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REVIEWS

A review on economically adsorbents on heavy metals removal in water and wastewater

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Abstract Heavy metals contamination in water has been an issue to the environment and human health. The persisting contamination level has been observed and concerned by the public due to continuous deterioration of water quality. On the other hand, conventional treatment system could not completely remove the toxic metals in the water, thus alternative purification methods using inexpensive materials were endeavor to improve the current treatment process. Wide ranges of low cost adsorbents were used to remove heavy metal in aqueous solution and wastewater. The low cost adsorbents were usually collected from agricultural waste, seafood waste, food waste, industrial by-product and soil. These adsorbents are readily available in a copious amount. Besides, the pretreatment are not complicated to be conducted on the raw products, which is economically sound for an alternative treatment. The previous studies have provided much evidence of low cost adsorbents' efficiency in removing metal ions from aqueous solution or wastewater. In this review, several low cost adsorbents in the recent literature have been studied. The maximum adsorption capacity, affecting factors such as pH, contact times, temperature, initial concentration and modified materials were revised and summarized in this review for further reference. Comparisons of the adsorbent between the modified and natural products were also demonstrated to provide a clear understanding on the kinetic uptake of the selected adsorbents. Some of the natural adsorbents appeared as good heavy metal removal, while some were not and require further modifications and improvements to enhance the adsorption capacity. SWOT analysis (strength, weakness, opportunities, threat) was also performed on the low cost adsorbents to identify the advantages of using low cost adsorbents and solve the weaknesses encountered by the utilization of low cost materials. This tool helps to determine the potential quality of low cost materials in the application for water and wastewater treatment.

Keywords Low cost · Adsorbents · Heavy metals · Adsorption capacity · SWOT analysis

1 Introduction

The demand on water usage is high as the domestic and industrial activities are increasing rapidly especially in the manufacturing sectors. However, on the other hand, the waste litters handling are not well managed, and at the same time the amount of waste being dumped are greatly increasing. This problem has affected the water quality causing the water quality to

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deteriorate (UNESCAP 2000). Discharge effluent from the industrial and agriculture activities are high in volume that such heavy metals, including organic compounds and some effluents were drained without proper treatment, which directly affected the water quality (UNESCAP 1999). Surface water and groundwater were also affected by the nonpoint sources pollutants contributed by agricultural activities, where the usage of chemical fertilizers leaches into groundwater and runoff. The high demand on the agricultural products led to the high utilization of fertilizer in order to yield a high number of productions (Dzikiewicz 2000). It has been found that exposure to staggering level of heavy metal such as arsenic to humans lead to arsenic poisoning (Reddy et al. 2013; Tandon et al. 2013; Vadahanambi et al. 2013). The arsenic pollution in the West Bengal districts of India, which caught the world concern, has affected so many people who are living within that area (Das et al. 1995). The inorganic arsenic in drinking water had caused the people who consumed the polluted drinking water to be suffered with health problem (Basu et al. 2001).

Many studies found that the natural surface water and groundwater are contaminated by heavy metals either due to anthropogenic sources or natural geological reason (Sarmani 1989; Karadede and Unlu 1999). Trace metals were found to be accumulated in the aquatic life and these protein sources were then entered the human's bodies through food cycle (Basu et al. 2001). The accumulation of Cu, Zn, Pb, Cd and Hg in the pink shrimp are said to be due to the release of industrial discharge into the river water according to Biney and Ameyibor (1992).

Many types of treatments have been introduced to remove the toxic metals in the water ranging from the expensive chemical treatment to the low cost biosorption. The chemical treatment for wastewater required mixture of chemical compound to reduce the metals pollutant, yet the removal could not achieve as expected in the guideline (Sanchez et al. 1999; Ismail et al. 2013). Some treatment methods developed such as coagulation, flocculation, reverse osmosis, membrane separation, ion-exchange, solar photo degradation and ozonation were conducted either to remove or degrade the pollutants in wastewater to harmless form (Pal et al. 2012; Lim and Aris 2013; Prieto-Rodríguez et al. 2013). The hazardous state of the toxic metals is changed into a stable and safe state so that the threat to human and the environment is reduced.

Conventional methods for treating wastewater were long conducted to provide a better water quality and to treat it before being released into the water bodies. Among the conventional water treatment conducted were chemical precipitation, coagulation, flocculation, ion exchange, membrane filtration and activated carbon (Bailey et al. 1999; Sud et al. 2008; Demirbas 2008; Chiban et al. 2012; Dave et al. 2012). However, the conventional methods were rather expensive, as higher cost was needed during the operation. Moreover, the conventional methods tend to have residuals and incomplete removal of the pollutants posing another problem (Chiban et al. 2012). Recently, scientists have been studying on using inexpensive adsorbents to remove heavy metals in the water. The low cost adsorbents used to practice adsorption activities were usually waste products from another production like agriculture, industrial and food production, which can be obtained abundantly.

The common removal or stabilizing agents used to stabilize heavy metals based on the recent studies are agricultural waste, oyster shells, crab shells, kaolinite, portland cement, fly ash, mussel shells, lime and Fe nanoparticles (Lee et al. 1997; Yoon et al. 2003; Vijayaraghavan et al. 2004; Moon et al. 2008; Tang et al. 2011; Seco-Reigosa et al. 2012). The main component in some natural products is calcium carbonate ($CaCO_3$), which can be used to immobilize the hazardous components of the heavy metals (Bothe and Brown 1999; Ismail et al. 2013). The agricultural waste products have the ability to remove the metals in aqueous solutions effectively (Ferro-Garcia et al. 1988; Demirbas et al. 2002; Wu et al. 2013). Among the agricultural wastes, which were popularly used to remove heavy metals were rice husk, palm fruit shell, fruit seed, nut shells, and fruit peels (Tan et al. 1993; Saifuddin and Kumaran 2005; Johan et al. 2011; Lim et al. 2012; Olu-owolabi et al. 2012; Omri and Benzina 2013; Sugashini and Begum 2013). These adsorbents were called as low cost based on the economic value, abundant supply and can be easily collected from the industrial, agricultural and food-processing production (Bailey et al. 1999; Jain et al. 2013). The low cost adsorbents, which review in present paper, are shown in Fig. 1.

