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Original Article

Cadmium ion removal using biosorbents derived from fruit peel wastes

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Abstract

The ability of fruit peel wastes, corn, durian, pummelo, and banana, to remove cadmium ions from aqueous solution by biosorption were investigated. The experiments were carried out by batch method at 25°C. The influence of particle sizes, solution pH, and initial cadmium ion concentrations were evaluated on the biosorption studies. The result showed that banana peel had the highest cadmium ions removal followed by durian, pummelo, and corn peels at cadmium ions removal of 73.15, 72.17, 70.56, and 51.22%, respectively. There was a minimal effect when using different particle sizes of corn peel as biosorbent, while the particle size of the others had no influence on the removal of cadmium ions. The cadmium ions removal increased significantly as the pH of the solution increased rapidly from 1 to 5. At pH 5, the cadmium ions removal reached a maximum value. The equilibrium process was best described by the Langmuir isotherms, with maximum biosorption capacities of durian, pummelo, and banana peel of 18.55, 21.83, and 20.88 mg/g respectively. Fourier Transform Infrared Spectroscopy revealed that carboxyl, hydroxyl, and amide groups on the fruit peels' surface and these groups were involved in the adsorption of the cadmium ions.

Keywords: fruit peel, cadmium ions, biosorption, wastewater treatment, biosorbent

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1. Introduction

Heavy metal released in the environment has been increasing continuously as a result of industrial activities and technological developments and poses a threat to human health (Zhexian *et al.*, 2006). Cadmium is a priority toxic pollutant in wastewater that is introduced into water from metal mining, melting, plating, batteries, pesticides, oil paint, pigments, and alloys (Xiaomin *et al.*, 2007 and Guiqiu *et al.*, 2008). Large dosages of cadmium in humans can result in tubular and glomerular damage and cause proteinurea and anemia (Michael *et al.*, 2005). The presence of cadmium ions in the environment has become a potential threat to plant, animal, and human life due to their bioaccumulation tendency and toxicity and therefore they must be removed from municipal and industrial effluents before discharge into natural streams (Schiewer *et al.*, 2007).

The traditional heavy metal treatment includes chemicals and processes in surface chemistry such as precipitation, adsorption, membrane processes, ionic exchange, floatation, and others (Selatnia *et al.*, 2004 and Rosangela *et al.*, 2007). However, these techniques are rather expensive and are not selective enough to allow the recovery of very dilute heavy metals present in the effluent (Mohammad *et al.*, 2000). These disadvantages, together with the need for more economical and effective methods for the recovery of metals from wastewater, have resulted in the development of alternative separation technologies. One alternative is biosorption (Mack *et al.*, 2007 and Volesky, 2007), where certain types of biomass are able to bind and concentrate metals from even very dilute aqueous solutions. A biosorption process offers a number of advantages when compared to the conventional methods currently used. These include low operating costs, minimization of the volume of chemical and/or biological sludge to be handled, and high efficiency in detoxifying effluents (Marques *et al.*, 1999 and Cho and Kim,

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2003). The mechanisms of biosorption are generally based on physico-chemical interactions between metal ions and the functional groups present on the cell surface, such as electrostatic interactions, ion exchange, and metal ion chelation or complexation (Ozer *et al.*, 2004). Functional groups most commonly implicated in such interactions include carboxylate, hydroxyl, amine and phosphoryl groups present within cell wall components such as polysaccharides, lipids, and proteins (Dziwulska *et al.*, 2004). In this case, factors like pH, size of biosorbent, ionic strength and temperature influence the metal biosorption (Volesky and Schiewer, 1999).

Therefore, the purpose of this study was to explore the feasibility of using fruit peel waste for cadmium ion removal from aqueous solutions. Four fruit peel wastes of corn, durian, pummelo, and banana were used in this study. The effects of particle size and pH on their biosorption capacities were studied.

2. Materials and Methods

2.1 Biosorbents preparation

Fruit peel wastes of corn, durian, pummelo and banana were collected and were washed several times with deionized water, which was prepared by the technique of reverse osmosis, to remove heavy metals. The washed materials were cut into small pieces (1-2 cm) and were dried at 60°C in a hot air oven (Memmert Model 600) until they reached a constant weight, which was accomplished after 48 hrs. In the final stage these materials were ground and screened with sieves having the cut of size of > 850, 600-850, 425-600, 300-425, 212-300, and 150-212 μm.

