Coagulant Property of *Moringa oleifera* in the Treatment of Wastewater – A Review

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Abstract

Moringa oleifera is an average size tree with approximate height between 10 to 15 m. It is cultivated in many regions of the world and has been reported to possess multiple utilities ranging from medicinal uses to its natural coagulating property in the treatment of water and wastewater. The seed powder of *Moringa oleifera* had also been used in the removal of dyes, organic pollutants, drugs, heavy metals, fluoride ion, etc. A review of the coagulant property of *Moringa oleifera* in the removal of contaminants is presented with a focus on recent researches and suggested future research works using this natural coagulant.

Keywords: *Moringa oleifera*, coagulation, heavy metals, dyes, drugs, organic pollutants, disinfection.

1. Introduction

Wastewater can simply be regarded as any water which has been adversely affected by human day to day activities. There are number of routes through which waste can get into water; such activities include domestic, industrial, commercial or agricultural activities, surface runoff or infiltration. Wastewater can be purified by subjecting it to series of treatments so as to remove organic materials, unacceptable amount of pathogenic organisms, heavy metals and toxic substances present in it (Usharani *et al.* 2010). Untreated wastewater constitutes environmental and health hazards and must therefore be treated to prevent further harm to human health especially children below the age of five and the elderly above the age of sixty five which are more vulnerable (WHO 2006). A report by the World Health Organization (WHO) revealed that about 1.2 billion people lack access to safe drinking water while 3 out of every 5 persons lack clean and safe drinking water (Pritchard *et al.* 2009). Waterborne diseases such as diarrhea, cholera, dysentery and typhoid are fast becoming endemic in developing countries. A global estimate carried out by the WHO revealed that about 80% of diseases and sicknesses in the world are caused by lack of proper sanitation, use of polluted water or unavailability of drinkable water (Yongabi *et al.* 2011).

The conventional and well documented means of wastewater treatments include reverse osmosis, filtration, ion exchange, UV sterilization, coagulation, etc. However, these conventional techniques are quite expensive to run and maintain, non-eco-friendly and unsustainable especially in developing countries (Pritchard et al. 2009). Excessive exposure to disinfection byproducts of chlorine i.e. trichloromethane has been associated with an increased risk of bladder cancer (Villanueva *et al.* 2004), while aluminum sulphate (alum) has been linked to increase in the risk of Alzheimer's disease which causes cognitive failure in humans leading to disorder of thought, defective reasoning and impaired memory (Lukiw 2010, De

Sole *et al.* 2013). Furthermore, the unavailability and when available; the high cost of purchasing chemicals used for disinfection and coagulation of wastewater makes treated water very scarce and expensive especially for people in west Africa.

Research has shown that about 125 litres of drinkable water is required for consumption daily by an individual, yet, many people especially those in developing countries cannot boast of 25 litres of clean water in a day (Yongabi *et al.* 2010). This has therefore led to simple wastewater treatment using natural coagulant such as *Moringa oleifera* seeds which have been shown to possess both phytocoagulant and phytodisinfectant properties (Yongabi et al. 2010). The phytodisinfectant property of *Moringa oleifera* seeds is due to the presence of pterygospermin; a phytochemical present in the seed (Kurup *et al.* 1954). Two major mechanisms have been proposed for the coagulation property of *Moringa oleifera* seeds. The first mechanism is the adsorption and charge neutralization mechanism (Gassenschmidt *et al.* 1995, Ndabigengesere *et al.* 1995), while the second mechanism is inter-particle bridging (Muyibi and Evison 1995). Due to the small size of the *Moringa oleifera* coagulant protein (6.5 – 13kDa), an inter-particle bridging effect may not be the likely coagulation mechanism. The high positive charge and very small size of *Moringa oleifera* coagulant protein may suggest that destabilization mechanism could be charge neutralization and surface adsorption (Toma and Serawit 2014).