The purpose of this review is to provide potential low cost materials used in the recent studies and also to compare the efficiency of various low cost materials in removing the metals. This review can serve as

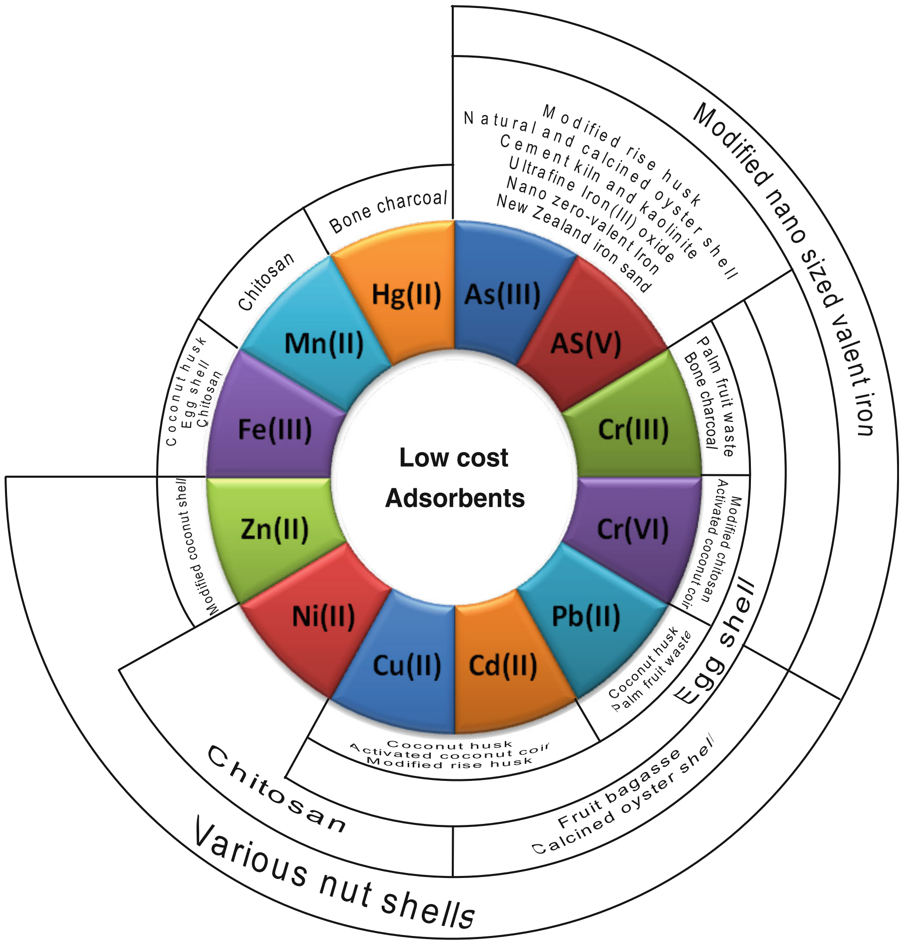

Fig. 1 Low-cost adsorbents reported in this review

examples for further studies on adsorption of heavy metals using low cost products. Besides, the reviewed low cost materials can include for the conventional wastewater treatments with a better economic aspect and environmental sound method. Integration of ideas and knowledge upon the current technology can provide a better wastewater treatment system and produce no secondary by-products of the treatment activities. In this review, SWOT analysis was applied on the low cost adsorbent used in literature. SWOT analysis of low cost adsorbent was applied to provide a perceived sight of an alternative water treatment method. Findings of SWOT for application of low cost adsorbents in removing heavy metals from water and wastewater also contribute a better understanding of new water management and help in sustainable water management.

2 Low cost adsorbents

2.1 Nano zero-valent iron particles and minerals

Recently, many studies have shown that arsenic can be stabilized using agents such as cement kiln dust (CKD), New Zealand iron-sand, magnetite, activated carbon, hematite, kaolinite, zeolite and natural products like oyster shells (Jing et al. 2003; Singh and Pan 2006; Moon et al. 2008; Yadanaparthi et al. 2009; Ok et al. 2010; Kim et al. 2011; Moon et al. 2011; Panthi and Wareham 2011).

pH plays an important role in affecting the adsorption rate, where the change of solution pH directly contribute to the available sites of the adsorbents. Panthi and Wareham (2011) revealed that at pH 7, New Zealand iron-sand has the maximum removal of

4 mg/L As(III). At pH 3, 63.0 % of As(III) was being adsorbed by the New Zealand iron-sand and at pH 11, the adsorption had reduces to 44 %. On the other hand, adsorption for As(V) showed a good match in acidic pH, approximately 97.6 % of As(V) was eliminated in the pH range of 3.0–6.0 (Panthi and Wareham 2011). The pH effects also occurred on the precipitation of arsenic on a calcium source (Bothe and Brown 1999). However, for the contact time between this adsorbents and arsenic, it did not reach 100 % removal. For the first 24 h, only 77.3 % of 4 mg/L As(III) has removed by this adsorbent, while for 4 mg/L of As(V) only 57.6 % has removed after the first 36 h (Panthi and Wareham 2011). Long contact times are required in order to achieve more metals removal.

According to Kim et al. (2011), a higher percentage of arsenic was immobilized by nano-sized magnetite coated with sodium dodecyl sulfate, follow by the magnetite, zero valent iron and the nano-sized zero valent iron coated by sodium dodecyl sulfate. The surface coated on the nano-sized particle has made the mobility of magnetite to be effective in the arsenic contaminated soil where the magnetic component of the magnetite has coated with negative charge of sodium dodecyl sulfate to prevent aggregation in the soil particles (Kim et al. 2011). Aredes et al. (2012) studied that magnetite, goethite and laterite could remove 100 % of As(V) (5 ppm) in water while hematite only adsorbed 20 % As(V) due to saturation, yet the hematite amount added to the arsenic solution was only half of the amount of magnetite, goethite and laterite. Aredes et al. (2012) also conducted a simple household treatment test on 20 ppm of As(V) solution, with just manual shaking of 100 ml As(V) and 5 g of laterite, the initial As(V) solution were reduced to 1 ppm in 10 min, proving that natural iron material is a great source for easy and fast water treatment application. The zero-valent iron loaded with a high amount of Fe with proportion of 95 % iron concretion, 2.5 % of carbon and 2.5 % of lime, and 90 % iron concretion, 5 % of carbon and 5 % of lime have higher removal of arsenic (500 ppm) which is about 99.95 and 99.94 % of removal as compared to the zero-valent iron loaded with lower Fe but substitute in other materials like carbon and lime with removal of 50.27 % only (Alshaebi et al. 2009). However, pH also constituted to the arsenic removal, low pH remove higher amount of arsenic than high pH especially when the pH value is more than 7 (Alshaebi et al. 2009). The stabilized state of iron nano particles also affects the reduction of the chromium in water and soil. Cr(VI) is effectively reduced to Cr(III) by using the zero valent iron particles stabilized with carboxymethyl cellulose, the harmful chromium form was changed to a less toxic form by this reducing agent with 90 % of 34 mg/L Cr(VI) being reduced in water, when the Fe dosage increased up to 0.12 g/L (Xu and Zhao 2007). This study showed that the stabilized iron nano particles had a better kinetic uptake for Cr(VI) as compared to Cao and Zhang (2006) which used non scale iron particles (Xu and Zhao 2007). However, the carboxymethyl cellulose used as stabilizer for iron nanoparticle is prone to decompose due to its organic structure, which can weaken the effectiveness of iron nanoparticle in stabilizing Cr(VI) (Xu and Zhao 2007). Magnetite is also one of the famous adsorbent for heavy metal uptake. The magnetite reduced from hematite by therma-mechanical reduction at 570 °C to powder mass of 25/1 for 15 h was observed to have a better removal of Cr(VI) at pH 2 than the magnetite reduced at 600 °C (Javadi et al. 2012). This is due to the small crystalline particles generated in the longer mechanical time. However, the removal percentages of Cr(VI) by nanocrystalline magnetite were not high at most only 40 % of removal for initial Cr(VI) of 20 mg/L at pH 2 (Javadi et al. 2012). The adsorbents dosage needs to be increased in order to observe a higher removal percentage for this study because the increased of dosage can provide more binding surfaces for the metal ion (Singha and Das 2011).