2.2 Comparison of biosorption capacities

The percent of cadmium ions removal by fruit peel materials of corn, durian, pummelo, and banana peel were determined by contacting 0.1 g of 150-212 μm fruit peel materials with 100 ml of cadmium nitrate tetrahydrated ($Cd(NO_3)_2.4H_2O$, 50 mg/l; cadmium concentration 18.2 mg/l) on an shaker incubator (Vision Scientific VS-84805F) at 150 rpm for 24 hrs and the temperature was kept constant at 25°C. A pH value of 5.0 was maintained throughout the experiment by adding 0.1 N NaOH or 0.1 N HNO_3. At this pH value there would be little competition between cadmium ions and protons compared to lower pH values (Annadurai *et al.*, 2003), and therefore the height of metal uptake.

2.3 Effect of variation of particle size

The effect of particle size was determined by the agitation of 0.1 g of fruit peel materials of corn, durian, pummelo, and banana peel and 100 ml of $Cd(NO_3)_2.4H_2O$ solution concentration 50 mg/l using shakers with different particle sizes as follows: > 850, 600-850, 425-600, 300-425, 212-300, and 150-212 μm. Agitation contact time was kept for 24 hrs, which is sufficient to reach an equilibrium with a constant agitation speed of 150 rpm at 25°C. A pH value of 5.0 was maintained throughout the experiment by adding 0.1 N NaOH or 0.1 N HNO_3 before and during the experiment.

2.4 Effect of variation of the solution pH

The effect of initial solution pH was determined by the agitation of 0.1 g of fruit peel materials of corn, durian, pummelo, and banana peel and 100 ml of $Cd(NO_3)_2.4H_2O$ solution concentration 50 mg/l using a shaker at a different solution pH ranging 1 to 8. Agitation contact time was kept for 24 hrs, which is sufficient to reach equilibrium with a constant agitation speed of 150 rpm at 25°C. The pH was adjusted by adding 0.1 N NaOH or 0.1 N HNO_3 before and during each experiment. The pH was measured by using a pH meter (WTW. inoLab pH level 1).

2.5 Determination of equilibrium isotherms

The equilibrium isotherms were determined by contacting a constant mass of 0.1 g of 150-212 μm fruit peel materials of durian, pummelo, and banana peels with a range of different concentrations of $Cd(NO_3)_2.4H_2O$ solution 25-100 mg/l (cadmium concentration 9.1-36.4 mg/l). Agitation contact time was kept for 24 hrs, which was sufficient to reach equilibrium with a constant agitation speed of 150 rpm at 25°C. The pH values of 3.0 and 5.0 were maintained throughout the experiment by adding 0.1 N NaOH or 0.1 N HNO_3 before and during the experiment.

2.6 Cadmium analysis

After biosorption, biosorbents were separated from the solution by passing through a Watman 0.45 μm GF/C filter and the filtrate was subjected to residual cadmium concentration determination. The initial concentration and the residual concentrations of the cadmium ions were analyzed by Flame Atomic Absorption Spectrophotometer (Perkin Elmer AAnalyst 200).

The percentage of cadmium ions removal by fruit peel materials was calculated using the equation:

$$\% R = (Ci - C_e)*100/Ci \qquad (1)$$

Where;

R = cadmium ions removal;
Ci = initial cadmium ions concentration in solution used (mg/l);
C_e = cadmium ions concentration in solution (mg/l) at equilibrium

All experiments were performed in triplicate and mean values were presented. Standard deviation and analytical errors were calculated and the results were shown as the means with relative standard deviations of less than 8%.

2.7 FT-IR spectroscopy

The samples of the biomass before and after cadmium ions biosorption were analyzed with a FT-IR (Perkin Elmer System 2000) spectrometer under ambient conditions.

