
TDS (mg/l)	500	2000
DO (mg/l)	5.5	15
Alkalinity (mg/l)	200	600
BOD (mg/l)	5	> 10
COD (ppm)	–	10
TSS (mg/l)	500	2000

Effect of contact time and RPM on Cu and Pb with Banana peel: The initial amount of Cu in water is 2.06 mg/l, it was reduced to 0.64 and 0.72 mg/l with respect to RPM 100 and 150 in 2 h, its percentage of removal is 68.9% and 65% and in 4 h, it is reduced to 0.71 and 0.66 mg/l with respecf'to RPM 100 and 150 and its percentage of removal is 65.5% and 67.9% Where the initial amount of Pb in water is 0.24 mg/l, it was reduced to 0.079 and 0.073 mg/l with respect to RPM 100 and 150 in 2 h and its percentage of removal is 67.08% and 69.58%, in 4 h, it is reduced to 0.081 and 0.068 mg/l with respect to RPM 100 and 150 and its percentage of removal is

Table 7. Treatment of water sample with banana and citron peel using AAS

Sample Name	Initial Conc. (mg/l)	Contact Time in h. (CT)	RPM	Final Conc. (mg/l)	Adsorption (mg/l)	Percentage of Removal
C_1	2.06	2 h	100	0.64	1.42	68.93
(Cu)+Banana			150	0.72	1.34	65.04
		4 h	100	0.71	1.35	65.53
			150	0.66	1.40	67.96
C_2 (Cu)+Citron		2 h	100	0.63	1.43	69.41
			150	0.64	1.42	68.93
		4 h	100	0.60	1.46	70.87
			150	0.57	1.49	72.33
C_1 (Pb)+Banana	0.24	2 h	100	0.07	0.16	67.08
			150	0.07	0.16	69.58
		4 h	100	0.08	0.15	66.25
			150	0.06	0.17	71.66
C_2 (Pb)+Citron		2 h	100	0.11	0.13	54.16
			150	0.10	0.14	58.33
		4 h	100	0.09	0.14	59.58
			150	0.09	0.14	60.41

Table 8. Water sample analysis using AAS

Sample Name	Initial Conc. (mg/l)	Final Conc. (mg/l)	Adsorption (mg/l)	Percentage of Removal
C_1 (Cu)+Banana	2.06	0.64	1.42	68.93
C_2 (Cu)+Citron		0.57	1.49	72.33
C_1 (Pb)+Banana	0.24	0.06	0.17	71.66
C_2 (Pb)+Citron		0.09	0.14	60.41

66.25% and 71.66%, this is due to the presence of alkane, hydroxyl and carboxyl groups in banana peel, it have huge potential to adsorb Pb than Cu (Table 7). The use of plant material has been carried out by many workers, aquatic duck weed by Abubacker and Sathya (2017) water lettuce by Abubacker and Sathya (2016), Pomegranate peel by El-AshtOukhy (2007), water melon shell by Koel Banerjee *et al.* (2012). Activated carbon also been recognized as a highly effective adsorbent for the treatment of heavy metals in waste water (Ho Yuh-Sha *et al*., 2005). Banana peels as biosobent material was used by Annadurai *et al*. (2003) Suggested that the proteins found in the banana peels has the capacity to bind heavy metals, it could be a possible mechanism by which banana peels adsorbent effectively absorbed Pb and Cu as reported by Jamil *et al*. (2008), Lianga Sha *et al.* (2010), Hossain *et al*. (2012), Saman Khan *et al*. (2013) and Sudha *et al*. (2015) used citron peels as biosorbent material to remove metals from contaminated water bodies, the present study also another evidence that Pb an Cu metal contaminations in ground water have been rduced to greater extend by banana and citron peels.

Effect of contact time and RPM on Pb and Cu with Citron peel: With respect to the contact time 2 h, the initial amount of Pb in water is 0.24 mg/l it is then reduced to 0.11 and 0.1 mg/l at the RPM 100 and 150, its percentage of removal is 54.1% and 58.3%, in 4 h at RPM 100 and 150 the concentration is reduced to 0.097 and 0.095 mg/l, its percentage of removal is 59.5% and 60.04%. In case of Cu at the RPM 100 and 150, the initial concentration is fall down from 2.06 mg/l to 0.63 and 0.64 mg/l in 2 h, its percentage of removal is 69.4% and 68.9%, where in 4 h, its percentage ofremoval is 70.8% and 72.3% at the RPM 100 and 150 in this its concentration reduced to 0.6 and 0.57 mg/l, it is may be due to the presence of aliphatic alkyl halide groups in citron it have ability to adsorb more Cu than Pb (Table 7). Therefore, the banana and citron peels used to treat water sample shows better reduction of metal ion Cu and Pb, percentage of removal of C, (Cu) +Banana is 68.93%, C2 (Cu) + Citron is 72.33%, C1 (Pb) + Banana is 71.66% and C2 (Pb) + Citron is 60.41% (Table 8).