Ultrafine Fe_2O_3 (α-Fe_2O_3) nanoparticles which was proven by Tang et al. (2011) was able to remove As(III) and As(V) in a small amount. The As(III) with concentration of 0.115 mg/L was totally removed by 0.06 g/L Fe_2O_3 nanoparticles while for 0.095 mg/L of As(V) only 0.02 g/L of Fe_2O_3 nanoparticles (α-Fe_2O_3) was required for 100 % removal (Tang et al. 2011). This study has also demonstrated on the commercial Fe_2O_3 powder, and the results revealed that the Fe_2O_3 nanoparticles had two times higher removal percentage of As(III) and As(V) than the commercial Fe_2O_3 powder, which shows that the Fe_2O_3 nanoparticle has a great commercialize value (Tang et al. 2011). The α-Fe_2O_3 showed that modification can improve the adsorption capacity of adsorbent in removing the As(III) with the increased of surface areas for metal ions (Tang et al. 2011). Another study had enhanced the Fe nanoparticle by coating SiO_2 (Fe@SiO_2) on the surface, this Fe@SiO_2 successfully removed 100 % of 70 mg/L Cr(VI) in 180 min (Li et al. 2012a). The SiO_2 coated on the Fe nanoparticles restrained the jumble up of Fe particles and providing the Cr(VI) the ability to adsorb on the surface easily. The factors of preparing Fe@SiO_2 indeed are crucial as the speed injection of boron hydride and amount of tetraethylorthosilicate (TEOS) affect the Cr(VI) removal ability of Fe@SiO_2 (Li et al. 2012a; Ponder et al. 2000).

2.2 Calcium carbonate and seafood waste

Calcium carbonate and seafood waste were used to remove heavy metal due to the precipitation occur between the metal ions and carbonate from the adsorbents (Patterson et al. 1977; Sdiri and Higashi 2012). The cement kiln dust, which contains calcium carbonate, is one of the examples that were used in stabilizing harmful state of arsenic (Moon et al. 2008, Woolard et al. 1999). The cement kiln dust with mixture of kaolinite showed potential removal of As (III)and As(V) in the soil but the As(III) and As(V) in the mixture of cement kiln and montmorillonite are higher than the concentration in kaolinite after 1 and 7-day treatment (Moon et al. 2008). This showed that the clay type also played a role in the stabilizing the heavy metals as the kaolinite is a 1:1 type clay where the cation exchange is low and the arsenic anion can effectively removed from the soil. Positively charged kaolinites and other clay type are greatly suitable for anionic species of metals adsorption (Matusik and Bajda 2013). This clay type adsorbent can be used for removal of metals in aqueous solution. In Jiang et al. (2013), the 2:1 type clay used to adsorbed Cd(II) ions were also found to contain large amount of calcium and other negative charge minerals, which influenced the metal ions adsorption. The calcium content increased the pH, strongly decreased the mobility of metal ions such as Cd(II) ions in soil and water (Gerritse 1996).

Modifications on the natural clays were conducted in several studies to improve the metals adsorption efficiency. Djukic′ et al. (2013) discovered the removal efficiency of Serbian natural clay improved after undergo milling process. The Cd(II) and Ni(II) ions were reduced up to 87 % from the initial of 9.45 and 10.2 mg/L, respectively, due to the increased of surface area and the increased of exchangeable cation capacity (Djukic′ et al. 2013). Surface coating clay also is another interesting method to enhance the metal uptake ability. The clay coated with iron oxide showed a great removal of Pb(II) than the natural clay and natural sand (Eisazadeh et al. 2013). The pH of metal solution influences the removal efficiency of clay type. In Zhao et al. (2013), the Cr(VI) were highly removed in pH 2 because slight amount of natural components such as Al_2O_3 and Fe_2O_3 from the Akadama clay had leached out to bind with Cr(VI). This phenomenon was also observed by Helios-Rybicka and Wójcik (2012) where the iron oxide and manganese oxide were found to leach out from the kaolinite and immobilized the toxic metals. Combined of two types of adsorbents were done by El-Eswed et al. (2012) and Yousef et al. (2009), using kaolinite and zeolite tuff to remove multiple ions. The removal performance of kaolinite-zeolite tuff was successfully greater than the alone material (El-Eswed et al. 2012).

The uses of natural products from seafood are very popular in the recent researches as they can be easily obtained, cheap and the products can be recycled instead of being waste products. Many studies found that metal ions can be reduced by using natural oyster, crab and mollusk shells as its adsorbent (Lee et al. 1997; Rahman et al. 2008; Moon et al. 2011; Du et al. 2012; Ismail et al. 2013). The natural oyster shell could reduced 72 % of 4,779 mg/kg of arsenic in the mine tailing after 28 days of treatment with 30 % natural oyster shell weight to the sample; for the calcined oyster shell at 25 % weight, it effectively reduced 99 % of arsenic with the same contact period of 28 days (Moon et al. 2011; Seco-Reigosa et al. 2012). Best arsenic reduction results is shown by using the calcined oyster shell with the Portland cement treatment, which surpassed its natural product, and its combination with cement kiln dust achieved 95 % reduction of arsenic in 7 days period because of the pH and calcium contents in its combination with Portland cement (Moon et al. 2011). The natural products need to be enhanced or modified with other products in order to boost the reduction ability on the heavy metals, where the increase of available calcium on the adsorbents will improve the kinetic uptake of the heavy metals (Seco-Reigosa et al. 2012; Vieira et al. 2012). Besides, the calcined oyster shells and natural oyster shells also achieved a different heavy metals reduction (Liu et al. 2009; Ok et al. 2010). The natural oyster shells are consisting mainly $CaCO_3$, which is in the non-reactive form, while the calcined oyster shells changed the ordinary state of the natural oyster shell into CaO, which is the reactive form (Yoon et al. 2003; Ok et al. 2010). The surfaces of the modified shells were also improved with more porous and available site for adsorption of heavy metal ions (Liu et al. 2009). Study had shown that the calcined oyster shells had better stabilization of Cd and Pb in the soil than the unmodified oyster shells due to the quicklime, which changes the soil pH (Ok et al. 2010). This same adsorbent is applicable in the contaminated metal solution.