3. Result and Discussion

3.1 Comparison of biosorption capacities

The cadmium ions removal by fruit peel materials is shown in Figure 1. The experimental results indicated that all fruit peel materials adsorbed cadmium ions at different cadmium ions removal level. Banana peel had the highest cadmium ions removal of 73.15% followed by durian, pummelo, and corn peel at cadmium ions removal of 72.17, 70.56 and 51.22%, respectively. Banana, durian, and pummelo peel were selected for isotherm studies because of their higher mechanical strength compared to corn peel.

3.2 Effect of variation of particle size

The percentage of cadmium ions removal of fruit peel materials at different particle sizes is shown in Figure 2. It was seen that, as the corn peel size increases, the percentage of cadmium ions removed drops from around 51% for the 150-212 μm fraction to 31% for the > 850 μm fraction. This was probably due to an increase in the total surface area, which provided more sorption sites for metal ions (Ho *et al.*, 2001). The particle size of the others had no influence on the removal of cadmium ions. The higher biosorption level achieved by smaller particle size of the biosorbents may not be connected to the fact that smaller particle sizes give large surface areas. Although this was contrary to the expectation for an intraparticle diffusion controlled process (Demirbas *et al.*, 2004), it is necessary to point out that the two sizes of biomass were actually of the same thickness (dimension, which determines the diffusion distance). This was due to the size grading of ground biomass particle by standard sieves, which works on the length and width dimensions (Eneida *et al.*, 2002). This behavior has been reported by others (Kuyucak and Volesky, 1989), although Leusch *et al.* (1995) showed that larger biomass particles of *Sargassum fluitans* and *Ascophylum nodosum* had higher metal uptake than smaller particles in the case of cadmium, copper, nickel, lead, and zinc. This is possibly due to the thickness of corn peel being smaller than the others. Then, the influence of biosorbent size on metal uptake seems to be a function of both the type of biomass and the metal ion. The reason to explain this phenomenon seems unclear at this point. For this study it might be necessary to uncover the particle size effect. However, it might reach a ceiling limit of biosorption.

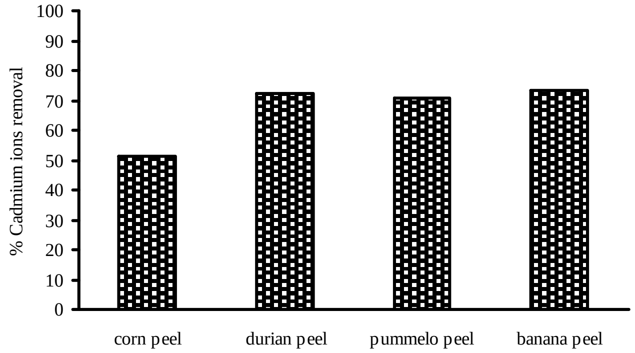

Figure 1. Comparison of percent cadmium removal by biosorbents derived from different fruit peel wastes Conditions: biosorbent 0.1 g; cadmium concentration 18.2 mg/l; particle size, 150-212 μm; contact time, 24 hrs; pH, 5; 150 rpm; 25+2°C.

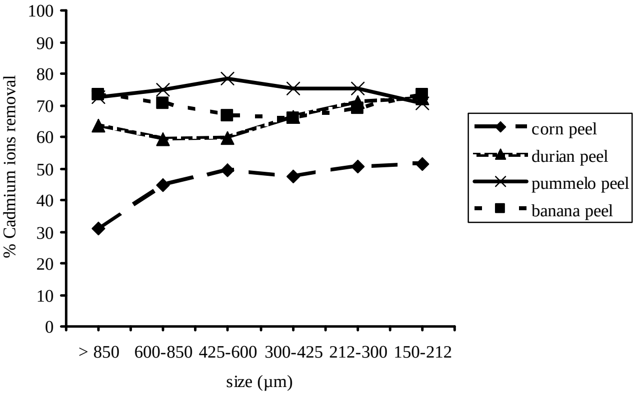

Figure 2. Effect of variation of particle sizes of biosorbents on percent cadmium removal Conditions: biosorbent 0.1 g; cadmium concentration 18.2 mg/l; contact time, 24 hrs; pH, 5; 150 rpm; 25+2°C.