FTIR Interpretation: There is a distinct change in the spectra resulting from the adsorption process A, when compare with A, there is a reduction of peaks corresponding to alkyl halides and alkanes (C-Br) and (C-H rock). The peak obtained in A_1 1015.54 cm^{-1}indicates there is a reduction and a distinct change in alkyl halides to aliphatic amines. There is a adsorption of nitro compounds in A_1 with respect to the peak 1535.19 cm^{-1} indicates the bonding of N-O asymmetric stretch. This shows the absorption of metal ions.

In contrast there is a change in the peak 1636.81 cm^{-1} in A_2 when compare with peak 1373.03 cm^{-1} in A, this shows that alkane group get transformed into 1 amine group due to the binding of metal ions with the hydroxyl and carboxyl groups present on pectin substance in banana. This shows better result in the absorption of Pb when compare with Cu. Spectrum reveals that B contain (C-Br Stretch) alkyl halides with respect to peak 615.81 cm^{-1}, aliphatic amines (C-N stretch) in the peak 1017.71 cm^{-1}alcohols, carboxyl groups (C-O stretch) in the peak 1310.89 cm^{-1}. Further there is a reduction in B_1, corresponding to the peak 1006.84 cm^{-1} and 1604 cm^{-1}, aliphatic amines (C-N stretch), amino groups (N-H bond). The peak get diminished in B_1 when compare with B, shows that there is a ligand binding between the alkane groups and metal ions, alcohols and carboxyl groups (C-O stretch) get diminished in the peak 1604.01 cm^{-1}.In other hand there is an increase in the peak at 1413.77 cm^{-1} and 1613.25 cm^{-1} shows the presence of (C-C stretch) aromatic and (N-H stretch) 1 amines in B_2. Similarly, the intensity of the peak 1023.03 cm^{-1} in C_1 shows the presence of (C-N stretch) aliphatic amines, (C=O) Carboxyl with respect to the peak 1633.02 cm^{-1}, the peak at 2921.18 cm^{-1} and 3335.41 cm^{-1} shows the presence of (C-H stretch) and (N-H stretch) of alkanes and 1,2 amides. Suddenly there is a distinct change in the spectra in C_2 at the peak 1010.31cm^{-1} and 1411.23 cm^{-1} shows the deformation of (C-O stretch) and (C-H bond) of alcohols, carboxylic acid and alkane groups. The analysis of FT-IR suggested that there is a absorption of compounds with change in functional groups shows the absorption of metal ions present in A, A_1, A_2, B, B_1, B_2, C_1, C_2 respectively.

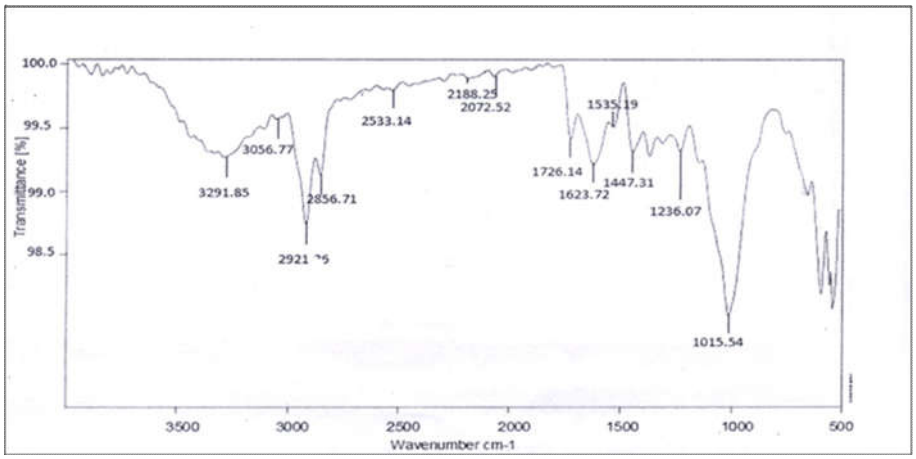

Fig. 1. represents FTIR spectra of pre-treated Banana peel (A)