2.0 Coagulant Properties of *Moringa Oleifera* Seeds

2.1 Removal of dyes

Dyes especially those from textile industry contribute immensely to environmental pollution (Huantian and Ian 2001) and hence the need for their removal. Ali and El-Mohamedy (2016) using jar test apparatus studied the effect of changes in pH, temperature and initial dye concentration on the removal of three dyes. They reported that optimization of coagulant dose, contact time (90 minutes) and pH (8-10) using *Moringa oleifera* seed powder for removal of three reactive dyes; Reactive Blue 19, Reactive Blue 81, and acid red 27 showed up to 90% colour reduction which was studied by measuring absorbance values using Double Beam UV Spectrophotometer. The use of *Moringa oleifera* seed powder in the removal of five azo reactive dyes (Remazol Black B-133, Procion Blue H-EXL, Procion Crimson H-EXL, Procion Navy H-EXL and Procion Orange MX-2R) from textile industry effluents was reported. The experiments were carried out using *Moringa oleifera* seed powder extracted using sodium chloride salt solution and the one extracted with water. Fast stirring was done at 150 rpm for 10 minutes then 20 rpm for 30 minutes and allowed to stand for 24 h. The results obtained show up to 94% dye removal for *Moringa oleifera* seed extract with salt and up to 80% dye removal for *Moringa oleifera* seed extract without salt; using UV-Visible spectrophotometric measurements for the study. The optimum concentration dose is 750 mg/L and optimum pH is 9, though pH adjustment seems not to affect the efficiency of dye removal (Grimau *et al.* 2014). Orange 7 which is an azo dye was effectively removed at optimum conditions of biosorbent dose (0.4 g), initial dye concentration (20 mg/L), initial pH 6, contact time (5 minutes) and temperature 25^{o}C (Reza and Seyedeh 2011). Chicago Sky Blue 6B (CSB) which is also an azo dye was effectively removed up to 98% using *Moringa oleifera* seed extract (Jesus and Jesus 2008). They reported that as the pH level elevates, the efficiency of the removal process declines because of the cationic character of the *Moringa oleifera* protein and hydrophobic links being enhanced. They further reported that temperature has no significant effect, while initial dye concentration negatively affects the percentage of dye removal, although complete removal increases with a higher initial dye concentration. Sanchez-Martin *et al.* (2009) concluded that *Moringa oleifera* seed powder is an interesting natural coagulant in the removal of dyes. They however commented that pH is a very important factor that must be considered in the cases of indigoid and azo dyes, while its influence is rather small in the case of anthraquinonic dyes. The coagulation property of *Moringa oleifera* seeds extract in the removal of dyes from water and wastewater is due to its proteinic nature (Jesus and Jesus 2008).

2.2 Removal of Drugs

Pharmaceutical ingredients (biologically active component of a drug) constitute one of the emerging environmental pollutants in the world. They are a group of new products or chemicals without defined regulatory status whose adverse effects on the environment and human health are still largely obscure (Deblonde *et al.* 2011). Tetracycline which is a common antibiotic has been detected in amount beyond acceptable level in wastewater due to increase in the use of antibiotics for bacterial infections treatment in livestock and man (Kim *et al.* 2008). Excessive intake of untreated or poorly treated wastewater contaminated with antibiotics can have harmful effects and can enhance hepatoxicity. Andrea *et al.* (2015) using jar tests reported that 0.5 g/L *Moringa oleifera* seed extract were effective in the removal of tetracycline which was measured using solid phase extraction (SPE) followed by high-performance liquid chromatography – mass spectrometry (HPLC-MS) in synthetic water with removal rate of up to 55%. Norfloxacin (NOX) which is also an antibiotic was reported to be removed effectively (80% - 90% removal) using *Moringa oleifera* pod husk as adsorbents which were either chemically treated or carbonized. FT-IR frequency shifts indicate that NOX was bound to the absorbents through hydroxyl, amine, carboxylic, hydrogen bonding, and aldehydic groups (Wuana *et al.* 2015). 4-nitroalanine which is commonly used as a starting material in the synthesis of pharmaceuticals, dyes, antioxidants, and gasoline (El-Shahat and Shehata 2014) was effectively removed using *Moringa oleifera* seeds as chelating agent. Electrochemical study of the chelating property of *Moringa oleifera* seeds in the removal of 4-nitroaniline evaluated using voltammetric (CV) and square wave (SQWV) revealed up to 70% efficiency (Zaroual *et al.* 2017).