3.3 Effect of variation of the solution pH

The variation of the solution pH is the one of the most important factors in the biosorption of the metal ions (Yu and Kaewsarn, 2002). This factor is capable of influencing not only the binding site dissociation state, but also the solution chemistry of the target metal in terms of hydrolysis, complexation by organic and/or inorganic ligands and redox potentials (Fiol *et al.*, 2006). Most research conducted on heavy metal biosorption indicates that the decrease in ion biosorption at acid pH may be due to the increase in competition with protons for active sites (Yu *et al.*, 1999, and Kaewsarn and Yu, 2001). At alkaline pHs, however, other effects may arise that also alter the process, such as the predominant presence of hydrated species of heavy metal, changes in surface charge or the precipitation of the appropriate salt (Blazquez *et al.*, 2005).

The effects of pH solution on the biosorption of cadmium ion by using fruit peel materials were studied and results are shown in Figure 3. The influence of the pH value was very similar for the four fruit peel materials employed. There was an increase in cadmium ions removal with increas-

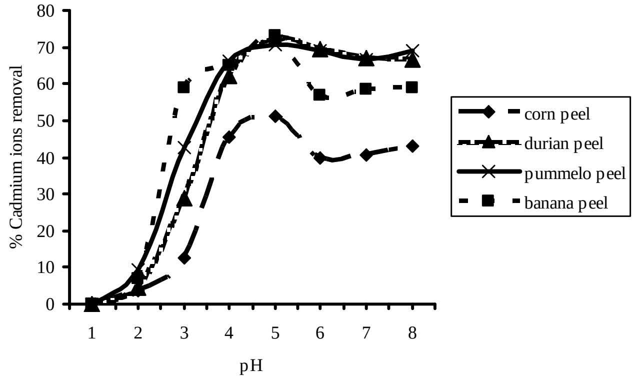

Figure 3. Effect of variation of solution pH on percent cadmium removal by biosorbents derived from different fruit peel wastes Conditions: biosorbent 0.1 g; cadmium concentration 18.2 mg/l; particle size, 150-212 µm; contact time, 24 hrs; 150 rpm; 25+2°C.

ing pH from 1-5. The cadmium ions removal increased rapidly. At pH of around 5, the cadmium ions removal leveled off at a maximum value. The biosorption of cadmium in a very highly acidic solution (pH 1) was observed to be negligible. According to several authors (Chang *et al.*, 1997 and Cruz *et al.*, 2004) the biosorption below pH 2 was slightly due to the competition of hydrogen ions for the active sites. It is clear that cadmium ions were effectively adsorbed in the pH 4-8 and the maximum biosorption of cadmium ions on fruit peel materials occurred at pH 5. The dependence of percent cadmium removal on pH is similar to the cadmium ions sorption on *Hydrilla verticillata*, and Cystine-modified biomass (Bunluesin *et al.*, 2007 and Junxia *et al.*, 2007). Then the biosorption capacity of cadmium ions decreases as the pH continues to increase. Due to the fact that proton (H^+) vies with cadmium ions in lower pH, the sorbent surface takes up more H^+, consequently reducing cadmium ions bind on the sorbent surface. In higher pH, the sorbent surface takes more negative changes, thus attracting greater cadmium ions. However, with a further increase in pH, the formation of anionic hydroxide complexes decreases the concentration of free cadmium ion, and thereby the biosorption capacity of cadmium ion decreased (Stephen and Sulochana, 2004).

3.4 FT-IR analysis

The FT-IR spectra of fruit peel materials before and after cadmium ion biosorption are shown in Figure 4 and 5. These spectra were obtained from scanning in the range of 400-4000 cm^{-1}. The influences of the peaks were very similar for the four fruit peel materials. The band of corn, durian, pummelo, and banana peel represented overlapping of O-H and N-H stretching vibration at 3388, 3369, 3365, and 3388 cm^{-1}, respectively. The band represented C=O stretching vibration of carboxylic acids at 1734, 1741, 1734 and 1731 cm^{-1}, respectively. The significant shifts of these specific peaks to the higher wave number after the cadmium ions biosorption suggested that chemical interactions between the cadmium ions and the amide groups occurred on the biomass surface. The influences of the shift peaks were very similar for the four fruit peel materials. The band of corn, durian, pummelo, and banana peel shifted to 3413, 3416, 3417, and 3416 cm^{-1}, respectively, and the band of corn and pummelo peel shifted to 1727 and 1737 cm^{-1}, respectively, indicating that hydroxyl, carboxyl, and amide groups were involved in the biosorption. The spectra of banana and durian peel at 1741 and 1731 cm^{-1}, respectively became smoother and a

