

Manganese in the source of groundwater in Malaysia and the method for the removal process: A review on the adsorption and membrane separation processes

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Abstract

In Malaysia, the quality of groundwater as one of the main sources drinking water is deteriorated due to the presence of a high level of manganese, which exceeds the allowable values for drinking water consumption. Manganese at concentration higher than 0.1 mg/L causes staining, high turbidity and bad taste problem in drinking water, and eventually can cause a depletion of brain dopamine and a syndrome of motor dysfunction and memory loss resembling Parkinson disease. Several methods have been used to eliminate manganese from the groundwater, which include precipitation, coagulation, ion exchange, oxidation and filtration, aeration, activated carbon adsorption, ionic liquid extraction and biosorption. Among those methods, adsorption is the most efficient and cheaper method to remove heavy metal as the operation is easily be controlled and the reversible adsorbents can be regenerated through a suitable process. Membrane filtration on the other hand particularly reverse osmosis and nanofiltration have been found to be a very effective and economical way to isolate components that are suspended or dissolved in a liquid. In addition to that, the combination of adsorption and membrane filtration process such as polymer enhanced ultrafiltration and adsorptive membrane respectively are currently attracted attentions. This paper provides a review on the adsorption process and membrane filtration process for manganese removal, with subsequently outlining the potential adsorbents to be incorporated in the fabrication of adsorptive membrane.

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1.0 Sources of drinking water in Malaysia

In Malaysia, 99% of its surface water supply and 1% of its groundwater supply are used for domestic (Azrina, 2011). The total Malaysian water resources are to be approximately 580 km³ per year and 30% for urban use (Azrina, 2011). Water supply primarily from groundwater and surface water was treated and distributed to customers as filtered drinking water, distilled mineral water for tap water, and drinking water. Bottled mineral water, bottled drinking water, and tap water are the main sources of drinking water in Malaysia (Chiba et al., 2006). Surface water supplies from Sungai Kinta, Sungai Langat, West Coast Peninsular Malaysia, and Sungai Selangor are extensively used for drinking water.

Based on study by Jamaluddin et al. (2013), in Malaysia, the population in Sabah and Kelantan have limited surface water supplies. As a result, groundwater is becoming the main supply of water for

domestic use. Air Kelantan Sdn. Bhd. (AKSB) has reported that groundwater as the source used for a total of six water treatment plants. Based on several reports, heavy metal concentration in the groundwater was comply to the Malaysia Drinking Water Quality Standard except for groundwater from Ampar Tenang, Rosob Village, and Machang (Rahim, 2010). The well water samples in Rosob Village, which is used for drinking water has a high concentration of manganese ions up to 409.5 µg/L as compared to 100 µg/L as outlined by the Malaysian Ministry of Health (Rahim, 2010).

2.0 Manganese

Manganese ion is an essential element for human body and enzyme activation (Wang et al., 2014). However, high level of manganese ion in brain causes a progressive, irreversible neurological disorder known as manganism. It can also cause depletion of brain

dopamine and a syndrome of motor dysfunction and memory loss resembling Parkinson disease (Mc Millan et al., 1999). As for the infants, high level of manganese ion in drinking water can retard their normal growth and intellectual development (Hong et al., 2014). Apart from its effect on human health, manganese ion at concentration higher than 0.1 mg/L causes staining, high turbidity and bad taste in drinking water. Therefore, the removal of manganese ion is vital in Malaysia, particularly where the source of drinking water is from the groundwater.

3.0 Methods of manganese removal

Several methods have been used to eliminate manganese ions from the groundwater, which include precipitation, coagulation, ion exchange, oxidation and filtration, aeration, activated carbon adsorption, ionic liquid extraction and biosorption (Mariana et al., 2015). However, there are drawbacks on the application of those methods such as high operational cost, generation of toxic material and generation of large amount of sludge. Membrane filtration either in combination with polymerization process, in situ adsorption and filtration or as a single unit operation, has become a promising alternative with additional advantage of environmentally friendly process due to no solvent is being used, no generation of sludge and mild process parameters.