In Tudor et al. (2006), clam, oyster, and lobster shells were used to carry out metal adsorption on 10,000 mg/L of Pb(II) and Cd(II), respectively with comparison of adsorption capacity of using limestone and chitosan. The clam, oyster and lobster shells had the best uptake capacity for Pb with >99.9 % after 1, 5 and 326 h; while for Cd, only clam shell had the best removal capacity which is >99.9 % after 24 and 168 h. This is due to the larger adsorption surface of lobster and oyster shell (Tudor et al. 2006; Du et al. 2012). However, shells and chitosan appeared to be effective in removing Hg in the water due to the organic constituent on the lobster shells surface and the chitosan enable Hg to bind with the organic compound (Tudor et al. 2006).

2.3 Egg shells

In recent times, carbonated shells from food industries have been commonly used in many studies to perform metal ions adsorption due to its abundances after food processing (Ahmad et al. 2012; Wang et al. 2013). Eggshells are one of the examples of carbonate shells used for metal removal. The natural chicken eggshells were used to remove 96.43 % of 7 mg/L of Fe(III) in the aqueous solution with the optimum temperature of 20 °C and dosage of 2.5 g/L with control pH 6 (Yeddou and Bensmaili 2007).

pH has always been the control factors for adsorption of heavy metals, depending on the metal, certain metals prefer more neutral pH and higher pH for effective removal to occur because of the negatively charged surface which promote more adsorption to take place; while anionic metals prefer lower pH for better adsorption between adsorbents in aqueous solution (Kadimpati et al. 2012; Suteu et al. 2012). The calcined eggshells showed fast removal of 3 mg/L of Cd and Cr, respectively, as compared to the natural eggshell as the calcinations enhanced the adsorption of metal, but for Pb(II), natural eggshell was preferable in removing it from the wastewater due to the present of its soluble species (Park et al. 2007). The Cd and Cr were suitable in higher pH value uptake for these adsorbents than the Pb, which favored slightly lower pH (Park et al. 2007). Park et al. (2007) also conducted removal of Cd(II), Cr(VI) and Pb(II) in real wastewater by using the calcined eggshell which showed a great potential for application in the industrial wastewater since the metals could effectively reduce and the acidic wastewater has also shifted to neutral condition.

Efficiency of natural hen and duck eggshells were compared with the boiled hen and duck egg shells in removing lead (2.365 mg/L) in the battery wastewater by Arunlertaree et al. (2007). This study focused on the optimum pH, contact time and adsorbent dosages obtained from the experiment, showed the ability of egg shells in removing 100 % of lead in real wastewater (Arunlertaree et al. 2007). In fact, the natural duck shells had shown to have higher adsorption capacity, which is 1.643 mg/g, compared to the natural hen egg shells, which is 1.457 mg/g. This can be explained by the volume of pores on natural duck egg shells which were more and higher with level of protein fibers than natural hen egg shell (Arunlertaree et al. 2007). The protein fibers were not broken down in its natural state, making it held lead ions better than the boiled egg shells in acidic condition (Arunlertaree et al. 2007). Both Park et al. (2007) and Arunlertaree et al. (2007) proved that eggshells are very suitable for battery wastewater treatment with high removal efficiency; natural characteristics of eggshells can act as pH adjuster, which makes this adsorbent, has budgetary and environmental friendly values.

2.4 Chitosan

Chitosan is well known for having the ability to remove heavy metals in the water based on the previous study (Bailey et al. 1999; Saifuddin and Kumaran 2005; Pontoni and Fabbricino 2012). Cu(II) with initial concentration of 200 ppm was found to bind effectively with the chitosan with its small ionic diameter of 70 pm, which demonstrated 99.25 % of Cu(II) removal in aqueous solutions (Uzun and Güzel 2000). In contrast, Mn was found to be the least being removed among Fe, Ni and Cu due to its bigger ionic diameters and its stable complex which only form hydroxide and carbonate in alkaline condition (Uzun and Güzel 2000; Choi et al. 2007).

In the recent studies, chitosan is often combined and modified with other adsorbents to promote its effectiveness. Chitosan coated acid beads were able to reduced 65 % of 20 mg/L Cr(VI) at pH 1–92 % removal at pH 5 (Nomanbhay and Palanisamy 2005). Chromium and arsenic are pH dependent metals, the changes of the pH will directly affect the adsorption performance (Nomanbhay and Palanisamy 2005, Singha and Das, 2011). Adsorbent dosage also influenced the adsorption activities, 86 % of 20 mg/L of Cr(VI) was removed by 13.5 g/L of chitosan coated acid beads, 64 % removal of Cr(VI) with 18 g/L of chitosan coated beads, and 52 % Cr(VI) removal by 24 g/L of commercial activated carbon (Nomanbhay and Palanisamy 2005). The optimum agitation speed enables homogeneous surface binding to occur at the maximum condition (Nomanbhay and Palanisamy 2005; Bhuvaneshwari et al. 2012).

3 Agricultural waste

3.1 Coconut husk

Coconut husks are waste product of coconuts and it is usually available in abundance in the tropical countries. The high tannin in the coconut husk makes it a good metal adsorbent (Abdulrasaq and Basiru 2010). pH of the working solutions gave a significant influence in the adsorption process because 50 mg/L Fe(III), 50 mg/L Cu(II) and 10 mg/L Pb(II) were removed best in pH 5, where the removal capacities were almost 90 % and precipitation can be avoided. The adsorption of Fe(III) and Cu(II) using coconut husk also obeyed the Freundlich isotherm with R^2 value equal to 1, implicating the adsorption of Fe(III) and Cu(II) occurred due to the chemical bonding; while the Pb(II) showed better adsorption in Langmuir isotherm. The three metals also well described in the second-order kinetic which illustrated the occurrence of chemisorptions (Abdulrasaq and Basiru 2010). Modified coconut husk is proven to have a better adsorption of heavy metal. The coconut shell treated with acid and coated with chitosan has the best removal performance for 25 mg/L zinc with 93 % removal at pH 6, 30 g/L of adsorbent, 3 h contact time and solution temperature of 25 °C compared to the acid treated coconut shell and chitosan coated coconut because the coconut shell treated with acid and coated with chitosan has the most surface areas for Zn(II) to attach on (Amuda et al. 2007). The adsorption capacities of the modified coconut shells were also well represented by the Langmuir and Freundlich isotherm (Amuda et al. 2007).