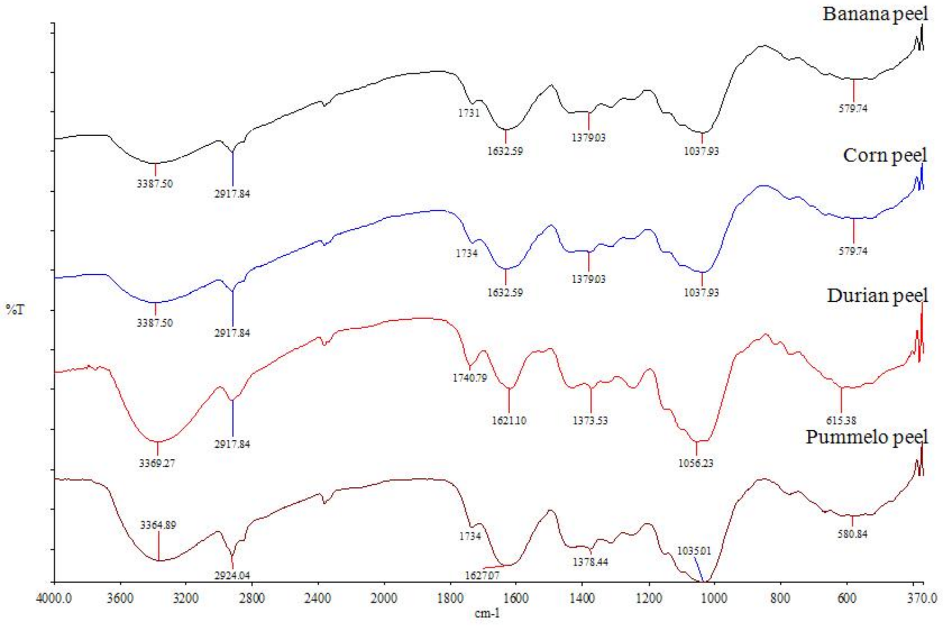

Figure 4. FT-IR of blank biosorbents derived from fruit peel wastes Conditions: particle size, 150-212 µm

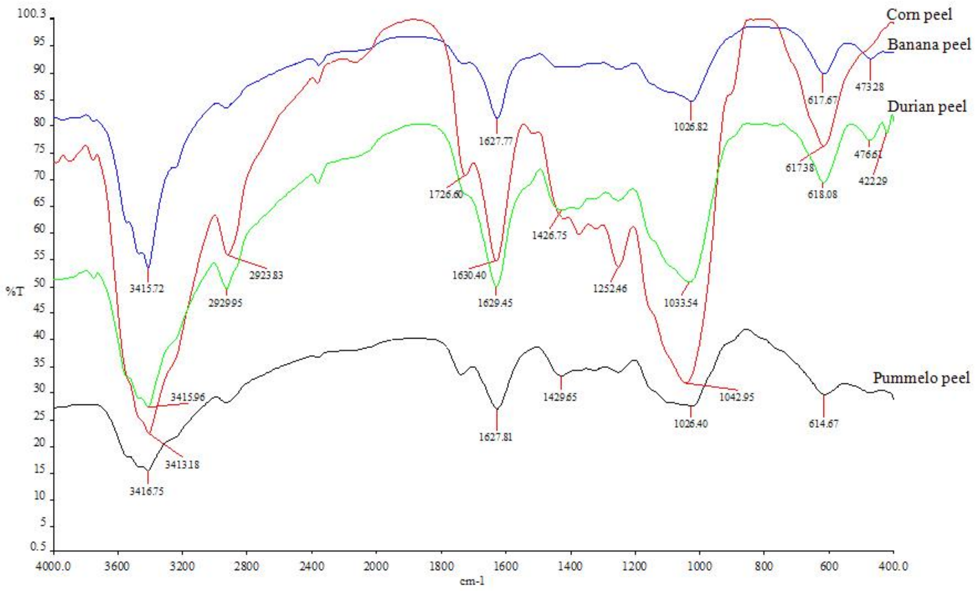

Figure 5. FT-IR of the biosorbents bound with cadmium ions Biosorption conditions: biosorbent 0.1 g; cadmium concentration 18.2 mg/l; particle size, 150-212 µm; contact time, 24 hrs; pH 5; 150 rpm; 25±2°C.