Therefore, the objective of this review is to discuss the previously reported research on the application of adsorption and membrane separation, respectively on the removal of manganese ions, with the aim to propose a new process or material for manganese ion removal.

3.1 Adsorption process

Despite the generation of large amount of sludge from this process, adsorption has been widely used to treat water containing manganese ion. Adsorption is a process involving the deposition of one substance on the surface of another material. The mechanism of adsorption can be one or a combination of several phenomena, which include formation of chemical complex at the surface of the adsorbent, electrical attraction (a phenomenon involved in almost all chemical mechanisms, including complex formation), and the rejection of a solute from the feed solution. The adsorption was attributed to a strong intermolecular bonding between molecules of the solvent and the solute. This mechanism could be explained by the

second law of thermodynamics, where the total energy of all bonds increases after the adsorption process has taken place (Woodard, 2001).

3.1.1 Polymer as adsorbent

There are few types of polymer that have been reported being used as adsorbents for manganese ion removal that include polyvinyl alcohol/chitosan dry blend hydrogel, polyhydroxyl-polyurethane foam, and polysodium acrylate-graphene oxide (Abdeen et al., 2015).

Polyvinyl alcohol/Chitosan hydrogel

It was reported that the blend hydrogel from polyvinyl alcohol/chitosan was able to remove 80% of manganese ion over 90 minutes of contact time (Abdeen et al., 2015). The polymer blending between polyvinyl alcohol/chitosan was prepared by ball milling method with the aim to produce a nanosized particle without using a cross linker. Poly(vinyl alcohol) (PVA) is widely used in polymer composite manufacturing due to it has high solubility in water, non-toxic behaviour, biocompatible, low cost, and high chemical stability (Koo et al., 2001 & Kao et al., 2009). Whereas, chitosan, which is extracted from crustacean shells, is a biopolymer that has high adsorption capacity for a wide range of heavy metals due to its relatively high proportion of active nitrogen sites (Muzzarelli, 1977).

Polyhydroxyl Polyurethane Foam (PPF)

In a study by Moawed et al. (2012), PPF at various loadings had been used to remove manganese ion. The amount used was in the range of 0.1 to 0.5 g where it was equilibrated with feedwater adjusted at the optimum pH. It was found that the maximum removal of manganese ions was achieved around 88–91% at pH 6–8 of the feed solution. It was found that electrostatic interaction to form hydrogen bonding is the main mechanism in the removal process of metal ions, where at pH 6–8, the adsorbent's surface was negatively charged that cause the adsorption of manganese ion. At pH lower than 5, the surface of PPF was positively charged.

Polyhydroxyl-polyurethane foam demonstrated a range of positive parameters, such as the low price for manganese ion removal and quick extraction kinetics. This integrated foam is theoretically can be used to extract the solid material for isolation

preconcentration, and to detect the trace manganese ion in environmental samples when combined with a spectrophotometer. This new adsorbent has a strong physical and chemical stability function, can be repeatedly used due to its high adsorption capacity, and a rapid balance in the recovery of the metal ions tested. Therefore, PFF was found as the suitable material for the pre-concentration and removal of manganese ion (Moawed et al., 2012).

Polysodium Acrylate-Graphene Oxide (PSA-GO)

Graphene as one type of carbon material, is widely applied as nano-sorbent for heavy metal (Khulbe et al., 2018) removal. It has been reported by Deepti et al. (2016) that the adsorption of manganese ions to PSA-GO gel was due to electrostatic interaction, where the adsorption process may involve physical and chemical interactions. The process of adsorption is often influenced by the organic matter present in the feed solution, ionic strength of the solution, and initial pH of the solution. The initial pH values of the solution affect the condition of the functional groups on the adsorbent and the metal ions in the existing form (Huang, 2012).