Adsorption of heavy metals on the natural product has also successfully removed the heavy metals from contaminated water. Coconuts coir activated carbon was used in Chaudhuri and Azizan (2011) to remove the Cr(VI) in the aqueous solution. The contact time, adsorbent dosage and pH factors affected the adsorption of Cr(VI), Cu(II) and Cd(II) by the coconut coir activated carbon (Chaudhuri et al. 2010; Chaudhuri and Azizan 2011). The Cr(VI) with 20 mg/L of concentration was removed 100 % with the 8 g/L of coconut coir activated carbon and the optimum adsorption occurred between pH 1–2 as the H^+ in the solution alter the adsorbents' surface charges (Chaudhuri and Azizan 2011). pH always influence the adsorbent's metal uptake, as such Cr(VI) ions are preferable to be removed in acidic condition (Aliabadi et al. 2006). The coconut coir activated carbon have also shown a better adsorption capacity compared to the commercial activated carbon and its adsorption of Cu(II) and Cd(II) which fits well in Langmuir isotherm (Chaudhuri et al. 2010). This study showed that coconut coir activated carbon is another good alternative for removing heavy metals in water.

3.2 Rice husk

Rice is one of the main food sources in some countries, and it has been growing largely especially in South East Asia countries. The waste produced from the rice packaging has been chosen as one of the popular low cost adsorbent to remove heavy metals in water because of its highly available amount from the production. The utilization the by-product of rice such as rice husk will positively contribute to economic yield (Chakraborty et al. 2011).

In Rehman et al. (2011), the ordinary rice husk was modified with polyaniline for a better adsorption performance. From all the observed factors, pH, contact time, agitation speed, temperature, it was found that the polyaniline/rice husk showed the highest removal of Cd(II) which were 93.08 % (20 min), 97.5 % (30 °C),and 94.45 % (100 rpm) as compare to the ordinary polyaniline and polyaniline/saw dust which have lower removal percentage (Rehman et al. 2011). The polyaniline/rice husk and polyaniline/saw dust also fits better in Langmuir isotherm, while simple polyaniline is well represented by Freundlich. This showed that the adsorption occurred on the monolayer of the polyaniline/rice husk and polyaniline/saw dust while for the simple polyaniline, the adsorption occurred on the heterogeneous surface (Rehman et al. 2011). Besides, the modified rice husk used throughout this study can be reused after regeneration of the adsorbents by using nitric acid (Rehman et al. 2011).

Rice husk was also being modified in Johan et al. (2011) by incinerating the rice husk with a microwave incinerator at 500 and 800 °C to turn it into ashes form. The removal of Cu(II) was affected by the pH, the removal capacity increased when the pH increased accordingly. The rice husk ash produced from 800 °C has revealed a better adsorption of Cu(II) with 71.4 % of 10 mg/L Cu(II) removal at pH 5 while for the rice husk incinerated in 500 °C, its removal capacity was 66 % (Johan et al. 2011). This can be explained when the structure of rice husk incinerated in 800 °C has exposed with more pores as compared to rice husk incinerated in 500 °C. This has allowed the adsorption to occur on the surface of the ash. The Freundlich isotherm and pseudo kinetic second order also well represent these adsorbents as the adsorption happened in the multilayer of the ashes (Johan et al. 2011).

In order to have an effective adsorption, rice husk was also treated both chemically and physically to improve its uptake performance. The activated rice husk treated with KOH and K_2CO_3 had proven that the adsorption capacity was 441.52 mg/g, which is higher compared to the other absorbent such as bamboo, cotton stalk, pine wood powder, coffee ground, and durian peel (Foo and Hameed 2011). This is due to increased of BET surface area, external surface area and pore size, which influenced the adsorption process (Foo and Hameed 2011).

Rice husk MCM-41 loaded on with Fe also found to be a great adsorbent for 3 mg/L As(V) and 4.5 mg/L As(III). The Fe content prepared by Wet Impregnated Technique (WIT) showed lower adsorption value than the Fe content prepared by Direct Hydrothermal Technique (DHT) (Wantala et al. 2012). However, with the increased of Fe in DHT, the adsorption of As(V) was lower due to the insufficient surface area (Wantala et al. 2012). pH also influenced the adsorption of As(V) where As(V) was highly adsorbed in the acidic form (Wantala et al. 2012). Wantala et al. (2012) also suggested that rice husk MCM-41 loaded with iron to be used on the real contaminated groundwater and regenerate the used adsorbent, while for the exhausted adsorbent need to be placed in a well-aerated sand filter to prevent the toxic leachability.

The rice husk activated with higher temperature imposed a higher yield of heavy metals because the pores on the surface increased which improves adsorption efficiency (Yahaya et al. 2010; Tseng et al. 2006). The different activation times and temperatures gave a significant effect on the rice husk activation ash which would affect the removal amount of the heavy metals. Yahaya et al. (2010) studied that the most favorable of activation time and temperature for rice husk activation ash to remove 50 mg/L of Cu(II) were at 737 °C and 1.82 h with the removal of 11.7 % of Cu(II).

3.3 Palm fruit

Palm oil plantations are one of the most lucrative agricultural activities in Malaysia. Tones of palm oil and products from palm fruits were produced every year. This massive industry has also generated agricultural waste from the beneficial production (Saifuddin and Kumaran 2005). In Ideriah et al. (2012), the removal of Cr by palm fruit fiber biomass at pH between 4 and 10 was very low, while Pb was removed 100 % at pH 10 and Cu was removed 100 % at pH 10 and 12. However, the maximum adsorption of Pb by okra waste was optimized at pH 5 (Hashem 2007). This showed that Pb is pH dependent but at the same time depends on the adsorption materials, which are used to remove heavy metals in aqueous solution (Hashem 2007; Ideriah et al. 2012). Issabayeva et al. (2010) has shown that Cu(II) is a pH-dependent because the Cu(II) at pH 3 was not effectively removed as compared to pH 5, Cu(II) was highly adsorbed by the palm shell activated carbon. The palm shell was observed to have better adsorption with single Cu(II) than with Cu(II) in complexing agents which were the malonic acid and the boric acid where the concentration of Cu(II) with the presence of these complexing agents was higher than the single Cu(II) in the aqueous solution (Issabayeva et al. 2010).