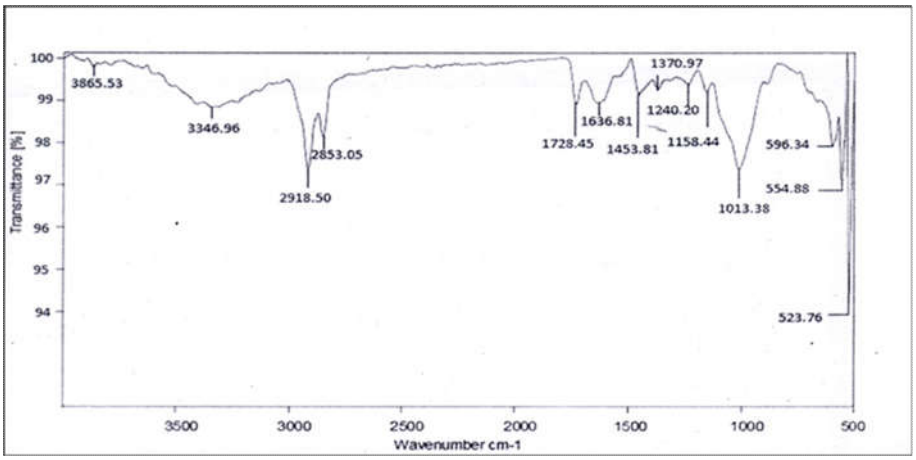

Fig. 2. represents FTIR spectra of treated Banana peel in Copper (A_1)

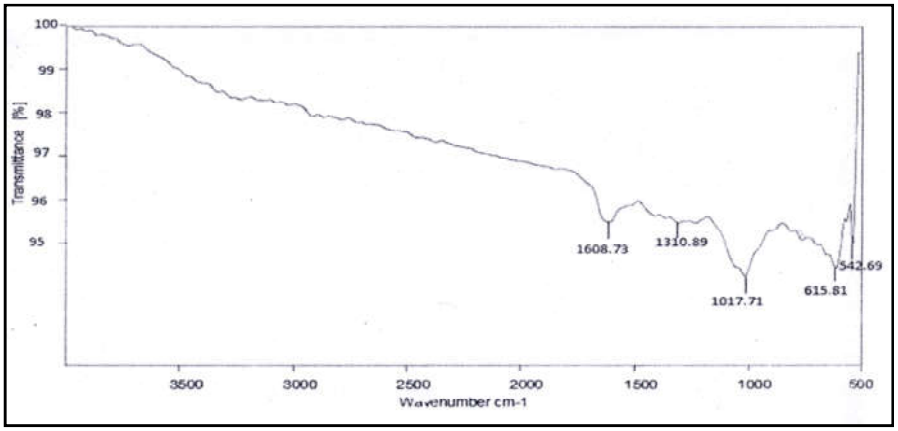

Fig.3 represents FTIR spectra of treated Banana peel in Lead (A_2)

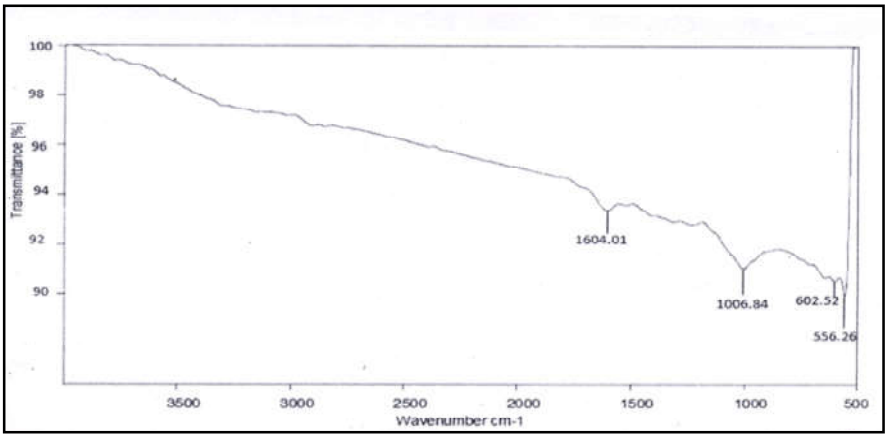

Fig. 4. Represents FTIR spectra of pre-treated Citron peel (B)

Fig. 5. Represents FTIR spectra of treated Citron peel in Copper (B_1)