2.3 Removal of Organic Pollutants

Organic pollutants constitute major environmental problem due to their toxic effects and potential accumulation throughout food chain and hence in the body (Gaspard and Altenor 2014). *Moringa oleifera* seed powder was used for adsorption tests using jar test apparatus in the removal of organic pollutants from slaughterhouse wastewater and the results showed considerable reduction of Chemical Oxygen Demand of 5,614 mg/L (64%) with optimized values of 7 g/L of seed powder, pH 9 and 8,772 mg/L of pollutant in wastewater. The pH, dissolved oxygen, and conductivity were measured using a multiparameter digital metre (HACH HQ 40d) while the COD value was determined using HACH DR/2010 and the measurements of settled sludge volume were performed with an Imhoff cone (Jorge del *et al.* 2015). The major constituent of humus soil, peat and coal is humic acid (Seckler and Saito 2014). It is also a major organic component of many streams, lakes and ocean water (Stevenson 1994). Santos *et al.* (2012) reported that a dose of 1 mg/L *Moringa oleifera* seed powder protein was effective (up to 98%) in the removal of both total organic carbon and dissolved organic carbon from analytical grade humic acid using UV-visible spectroscopy and zeta potential measurements to monitor coagulation activity. Teixeira *et al.* (2016) reported the use of *Moringa oleifera* seeds as natural coagulant in surface water treatment using jar test equipment. The results obtained showed that some of the UV_{254nm} compounds were removed while DOC was increased. UV_{254nm} Compounds removal was approximately 40% for low turbidity water and less than 40% for high turbidity water using 50 mg/L of the MO extracted with ethanol. A report by Bakare (2016) also revealed the effectiveness of *Moringa oleifera* seed extract as natural coagulant in water treatment using jar test apparatus. The water samples from river were analyzed before and after treatment with *Moringa oleifera* seed extracts at varying concentrations (50 mg/L, 100 mg/L, 150 mg/L, 200 mg/L, and 250 mg/L) for different parameters – pH, conductivity, turbidity, and total solids. He reported significant reduction in all parameters analyzed with increased doses of MO with the exception of pH which was fairly constant at all doses unlike synthetic coagulant which alters pH value.