3.4 Nut shell

Recently, agriculture wastes have been very popular among the other low cost bioadsorbents since the cost is inexpensive and the amounts can be easily collected in a large volume. Modified cashew nuts showed a great potential in removing Cu(II), Cd(II), Zn(II) and Ni(II) with the maximum adsorption capacity of 406.6, 436.7, 455.7 and 456.3 mg/g based on Langmuir isotherm (Kumar et al. 2012). Several factors such as pH, temperature, initial concentration, and contact time affect the adsorption kinetics. The best pH value for maximum adsorption of modified cashew nuts were pH 5 with 100 % removal for 100 mg/L of Cu(II), as for the optimum temperature was 30 °C while for the initial concentration, the lower metal concentrations achieved the highest adsorption compare to the high initial concentration due to the saturation state of the absorbents (Kumar et al. 2012).

The castor seed hull showed to have almost five times adsorption capacity than the activated carbon in comparison with the same fix contact time (300 min) and initial concentrations (5, 10, 20, and 30 ppm) (Sen et al. 2010). The uptake of Cd(II) in the aqueous solution by the castor seed hull were influenced by the pH where the adsorption at acidic state was lower and as the pH increased, the adsorption capacity improved due to the negative-charged surface which attracted the Cd(II) (Sen et al. 2010). pH also influenced the uptake of Cr(VI) by using cornelian cherry, apricot stone and almond shells where the removal capacity for 105 mg/L of Cr(VI) was 99.99 % with the optimum pH of 1 (Demirbas et al. 2004).

Comparison between the effectiveness of peanut hull carbon (PHC) and granular activated carbon (GAC) in removing Cu(II) had been conducted by Periasamy and Namasivayam (1996), showed that PHC performed a better recovery of Cu(II) in aqueous solution and wastewater. This study revealed that the PHC was able to remove 95 % of 20 mg/L of Cu(II) with 0.9 g/L of carbon concentration while GAC needed 13 g/L of carbon concentration. PHC also used 2.5 times shorter period to remove Cu(II) (10, 15 and 20 mg/L) compared to GAC (Periasamy and Namasivayam 1996). This study had proven that agricultural waste is a time effective and economic-wise absorbent.

The chestnut shell pretreated with 4 % of NaOH were found to be more efficient in removing Cd(II), Cu(II), Pb(II) and Zn(II) as compared to chestnut pretreated with acid formaldehyde, which causes the swelling of the adsorbent surface (Vázquez et al. 2009; 2012). 100 mg/L of Cd(II) was removed faster by using the chestnut shell pretreated with NaOH in comparison with Zn(II), Cu(II) and Pb(II) (Vázquez et al. 2012). This is due to the negatively charged surface of the pretreated chestnut shell, which attracts adsorption to occur. Pb(II) and Cd(II) were pH dependent metals, as the pH changes from 2 to 4, the higher amount of 20 mg/L of Pb(II) and 20 mg/L of Cd(II) were being adsorbed by walnut shells with the removal of >90 % for Pb(II) and almost 90 % removal for Cd(II) (Almasi et al. 2012).

Modified base-washed peanuts shells with citric acid demonstrated a better removal ability of Cd(II), Cu(II), Pb(II), Ni(II) and Zn(II) than some of the commercial resins such as Duolite GT-73 and carboxymethyl cellulose (Chamarthy et al. 2001). While for mixed metal ion solution of Cd(II), Zn(II), and Cr(III) in Cimino et al. (2000), Cr(VI) is found to be removed most compare to the other two metals. This study contributed that agricultural waste can be an alternative to remove metals in a cheaper cost with relatively simple modifications.

3.5 Fruit bagasse

Fruit bagasse is the fibrous residue from the extraction of the fruit juice. It is normally abundant from the food industries after massive of food and beverage production or packaging (Chakraborty et al. 2012). Studies have found that fruit bagasse contained high amount of hydroxyl and phenolic functional groups, have the ability in reducing heavy metals in water (Villaescusa et al. 2004; Farinella et al. 2007; 2008; Chakraborty et al. 2012). Grape bagasse from the wine production showed potential in removing cadmium and lead with adsorption capacity of 0.479 and 0.204 mmol/g estimated from Langmuir isotherm (Farinella et al. 2007).

Sugar cane treated with sulphuric acid and sugar cane activated carbon were used to remove Cd(II) (Krishnan and Anirudhan 2003). Sugar cane treated with 8.9 % of sulphuric acid had maximum adsorption of 98.8 % of 50 mg/dm^3 Cd(II) while for sugar cane activated carbon, 56.8 % of 50 mg/dm^3 Cd(II) were removed at pH 6 with (Krishnan and Anirudhan 2003). The pH affected the surface of the bagasse adsorbents by changing it to have negative charges and caused the adsorption of Cd(II) to increase after treated with higher percentage of sulphuric acid (Krishnan and Anirudhan 2003).

4 Bone charcoal

Bone charcoals were usually retrieved from the animal bone, and calcinations are conducted to change the original morphology of the bone for further utilization. The bone charcoal, which has the properties of carbon and calcium carbonate ($CaCO_3$), is capable to remove heavy metals like Cu(II) and Zn(II) and also some organic compounds (Wilson et al. 2003). Dahbi et al. (2002) has reported using bone charcoal from bovine to remove Cr(III) in wastewater where the removal of Cr(III) involved the reaction between the calcium in the bone charcoal. The study revealed that the use of camel bone charcoal in removing Hg(II) is due to the ion-exchange of mineral component on the bone charcoal, such as calcium and phosphate components (Hassan et al. 2008). pH also influenced the metal removal activities where the pH range affects the adsorption characteristics of the activated carbon. The Cr(VI) amount adsorbed by the activated carbon apricot stone decreased when the pH increased, Cr(VI) is preferably to be adsorbed in an acidic condition (pH 1) with the removal capacity of 99.99 %; while for cadmium, cobalt, Cr(III), nikel and lead, the amount adsorbed by activated apricot stone increased with higher pH (Demirbas et al. 2004; Kobya et al. 2005). The pH of the aqueous solution is directly affecting the metals adsorption activities for the pH dependent metals.

5 Gap and future perspectives on low cost adsorbents

The usage of low cost adsorbents has introduced a new alternative for water treatment system, and it also brings more opportunities in producing a better way of management. The future perspectives of low cost adsorbent can be determined by SWOT analysis where the strength, weakness, opportunities and threats of low cost adsorbents can be identified. The weakness and threats should be identified and changed to strength and opportunities to provide sustainable products in removing heavy metals in water (Praveena and Aris 2009).