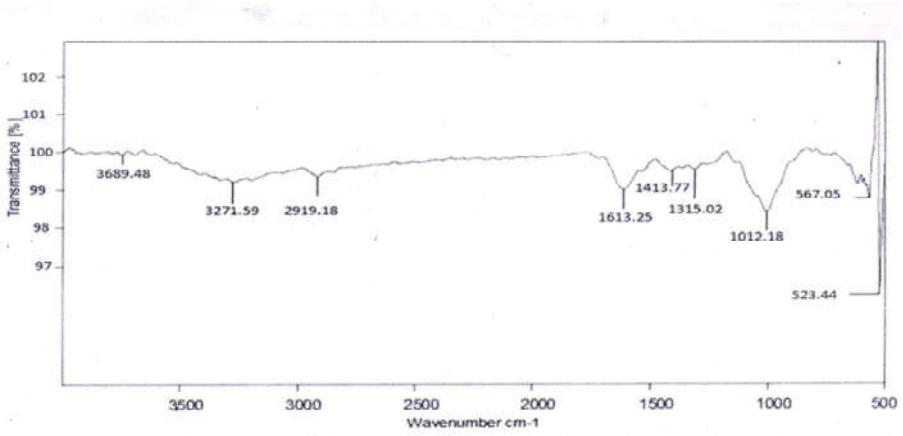

Fig. 6. Represents FTIR spectra of treated Citron peel in Lead (B_2)

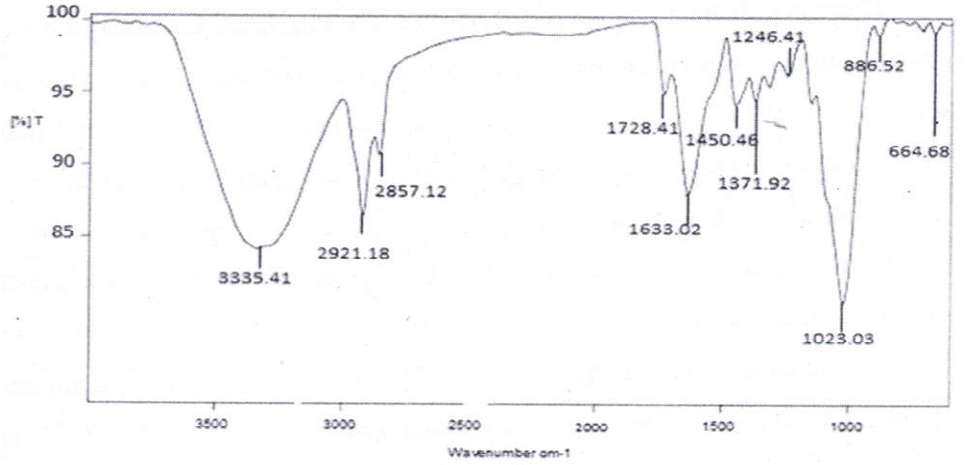

Fig. 7. Represents FTIR spectra of treated Banana peel in water (C_1)

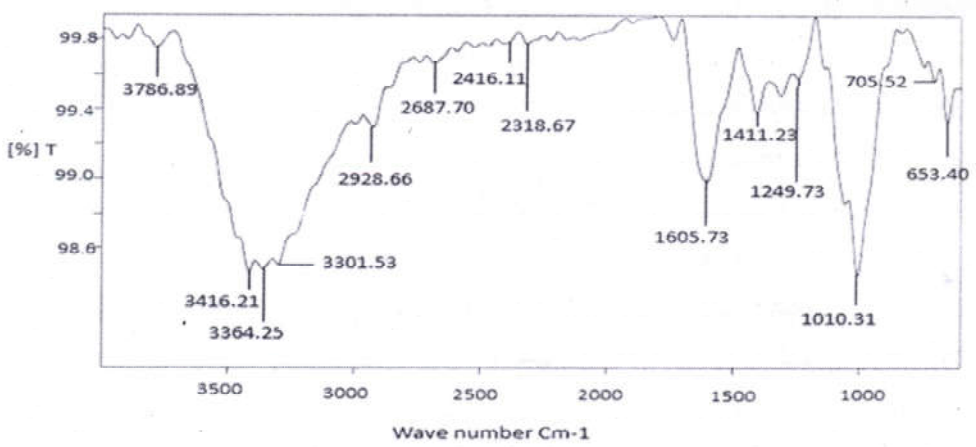

Fig. 8. Represents FTIR spectra of treated Citron peel in water (C_2)

The characterization result shows that banana peels act as good biosorbent in the adsorption of Pb whereas Citrus medica peels in Cu, because of the presence of high potential binding sites and functional groups. In the mean while there is a good reduction of metal ions inSemmandalam water sample with respective to banana and citron (*Citrus medica*) C_1, C_2.

Conclusion

Ground water is the largest and most important source of potable water for human consumption. It is also available everywhere can be drawn and made useful to day to day human activities. However, the ground water gets contaminated by the development of industries and their discharge, percolate and reach the ground water. The use of biosorbentmaterial of plant origin is a solution to remediate ground water contamination and it an ecofriendly approach.

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