2.4 Removal of heavy metals

A toxic heavy metal is any relatively dense metal or metalloid with potential toxicity, and constitutes environmental pollution (Srivastava and Goyal 2010). The term heavy metal is particularly applicable to cadmium, lead, mercury, and arsenic (Brathwaite and Rabone 1985), all of which appear in the World Health Organization's list of ten chemicals of major environmental concern. Other examples include; nickel, chromium, cobalt, copper, manganese, zinc, selenium, silver, thallium and antimony. *Moringa oleifera* seed powder has been reported to be very effective in the removal of heavy metals such as iron, copper, arsenic, cadmium and lead. (Matar *et al.* 2017) using a PB – 700 6 Paddle Jar Test apparatus for wastewater treatment reported that 1% *Moringa oleifera* seed cake was effective in complete removal of iron, up to 98% removal of cadmium and copper while removal of lead was up to 78.1% measured using Perkin Elmer AAnalyst™ 800 high – performance atomic absorption spectrometer (AAS). There was reduction in turbidity up to 85-94%, with no significant alteration in pH, conductivity, salinity and total dissolved solids after treatment. The amount of dissolved oxygen, chemical oxygen demand and biochemical oxygen demand also increased significantly. Aziz *et al.* (2016) carried out a study using a standard jar tester (Lovibond, Phipps & Bird) for batch tests for determining the removal of the target contaminants using *Moringa oleifera* seed powder. The pH was adjusted to 7.0 using 0.1M NaOH in the batch experiment. The experiment was carried out by rapid mixing at 150 rpm for 3 minutes, followed by slow mixing at 30 rpm for 30 minutes. Optimum biomass dose of 200 mg/L *Moringa oleifera* seed powder with a contact time of 30 minutes led to the removal of up to 81% of lead which was measured using Agilent 7700 series Inductively Coupled Plasma Mass Spectrometry (ICP-MS) present in contaminated drinking water. Sajidu *et al.* (2005) used the jar test apparatus to evaluate coagulation and flocculation processes. They reported that the powdery form of polyelectrolytes from *Moringa oleifera* whole seed kernel was used in the treatment of synthetic heavy metal contaminated water with optimum dose of 2 g/100mL and a contact time of 60 minutes. This caused the removal of 92% iron, 89% lead and 48% cadmium measured using Atomic Absorption Spectroscopy (Bulk Scientific Model 2001) using the standard methods of the Association of Official Analytical Chemists (AOAC). Basra *et al.* (2014) investigated the coagulation property of *Moringa oleifera* seed powder (SP) and its seed aqueous extract (SAE) using batch test in the removal of lead (Pb) and Chromium (Cr). They explained that amino acids present in *Moringa oleifera* seed proteins accept protons from water and release hydroxyl group ions causing the solution's pH to be basic with natural buffering capacity. The electrical conductivity (EC) of untreated sewage water was lower than treated water due to solubility of minerals. Further decrease in EC of sewage water with *Moringa oleifera* seeds was due to the presence of lower molecular weight water soluble proteins which carry positive charge. The results revealed that at optimum dose of 2 g/L and 40 mL/L for SP and SAE respectively and contact time of 3 hours; *Moringa oleifera* SAE was more effective in the removal of lead and chromium from sewage water. Muthuraman and Sasikala (2015) reported that 0.5 g of *Moringa oleifera* seed powder was effective in the removal of up to 88% Cr (VI) with contact time of 120 minutes at a pH of 5. They explained that *Moringa oleifera* seed powder adsorption and inter-particle bridging takes place to remove Cr (IV) from synthetic aqueous solutions. Coagulating property of *Moringa oleifera* cake residue in the removal of certain heavy metals was investigated by Eman *et al.* (2014). The wastewater treatment was performed using a PB-700 6 Paddle Jar Test apparatus. Heavy metal removal test was done using Perkin Elmer AAnalyst^{TM} 800 High-Performance Atomic Absorption Spectrometer (AAS) while conductivity, pH, TDS, DO and salinity were measured using multifunction PCD650 waterproof portable meter. The results obtained show that there was 100% removal of iron; copper and cadmium removal were up to 98% while the removal of lead was 82.17%. Optimization of time, temperature and pH ensured cadmium removal of up to 70.7% using *Moringa oleifera* seed powder as natural coagulant. Ndabigengesere *et al.* (1995) reported that the biosorption of heavy metals from water and wastewater using *Moringa oleifera* seeds under non-reducing condition is due to the present of cationic proteins in *Moringa oleifera* seeds. They further reported that the mechanism of coagulation is adsorption and charge neutralization of the colloidal charges. Gassenschmidt et al. (1995) corroborated this report when he isolated flocculating protein with molecular mass 6.5kDa, isoelectric point above pH 10 and flocculant capacity comparable to a synthetic polyacrylamide cationic polymer. The result of Fourier Transform infrared spectrum of *Moringa oleifera* seeds extract showed that it has many functional groups such as hydroxyl, carboxylic and carbonyl groups (Reddy *et al.* 2010). A change in absorption band of hydroxyl group showed that the degree of hydroxyl polymerization in lignocellulose was decreased by binding of nickel. This observation clearly indicates that biosorption of heavy metals occur at the functional groups present on the surface of plant extract (Reddy et al. 2010). Obuseng *et al.* (2012) observed shift in the absorption peaks of hydroxyl and carboxylic acid groups of *Moringa oleifera* seed extract when used in the removal of heavy metals.