5.1 Strength of low cost adsorbents

"Low cost" by the words itself has explained the financial meaning of inexpensive and affordable. The strengths of low cost adsorbents based on SWOT analysis are cheap in term of cost, abundant amount in the environment and easily obtain, as most of the adsorbents are waste products. The low cost materials used to perform heavy metals adsorption in this paper are basically the waste or by-products of manufacturing and food industries. One of the examples is mollusk shell, which can be easily obtained from either the food industries or collect from the coastal area (Moon et al. 2011; Seco-Reigosa et al. 2012; Suteu et al. 2012). These products will become waste after either being consumed by humans and cause odor and aesthetic problem if these wastes are not well-managed (Ok et al. 2010; Seco-Reigosa et al. 2012; Suteu et al. 2012). By using these materials to conduct heavy metals removal, will help to solve the pollution problem, furthermore, less cost is needed to pre-treat the materials before the adsorption activities (Moon et al. 2011; Ok et al. 2010).

Besides, the by-product from agricultural production like rice husk, rice bran, rice straw, palm oil husk, nut shell, apricot stone, fruit baggasses, sugar cane baggases and coconut husk are also easily available as these are the wastes from the agricultural production (Chamarthy et al. 2001; Chang et al. 2012; Demirbas et al. 2004; Farinella et al. 2007; Ferro-Garcia et al. 1988; Hashem 2007; Ideriah et al. 2012; Johan et al. 2011; Kakalanga et al. 2012; Kobya et al. 2005; Krishnan and Anirudhan 2003; Rehman et al. 2011). These products are so abundance as they are generated from the daily food production. Moreover, these products have a great ability in removing heavy metals in water and no secondary product or sludge is generated. This ability has made these adsorbents bonus in water treatment system. Besides, some studies have also conduct regeneration of the used adsorbents which also make the low cost adsorbents can be reused again, making these materials a new alternative for water management (Rehman et al. 2011; Bhuvaneshwari et al. 2012). At the same time, regeneration of the used adsorbents promotes sustainability water treatment products.

5.2 Opportunities of low cost adsorbents

SWOT analysis also focuses on the opportunities of low cost adsorbents in water treatment system. The

Table 1 Summary of SWOT analysis for low cost adsorbents

Strengths	Weaknesses
Cheap and inexpensive materials (Bailey et al. 1999; Gupta and Suhas 2009; Kurniawan et al. 2006)	Usually the adsorbents are not in readily form
By-products from agricultural and industrial activities (Chamarthy et al. 2001; Chang et al. 2012; Demirbas et al. 2004; Farinella et al. 2007; Ferro-Garcia et al. 1988; Hashem 2007; Ideriah et al. 2012; Johan et al. 2011; Kakalanga et al. 2012; Kobya et al. 2005; Krishnan and Anirudhan 2003; Rehman et al. 2011)	Pre-treatment and pre-washing processes are needed (Tudor et al. 2006)
Abundant in the environment (Chang et al. 2012; Kurniawan et al. 2006)	Some materials do not appear to be good adsorbents in natural form (Moon et al. 2011)
High ability in removing heavy metal from water (Saravanane et al. 2001; Zahra 2012)	Some adsorbents are only suitable for certain metal ions (Ahmad et al. 2012)
Opportunities	**Threats**
Costs for water treatment system can be reduced (Patil 2012)	May cause insufficient of adsorbents due to the high demand in dosage
Long hour of contact period can be reduced (Periasamy and Namasivayam 1996)	Extra chemical costs may be needed for pH adjusting to provide suitable condition for optimum adsorption to occur
Modification and enhancement of low cost adsorbents can turn into marketable products	
Improvement and potential replacement for the conventional water management system (Gupta and Suhas 2009; Patil 2012)	

studies on low cost adsorbents have reported for cost and time-efficient, this can be used to replace the expensive and time-consuming treatment methods (Arunlertaree et al. 2007; Champagne and Li 2009). The low cost adsorbents can be pre-treated or modified to improve the current efficiency in order to produce a marketable product for the water management system and the manufacturing industries (Hsien and Liu 2012). Besides, the low cost adsorbents are environmentally friendly product, making it a better choice for water management (Kırbıyık et al. 2012). The operational cost can be reduced with the addition of adsorbents to the traditional water treatment system (Martinez-Juarez et al. 2012).

5.3 Weakness of low cost adsorbents

The weakness of low cost adsorbents has pointed out by SWOT analysis, as the low cost adsorbents are usually not readily available. The low cost adsorbents usually present as raw materials (Liu et al. 2009). Cleaning and pre-treatments are needed in order to eliminate the residue of contaminants on the surface of the low cost adsorbents (Hadi 2012; Liu et al. 2009). Some low cost materials do not appear to be good adsorbents in its natural form, modification and enhancements are needed to change the original state to reactive form for a better adsorption result (Liu et al. 2010b; Park et al. 2007; Quintela et al. 2012). Besides, some adsorbents in small amount or too much dosage cannot remove metal ions in water due to the insufficient binding surfaces and aggregation (Yeddou and Bensmaili 2007). Some adsorbents are particularly effective on selected metal ions while failed on the other metal ions (Ahmad et al. 2012). Enhancement and modification can be done in order to produce better adsorbents for metal ions uptake (Chamarthy et al. 2001; Liu et al. 2012).

5.4 Threat of low cost adsorbents

The over-explored of low cost adsorbents is the threat toward low cost adsorbents. When the low cost adsorbents are being widely used, the demand will be higher than the supply. This will also caused problem to the industries due to the insufficient supply. Besides, some metal ions are pH dependent and it will cause the treatment system to use more chemical to adjust the suitable pH for the adsorbents. Desorption of metal ions can be done in order to regenerate the adsorbents for the further usage (Bhuvaneshwari et al. 2012; Grover et al. 2012; Hsien and Liu 2012). By this

Table 2 Summary of adsorption capacity/removal efficiency at equilibrium for the adsorbents conducted in the previous studies