band of pummelo and banana peel at 1430 and 1427 cm^{-1}, respectively, appeared, which would result from the complications of cadmium ions with the functional groups from protein. These results indicated that carboxyl, hydroxyl and amide groups on the fruit peels' surfaces were involved in the biosorption of the cadmium ions.

3.5 Equilibrium isotherms

Biosorption isotherms can be generated based on numerous theoretical models where Langmuir and Freundlich models are commonly used to fit experimental data when solute uptake occurs by a monolayer biosorption (Yu and Kaewsarn, 1999 and Christian *et al.*, 2003). Langmuir isotherm assumes monolayer biosorption, and is presented by the following equation:

$$q_e = q_{max} bC_e / (1+bC_e) \qquad (2)$$

And the Freundlich isotherm has the form

$$q_e = K_F C_e^{1/n} \qquad (3)$$

Where q_e and q_{max} are the equilibrium and maximum uptake capacities (mg/g biosorbent); C_e is the equilibrium concentration (mg/l solution); b is the equilibrium constant; K_F and n are Freudlich constants characteristic of the system.

The linearization of Langmuir and Freudlich isotherms for the biosorbent from fruit peel materials are presented in Figure 6 and 7, respectively. The model parameters are tabulated in Table 1. The experimental data could better be described by the Langmuir isotherm than the Freudlich isothem. The regression coefficients (R^2) were 0.99, 0.97, and 0.94 for durian, pummelo, and banana peels, respectively. In contrast, the Freudlich isothem models were less precise, with a lower R^2 value of durian (0.81), pummelo (0.88), and banana peels (0.76) at pH 5. Based on the Langmuir isotherm, the maximum uptakes of cadmium were 18.55, 21.83, and 20.88 mg/g for durian, pummelo, and banana peels, respectively, arranged as durian peel < banana peel < pummelo peel. In addition, the maximum biosorption capacities for cadmium ion are obtained in this study using different low-cost biosorbents reported in the literature. The results show that the biosorption capacities for cadmium ion using fruit peel wastes were greater than the ones, which had been found using other adsorbents listed in Table 2. The solution pH had a significant effect on biosorption equilibrium; at pH 5 the uptake capacity was consistently higher than at pH 3 (Table 1). Similar results were observed for orange peel (Schiewer *et al.*, 2008), lemon, and grapefruit peels (Schiewer and Santosh, 2007). This occurred because of a decrease in proton concentration at pH 5 and thus decreased the competition between protons and cadmium ions for the same binding site (Kaewsarn and Yu, 2001).

4. Conclusions

In this study, the use of fruit peels of corn, durian, pummelo, and banana peel, which are waste products from agriculture, is environmental friendly and shows they can adsorb cadmium ions from aqueous solutions. The fruit peel material is a potential candidate for biosorption and further studies will help to evaluate the economic use of this biosorbent. The biosorption capacity was dependent on the type