It was observed that 84% manganese ion adsorption occurred at the initial pH above 3 (Deepti et al., 2006). The positively charged metal ions will be difficult to be absorbed on the PSA-GO gel surface which is also positively charged at a low pH range of 2 to 3 due to the repulsion of electrostatic (Alslaibi et al., 2015). Thus, the mixture of amine functional groups on the PSA-GO gel and metal ions will be prominent, resulting in the increase of metal ions adsorption.

3.1.2 Inorganic polymer and natural adsorbent

Besides using the above polymers, inorganic adsorbents such as zeolite, activated carbon, clays, agricultural residue, bio adsorbent such as non-living biomass, algal biomass, and microbial biomass (Fu et al., 2011; Samiey et al., 2014) have been used in the manganese ion removal process. Among them, adsorption by using activated carbon is the most efficient method where 99.42% removal was achieved by using granular activated carbon (Deepti et al., 2016). However, as the activated carbon is expensive, therefore the use of lower cost adsorbent is highly preferred. These adsorbents, which are generated from natural renewable resources such as agricultural residues are becoming attracted in the treatment of the

heavy metal-polluted water. These residues include rice husk ash, peanut husk, banana peels, potato peel, sugarcane bagasse, yam peel, orange peel, sunflower seed shell and tamarind fruit shell (Rudi et al., 2020).

On the use of inorganic polymer, there was a study conducted on the removal of manganese ions from aqueous solution by adsorption on the surfactant-modified alumina (SMA). The anionic surfactant used was sodium dodecyl sulphate (SDS). It was found that the increase in the pH of manganese ion solution from 4.04 to 8.05 along with the increase in the agitation speed from 110 to 150 rpm resulted to the enhancement in the adsorption capacity from 43.48 to 62.02%. Furthermore, the increase in process temperature and adsorbent dosage also enhancing the rejection of manganese ion. These situations were due to the increased availability of adsorption sites and endothermic process, respectively where the adsorption process is fast and follows Freundlich isotherm model.

Belviso et al. (2015) also explored on the potential of fly ash and zeolite, respectively as high-efficiency adsorbents to remove manganese ions. With the adjustment of high pH for the solid/liquid mixtures, both materials were found to efficiently remove manganese ions but fly ash is superior as compared to zeolite that might be due to metal sequestration by zeolite.

Vistuba et al. (2013) investigated on coal as adsorbent for simultaneous removal of iron and manganese from aqueous solutions. It was found that the removal of ferum ions was higher due to its smaller size as compared to manganese ion, which made it adsorbed more easily into the small pores of the adsorbents. The kinetic data and selectivity of ferum ions followed pseudo-second order model. The results showed that coal could be efficiently used for the removal of iron and manganese from aqueous solutions.

Adekola et al. (2016) also reported on the attempt to remove ferum ion and manganese ion simultaneously from a pure solution and actual raw water respectively through adsorption process by using rice husk ash. Result showed that the removal of manganese ions achieved 100% as compared to 70% for ferum ion. This result shows the effectiveness of using rice husk ash for manganese ions removal.

Based on the above findings, adsorption has been evidenced as cheaper and efficient method to remove manganese ions from a wastewater due to its flexible

operation and the reversible adsorbents can be regenerated (Fu & Wang, 2011). The right choice of adsorbents is important to increase the process efficiency. However, this process may lead toward producing large amount of sludge due to difficulties in recycling and recovering some of the adsorbents from water (Fu & Wang, 2011; Samiey et al., 2014).

3.2 Membrane filtration

Membrane filtration, as an alternative to traditional water treatment, becoming increasingly desirable. The use of membrane for the removal process has become a promising alternative with the additional advantage of environmentally friendly process. Loose membranes, such as ultrafiltration (UF) and microfiltration (MF) membranes, are superior to traditional particulate and colloid removal processes that are the main cause of turbidity and are essential carriers of pathogenic microorganisms and metals (Thanuttamavong et al., 2002). However, these membranes are not effective in the removal of solutes such as iron and manganese ions. The effective removal could be achieved through the use of reverse osmosis (RO) and nanofiltration (NF) but at the expense of substantially higher energy consumption due to the use of high pressure.