Types of materials	Adsorption capacity (mg/g)/removal efficiency (%)											References
	As(III)	As(V)	Cr(VI)	Cu(II)	Cd(II)	Zn(II)	Ni(II)	Pb(II)	Co(II)	Mn(II)	Fe(III)	
Coconut shell			20.0 mg/g									Alaerts et al. (1989)
Coconut shell			6.0 mg/g									Selomulya et al. (1999)
Dust coal			4.4 mg/g									
Hazelnut shell			177.0 mg/g									Cimino et al. (2000)
Resin-supported ferragel			28 mg/g, 90 %									Ponder et al. (2000)
Bituminuous coal			44.4 mg/g									Hamadi et al. (2001)
Sawdust			1.9 mg/g									
Petroleum pitch			23.7 mg/g									Park et al. (2003)
Bone charcoal				27 ± 1.3 mg/g		24.5 ± 3.0 mg/g						Wilson et al. (2003)
Almond shell			20.0 mg/g									Demirbas et al. (2004)
Apricot shell			21.0 mg/g									
Hazelnut shell			170.0 mg/g									Kobya (2004)
Hevea brasiliensis sawdust			44.1 mg/g									Karthikeyan et al. (2005)
Terminalia arjuna nuts			28.4 mg/g									Mohanty et al. (2005)
Palm shell charcoal coated with chitosan			154 mg/g									Saifuddin and Kumaran (2005)
Pine leaves (pH 2, 15 mg/g Cr(VI)			99 %									Aliabadi et al. (2006)
Nano scale zero valent iron			1.75 mg/g, 24–38 %									Cao and Zhang (2006)
Chitosan-coated acid treated coconut shell carbon						60.41 mg/g						Amuda et al. (2007)
Chitosan-coated coconut shell carbon						50.93 mg/g						
Acid treated coconut shell carbon						45.14 mg/g						
Natural hen egg shell								1.457 mg/g				Arunlertaree et al. (2007)
Boiled hen egg shell								1.092 mg/g				
Natural duck egg shell								1.643 mg/g				
Boiled duck egg shell								1.236 mg/g				
Fly ash(pH 8.5, 1.5 mg/L Mn)										92.20 %		Sharma et al. (2007)

Table 2 continued

Types of materials	Adsorption capacity (mg/g)/removal efficiency (%)											References
	As(III)	As(V)	Cr(VI)	Cu(II)	Cd(II)	Zn(II)	Ni(II)	Pb(II)	Co(II)	Mn(II)	Fe(III)	
Sodium carboxymethyl cellulosestabilized Fe nanoparticles			255 mg/g, 90 %									Xu and Zhao (2007)
Scrap iron			19 mg/g, 25 %									Gheju et al. (2008)
Zerovalent iron nanoparticles			180 mg/g, 98 %									Li et al. (2008)
Manganese-Coated Sand (pH 9)										90 %		Lee et al. (2009)
Manganese-Coated Sand (Birm commercial sand) (pH9)										100 %		
Coconut husk				92 % ± 2.8				94 % ± 3.2			94 % ± 1.4	Abdulrasaq and Basiru (2010)
Nano-scale Fe^0 particles supported on a PAA/PVDF membrane			181 mg/g									Li et al. (2010a), (b)
Entrapment of nanoscale zero-valent iron in chitosan beads			98.40 %									Liu et al. (2010a)
Palm shell			12.6 mg/g									Owlad et al. (2010)
Granular activated carbon					1.385 mg/g							Sen et al. (2010)
Castor seed hull					6.983 mg/g							
Bituminous coal			27.8 mg/g									Chaudhuri and Azizan (2011)
Coconut coir			38.5 mg/g									
MicrowaveIncinerated Rice Husk Ash (500 °C)				3.279 mg/g								Johan et al. (2011)
MicrowaveIncinerated Rice Husk Ash (800 °C)				3.479 mg/g								
Polyaniline					0.165 mg/g							Rehman et al. (2011)
Polyaniline/saw dust					4.739 mg/g							
Polyaniline/rice husk					5.128 mg/g							
Fly ash-based geopolymer (35 °C, pH 4)								183.605 mg/g				Al-Zboon et al. (2011)
Silica fume supported-Fe^0 nanoparticles			88 %									Li et al. (2011)

Table 2 continued

Types of materials	Adsorption capacity (mg/g)/removal efficiency (%)											References
	As(III)	As(V)	Cr(VI)	Cu(II)	Cd(II)	Zn(II)	Ni(II)	Pb(II)	Co(II)	Mn(II)	Fe(III)	
New Zealand Iron sand		97.60 %										Panthi and Wareham (2011)
Ultrafine Fe_2O_3 nanoparticles		100 %										Tang et al. (2011)
Nano hydroxyapatite (n-HAp) (pH7)			98 %									Asgari et al. (2012)
Phanerochaete chrysosporium loaded with nitrogen-doped TiO2 nanoparticles					84.20 %							Chen et al. (2012)
Jojoba oil (20 mg/L Cu, Pb)				95 %				98 %				El Kinawy et al. (2012)
Banana peel (5 mg/L Cu)				85 %								Hossain et al. (2012)
Banana peel (10 mg/L Cu)				88 %								
Modified cashew nut shell				406.6 mg/g	436.7 mg/g	455.7 mg/g	456.3 mg/g					Kumar et al. (2012)
Fe@SiO_2			467 mg/g, 64.18 %									Li et al. (2012a)
Spent grain (pH9)					92.63 %							Li et al. (2012b)
Activated carbon orange peel					28.67 mg/g				45.44 mg/g			Moreno-Pirajan and Giraldo (2012)
Duckweed (pH6, 20 mg/L Pb, 10 days)								89 %				Singh et al. (2012)
Fe loaded on rice husk RH-MCM-41	156 mg/g	1,111 mg/g										Wantala et al. (2012)

way, the low cost adsorbents, which obtained from the environment, can be conserved. Table 1 summarizes the SWOT analysis for the low cost adsorbents reported in this paper.

6 Conclusion

Wide ranges of waste products have reusable value instead of being disposed. From this review, many low cost materials have proven to have the ability in removing heavy metal. In addition, some of the wastes were enhanced and modified in order to improve the adsorption capacity. Table 2 shows various adsorbents used in the recent studies, the adsorption capacity and removal efficiency of the selected adsorbents. Comparison can be seen from Table 2 as different low cost adsorbents were used to remove the same heavy metals had dissimilar removal capacity. The low cost adsorbents have shown a great potential in water treatment application as the adsorbents can be alternatives choices to replace the current expensive chemical cost and conventional operations, which need more contact time into remove the heavy metals. Based on the SWOT analysis output, the low cost adsorbents have promising value for water management and economy. The low cost adsorbents can replace the high cost treatment system and at the same time present better results as compare with the conventional methods. Opportunities on the utilization of low cost adsorbents are greatly available with the further modification to change the materials into valuable and marketable products. The weakness and threats of the low cost adsorbents analyzed by SWOT can be encountered by further studies on modification, enhancement and regeneration of the materials. Therefore, low cost adsorbents are highly recommended to be selected for the sustainable water management.

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