2.5 Removal of fluoride ions

High level of fluoride ions in the environment can cause serious harm in humans and animals (Wang *et al.* 2004). Fluoride content in drinking water has been associated with various health challenges such as dental caries especially in children (Mei *et al.* 2003). The removal of excess fluorine in water is therefore very important. Amir *et al.* (2014) reported the use of the natural coagulant *Moringa oleifera* seed extract in the removal of fluoride from water. The effect of *Moringa oleifera* dosage, initial fluoride concentration, and pH were determined and modeled using the Box-Behnken design response surface methodology. The results obtained showed that increasing the MO dose resulted in increase in fluoride removal while changes in pH have no significant effect on fluoride removal. Coagulant dosage of 900 mg/L ensured 85.4% removal of 8 mg/L of fluoride as rightly predicted by the model. A report by Ravikumar and Sheeja (2014) revealed the effectiveness of *Moringa oleifera* seed cake and its composite coagulants in the removal of fluoride from aqueous system. They observed that coagulation with aqueous extract of *Moringa oleifera* seed cake powder at optimum concentration of 2.5 g/L reduced the fluoride concentration of fluoridated water below 1 mg/L but the turbidity after coagulation was very high which was removed by double filtration. However, when composite coagulants prepared by adding alum and starch to the *Moringa oleifera* extract was used, the fluoride concentration reduced below 1 mg/L and turbidity was within the standard limit of drinking water.

2.6 Sludge conditioning

Sludge conditioning is a process whereby solids of sludge are treated with chemical or mechanical means to prepare the sludge for dewatering process. Mechanical methods for treatment of sludge include centrifugation, belt filter presses and gravity drainage while chemical treatments involve the use of alum, ferric chloride, lime and polyacrylamides (organic polymer). Inorganic chemicals have been found to increase the dry solids of processed sludge by 20-30%, while polymers do not (Metcalf. and Eddy 2003). This makes polyelectrolytes advantageous in terms of sludge handling costs and management. *Moringa oleifera* seed extract which contain cationic protein has been studied for sewage sludge conditioning (Ademiluyi 1988). A report by Ghebremichael and Hultman (2004) that compared the effectiveness of an extract from *Moringa oleifera*, alum and synthetic polyelectrolytes (Praestol 2540 TR which is anionic and Praestol 650 TR which is cationic) for conditioning alum sludge from Lovo drinking water treatment plant in Stockholm using CST, SRF, shear strength and sand drainage tests for comparison. The results obtained indicated that *Moringa oleifera* showed comparable conditioning effect as alum while synthetic polyelectrolytes were more effective than *Moringa oleifera* and alum. Conditioning using combination of *Moringa oleifera* and alum showed better dewatering characteristics than *Moringa oleifera* alone and showed similar results as alum. CST, SRF and sand drainage results obtained showed that optimum doses for *Moringa oleifera*, alum and polyelectrolytes were 125, 63, and 1.8 kg/t respectively. Marwan *et al.* (2016) carried out a research to find an optimum mixture ratio that consists of natural (*Moringa oleifera*) and chemical (Aluminium sulfate) coagulants, and using it for sludge dewatering. Optimization of process condition (first optimization) was done using three factors: *Moringa oleifera* dosage, pH, and mixing time while the second optimization was done to determine optimum mixture ratio between *Moringa oleifera* and alum using Quadratic Mixture Model Optimization. The specific resistance to filtration (SRF) was used as response for both optimization steps and the results obtained were analyzed using Analysis of Variance (ANOVA) approach. The results obtained indicate that optimum value obtained from the first optimization was 1.1E+11 mg/kg at dosage of 235.58 mg/L, pH of 6.5, and mixing time of 21.2 minutes at R^2 = 95.8%. For the second optimization, the optimum SRF value was 0.8E+11 m/kg for the ratio of 50:50 for alum and *Moringa oleifera* seed extract. Using mixed coagulant at ratio of 50:50 for alum and *Moringa oleifera* seed extract, sludge dewatering efficiency can be high with 50% elimination of alum use.