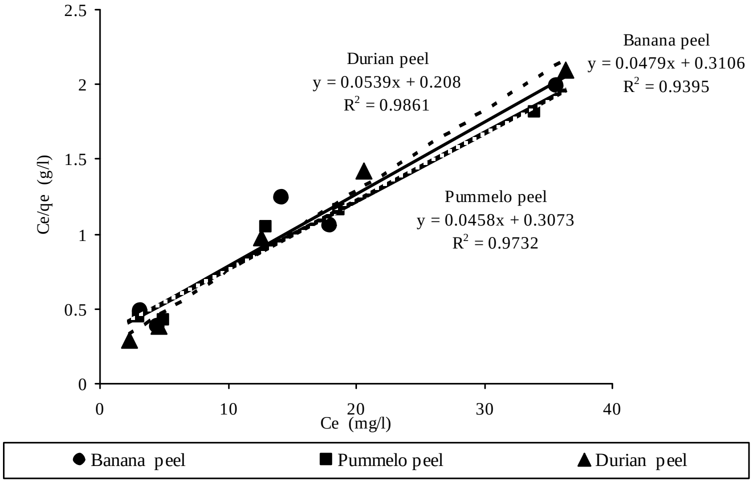

Figure 6. Comparison of linearity of adsorption isotherm of biosorbents derived from different fruit peel wastes based on Langmuir model Conditions: biosorbent 0.1 g; cadmium concentration 9.1-36.4 mg/l; particle size, 150-212 µm; contact time, 24 hrs; pH 5; 150 rpm; 25±2°C.

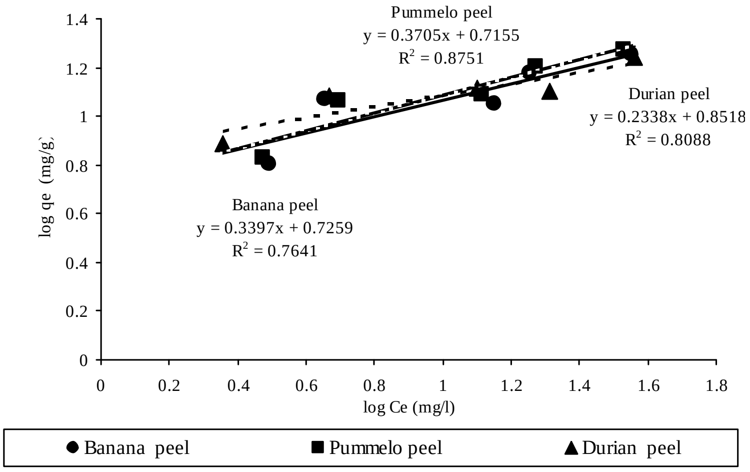

Figure 7. Comparison of linearity of adsorption isotherm of biosorbents derived from different fruit peel wastes based on Freundlich model Conditions: biosorbent 0.1 g; cadmium concentration 9.1-36.4 mg/l; particle size, 150-212 µm; contact time, 24 hrs; pH 5; 150 rpm; 25±2°C.

Table 1. Langmuir and Freundlich isotherm constants for cadmium ions adsorption onto adsorbents derived from various fruit peel wastes.

Biosorbent	Langmuir model			Freundlich model		
	b (l/mg)	q_{max} (mg/g)	R^2	k_f (l/g)	n	R^2
pH 3						
Durian peel	0.033	16.45	0.72	1.46	2.03	0.88
Pummelo peel	0.072	19.23	0.93	3.27	2.53	0.94
Banana peel	0.088	20.20	0.75	4.81	3.22	0.72
pH 5						
Durian peel	0.259	18.55	0.99	7.11	4.28	0.81
Pummelo peel	0.149	21.83	0.97	5.29	2.84	0.88
Banana peel	0.154	20.88	0.94	5.32	2.94	0.76

Table 2. Biosorption capacities of various biosorbents for cadmium ions.

Biosorbent	Biosorption capacity (mg/g)	Reference
Durain peel	18.55	This study
Pummelo peel	21.83	This study
Banana peel	20.88	This study
Papaya wood	17.22	Asma *et al.*, 2005
Cystine-modified biomass	11.63	Yu *et al.*, 2007
H. verticillata	15.00	Bunluesin *et al.*, 2007

of biosorbents and solution pH. There was minimal effect when using different particle sizes of corn peel as biosorbent while the particle size of the others had no influence on the removal of cadmium ions. This is possibly due to the thickness of corn peel being less than the others. At the pH of around 5, the cadmium ion removal leveled off at a maximum value. The equilibrium process was best described by the Langmuir isotherms model, with maximum biosorption capacities for durian, pummelo and banana peel of 18.55, 21.83, and 20.88 mg/g respectively. FT-IR spectroscopy revealed that carboxyl, hydroxyl, and amide groups on the fruit peels surface were involved in the biosorption of the cadmium ions.

Acknowledgments

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