Membrane systems in the treatment of drinking water have been used for the removal of dissolved organic and colour, softening, removal of particles and microbes, and desalination (Vandecasteele, 2003). Nanofiltration membranes are used to eliminate hardness, turbidity, dissolved salts, and microorganisms (Hilal et al., 2004). Ultrafiltration membranes could remove macromolecules and microparticles that typically contain organic colloids and inorganic particles.

Numerous studies have been conducted to explore membrane filtration capability in water treatment. However only limited number of studies focusing on the removal of manganese ion that contained in groundwater by using nanofiltration membranes. Previous studies have shown that high concentrations of manganese ions can be removed by a combination process of microfiltration and oxidation (Ellis et al., 2000). The efficiencies of removal have been increased by pre-chlorination with enhanced oxidation mixing, which has influenced microfiltration efficiency.

Large dose pre chlorination was needed to remove manganese ion by oxidation and ultrafiltration, while significant removal of ferum ion was possible even

without the addition of chlorine (Han et al., 2005). Nanofiltration process on the other hand, has become good candidate for recovery process of heavy metal ions due to its pore size, and distinctive surface electric charges that contributes to the ability to allow passage of monovalent ions but retain the divalent/multivalent ions (Li et al., 2019). As compared to reverse osmosis, nanofiltration uses lower operating pressure and able to remove the hardness elements such as magnesium and calcium, bacteria, virus, and colour (Deepti et al., 2016). There are only few studies have been reported on the use of nanofiltration for manganese ion removal. Al-Rashdi et al. (2011) reported the use of commercial nanofiltration (NF270) in the removal of manganese, in which the membrane was able to reject 89% of the ions. In an earlier study by Shaari et al., 2019, a novel thin film composite (TFC) membrane was fabricated from a polysulfone as the support layer with a thin layer made from a blend of polyvinyl alcohol (PVA)/chitosan and cross linked with tetraethyl orthosilicate (TEOS) to remove ferum ions. Result showed that more than 90% ferum ions was rejected by the TFC. The high rejection of ferum is attributed to the presence of chitosan as the adsorbent. This type of membrane separation reflects the integrated complexation process, where the adsorption and separation occur simultaneously on the membrane's surface.

3.3 Polymer enhanced ultrafiltration

Polymer enhanced ultrafiltration (PEUF), which involved a hybrid process of complexation followed by ultrafiltration process has been widely used and proven to effectively separate heavy metals from wastewater (Barakat et al., 2010). There are few types of water-soluble polymeric ligands that have been found as effective material to form complexes with heavy metal ions. These polymers are chitosan and chitin as biopolymer and synthetic macro ligands such as carboxymethyl cellulose, poly(acrylic acid), polyvinyl alcohol and polyvinyl ethylenimine (Barakat et al., 2010). In the complexation process, the heavy metal ion forms a high molecular weight compound with the polymeric ligand in the feed water. In the subsequent membrane filtration process, the compound was prevented to permeate through the ultrafiltration membranes due to its large molecule size.

The removal of manganese ions using PEUF was reported by Qiu et al. (2014) where co-polymer of maleic acid and acrylic acid were used as polymers

with polyvinyl butyral hollow fibre as the ultrafiltration membrane. The process successfully removed 99.6% of manganese ions where the complexation process was very rapid and completed in 5 min with pH 6 as optimum condition and the mass ratio of polymer to manganese ion solution was 6.

In a study to eliminate manganese ions by using an ultrafiltration membrane with different pH values, Choo et al. (2007) found that the percentage of manganese removal increased by adding poly(acrylic acid) (PAA) as an adsorbent prior to the filtration process. The efficiency of manganese ions rejection was dependent on the PAA concentration, but not on PAA molecular weight, which suggest that the interactions between carboxyl groups in PAA and manganese were not influenced by the length of the polymer chain. It was also found that the efficiency of manganese ions removal increases at higher pH of the feed solution (pH >7) along with 80% PAA concentration because of the increase formation of PAA-Mn complexes at high COOH:Mn ratios. Besides that, the manganese ions could form precipitate at pH >7. The effect of calcium ion as water hardness on manganese removal performance was also evaluated. When the calcium ion was present in the feed solution, it interferes the interactions between PAA and manganese ions that cause reduction in manganese ion removal. The efficiency of manganese ion removal was 60% at pH 7 with a molar ratio COOH:Mn of 27 without the presence of calcium ion, and it decreases to 25% with the presence of 0.5 mM calcium ion.