2.7 Treatment of dairy industrial wastewater and distillery spent wash

Wastewater generated by dairy industry is generally considered to be the largest source of food processing wastewater in many countries. These industries' wastewater is usually characterized by high BOD, COD, and nutrients. Patil and Ms-Manika (2015) reported that wastewater from dairy industry with initial pH, turbidity and COD values of 7.41, 289.5 NTU and 10000 mg/L respectively were treated with *Moringa oleifera* seed powder. The results obtained show that reduction of turbidity is

61.60%, COD reduction of 65% at 8000 mg/L with optimum dosage 0.1 g/500mL and optimum pH of 9. Parmar *et al.* (2012) reported that *Moringa oleifera* is an effective natural coagulant in the treatment of wastewater from dairy industry. Their results showed that turbidity, BOD and COD reductions were 75.29%, 60.17%, and 40.15% respectively. The optimum dosage was 100 mg/L at optimum pH of 9.

Distilleries are among the most highly polluting industries with reference to pollution of water. The quantity of wastewater generated from distilleries is large and it is characterized by high pollution load. Prasad (2009) using optimum response surface methodology (RSM) showed the effectiveness of *Moringa oleifera* seeds in colour removal from distillery spent wash. He studied the effects of dosage, pH, and concentration of salts for an optimized condition of colour removal from the distillery spent wash. The dosage (20 and 60 mL), pH (7 and 8.5) and concentration of 0.25 M had been found to be the optimum conditions of maximum 56% and 67% colour removal using sodium chloride (NaCl) and potassium chloride (KCl) respectively according to response surface methodology (RSM) analysis. The actual colour removal at optimal conditions was found to be 53% and 64% respectively for sodium chloride (NaCl) and potassium chloride (KCl) salts. These values are very close to those predicted by response surface methodology (RSM). David *et al.* (2015) carried out a study on performance evaluation of *Moringa oleifera* seed extract (MOSE) in conjunction with selected chemical coagulants for treating distillery spent wash. They studied coagulation-flocculation method of degradation of pollutants from the distillery spent wash. They assessed the effectiveness of the optimized dosage of chemical coagulants with or without the addition of *Moringa oleifera* seed extract (MOSE) in terms of percentage colour, COD and turbidity removal. At optimized dosages of ferric sulphate (2 g/L) and MOSE (40 mL/L), 97% colour removal was attained, closely followed by aluminium sulphate (1.5 g/L) and MOSE (40 mL/L) with 96.5% removal and calcium sulphate (4 g/L) with MOSE (40 mL/L) with 29.3% removal. They concluded that the treatment efficiencies were lower when the effluent was treated with chemical coagulants alone.

3.0 Conclusion

Moringa oleifera seed extract has been used extensively as coagulant for the removal of various contaminants from water and wastewater from different sources. *Moringa oleifera* seed extracts had been shown to be effective in the removal of heavy metals such as iron, cadmium, copper and nickel. The amino acids binding sites in the seed being responsible for the coagulation of heavy metals. The seed powder had also been used in the removal of dyes, organic pollutants, drugs, fluoride, sludge conditioning, treatment of dairy industrial wastewater and distillery spent wash. This shows that *Moringa oleifera* seed extract is an effective natural coagulant. Further research works are expected to focus on optimization of various parameters such as pH, contact time, surface area, particle size, etc which increase the effectiveness of *Moringa oleifera* as natural coagulant in the removal of various contaminants. Further study on the mode of attack of the seed extract on microbes in disinfecting contaminated wastewater should be extensively studied both alone and in combination with synthetic disinfectants. Furthermore, the effectiveness of *Moringa oleifera* seed powder in the removal of inner transition metals present in contaminated wastewater should be studied.

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