Although the polymer enhanced ultrafiltration process is a promising process for manganese removal, the drawback of the process lies with the use of large amount of polymeric ligand (Bessbouse et al., 2008). Besides that, severe fouling on a membrane's surface was anticipated after the deposition of the large amount of the complexed species, which also cause a decrease in the hydrophilicity of membrane's surface as a result from charge coupling in the complexes (Bessbouse et al., 2008).

3.4 Integrated complexation process

The simplification of the above method can be done by using an integrated complexation process, where the complexation and filtration steps are performed in a single unit operation (Bessbouse et al., 2008). The advantages of this process lie with a less amount of polymer used as compared to polymer enhanced ultrafiltration process and the polymer for the

membrane can be tailored-made according to targeted metal ion to be removed (Bingjie et al., 2013). The produced membrane is also known as adsorptive membrane. Chitosan is widely used for the removal of heavy metal ions due to the presence of amine and hydroxyl functionalities, which lead towards high binding capacity. As a natural amino polysaccharide, this polymer is becoming attracted due to its wide availability, and has unique functionalities (Salehi et al., 2016). Reiad et al. (2013) had conducted a study on the simultaneous removal of ferum ion and manganese ion from feed solution, where it was found that the affinity of a polymer blend chitosan (CS)/polyethylene glycol (PEG) membrane is higher towards ferum ion as compared to manganese ion. They also found that when the ratio of CS:PEG was increased, the potential to adsorb ferum and manganese ions also increase. This is due to the presence of high hydroxyl group in the polymer matrix of the membrane that increase the electrostatic interaction between the counter-metal ions in the solution and the negatively charged hydroxyl groups on the membrane's surface. When the initial metal ion concentration was increased, the diminishing pattern in metal adsorption was observed, where it is likely attributed to a decline in the affinity of hydroxyl functional groups with a growing degree of site occupation, accompanied by early and simple access to low metal-binding sites (Reiad et al., 2013).

4.0 Outlook for future research

Based on the above review, integrated complexation process was found as preferable process for the removal of manganese ions due to its simple and efficient process. In term of type of polymer, chitosan has been found to efficiently remove heavy metal ions. However, for efficiently remove the manganese ions, another type of adsorbent must be blended with chitosan to form the mixed matrix membrane. As reported by Adekola et al. (2016), rice husk ash was able to remove 100% of manganese ion from raw water, therefore the mixed matrix membrane formulated from a blend of chitosan and rice husk ash could become a preferable alternative for manganese ion removal. In Peninsular Malaysia, large amounts of biomass that is coming from agricultural residues are highly produced. From the major agricultural crops grown in Malaysia, rice accounts for 12.68% from the total percentage. There are not much uses of the agricultural residues, where mostly are disposed of by burning.

To protect the environment and saving of energy, the domestic waste, biodegradable waste, and natural fibres can be utilized to produce green composites by incorporating them into the polymer matrix (Sain et al., 2000). Rice husk is obtained from the rice which cover the outer layer of the rice (Arjmandi et al., 2015). The incineration process turns the rice husk to ash. The utilization of rice husk ash in the membrane formulation is an effort towards producing a value-added product from rice husk along with minimizing the environmental impact due to large disposal of rice husk. Although the usage of rice husk ash varies with different sieve sizes of rice husks, the main component of the rice husks such as silica has been proven to

improve the mechanical properties of polymeric materials (Arjmandi et al., 2015). During the treatment process, the pH of the feed solution containing manganese ion should be adjusted to pH 6-8 as the enhancement of adsorption process occurs within this range of pH.

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