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2 AUTHORS:

Sushil Kansal
Panjab University

Amit Kumari
Panjab University

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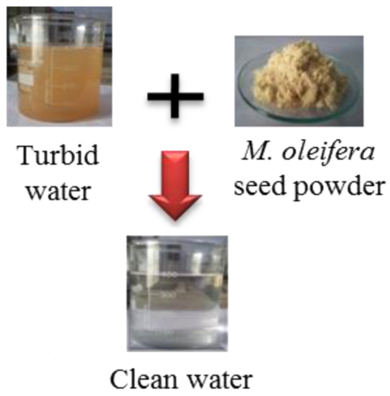
Potential of *M. oleifera* for the Treatment of Water and Wastewater

Sushil Kumar Kansal* and Amit Kumari

Dr. S. S. Bhatnagar University Institute of Chemical Engineering & Technology, Panjab University, Chandigarh-160014, India



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1. INTRODUCTION

"The hardest thing to see is what is in front of your eyes."

Goethe

The influx of significant quantities of organic, inorganic, and mineral substances into the environment has been increasing substantially over the past centuries due to natural calamities or by some anthropogenic activities such as excessive population growth, rapid industrialization, fast urban encroachment, and improved agricultural operations. Approximately 2 million tonnes of agricultural, sewage, and industrial wastes are being disposed of every day into the water courses worldwide. In developing countries, the major portion (~70%) of untreated water is discharged into the existing water bodies that lead to water pollution. According to the UN reports, the quantity of wastewater generated is nearly 1500 km^3 per annum.[1,2] As water is one of the basic necessities of life, addition of any natural or synthetic foreign material into water bodies affects all forms of life adversely. The detrimental effects of these contaminants with regard to health and environmental concerns are more prominent in underdeveloped countries especially in rural areas, where it is difficult to construct, operate, and maintain appropriate water/wastewater treatment systems.[3] Because of the restricted alternatives, the major source of water supply is surface water, which gets polluted due to the aforementioned reasons. Hence, diseases spread and people risk health problems and early deaths by consumption of polluted water. Increase in the mortality rate because of the use of contaminated water is an important issue for government and international institutions worldwide.[4] Of all of the diseases in developing countries, nearly 80% diseases are waterborne diseases, and the poor water quality has been found to be the key cause for the mortality of infants and children under five years of age.[5,6] Exposure to unsafe levels of arsenic and fluoride in drinking water leads to cancer and tooth/skeletal damage in millions of people. For example, in Bangladesh, it has been estimated that nearly 70 million people who consume groundwater are exposed to unsafe levels of arsenic (>10 μg/L).[7] An estimated 1.1 billion people still do not get clean and safe potable water, and these people are the world's poorest.[8] If the situation persists, it will lead to a substantial loss of human lives unless it is sincerely dealt with at all levels.

To mitigate the existing difficulties, steps should be taken toward the development of sustainable and cost-effective water treatment systems, which demand less maintenance and minimal operator's skills. Naturally occurring substances can be explored toward achieving safe and clean water supplies. Locally available materials have been used as coagulants and biosorbents in water treatment since ancient times. According to reports by Baker, the earliest historical accounts by ancient civilizations for the use of plants or their derivatives for water treatment have been registered in Sanskrit writings (400 A.D.), the Old Testament, and Roman records (77 A.D.).[9] Yet due to

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the lack of scientific understanding of their effectiveness and mechanism of action, they could not compete with commonly used chemicals, so their use diminished subsequently, and it remained limited to some remote areas of underdeveloped countries only.[10] Recently, however, a resurgence of interest in natural coagulants has emerged due to high cost factor, health related issues, and environmental impacts associated with synthetic organic/inorganic coagulants.[11−20] Various plant-based materials have been identified as effective coagulants. The major merits of plant-derived coagulant materials when used as point-of-use (POU) technology in water treatment methods are obvious; these are less expensive, do not alter the pH of treated water, and the sludge they produce is less voluminous and readily biodegradable. The history of using powdered roasted maize grains (*Zea mays*) to settle impurities by Peru soldiers is long. The use of Nirmali tree seeds (*Strychnos potatorumas*) for clarifying turbid river water in India about 4000 years ago is also well documented.[21] In Chili, the sap of tuna cactus (*Opuntia fiscus indica*) is being used extensively to purify water. Tunafloc A and B are commercially available products.[22] In India, seeds of Strychnos potatorun are still being used in some remote areas of Maharashtra and Tamil Nadu for clarifying turbid water.[23] Some other plant-based effective coagulants include Okra,[24] *Cactus latifaira* and *Prosopis juliflora*,[25] tannin from valonia,[26] orange peels, apricot, peach kernel, and beans,[27] and maize.[28] One of the natural coagulants identified from animal origin is chitosan.[29]

Of all of the plant materials investigated, the seeds of *Moringa oleifera* Lam. (*M. oleifera*) have drawn special attention as it treats water by acting both as a coagulant and as an antimicrobial agent.[8,10,15,30−43] Investigations have been carried out since the early 1970s to evaluate its efficacy for water treatment.[44]

M. oleifera is cultivated across whole of the tropical belt for different purposes. This tropical plant belongs to the family Moringaceae and is one of the 14 different species of this family so far identified.[14,15] It is a cosmopolitan, drought tolerant tree available throughout the year. It can grow well even on the soil with relatively low humidity.[45] *M. oleifera* being native of the western and sub-Himalayan tracts, India, Pakistan, Asia Minor, Africa, and Arabia, is now found in Cambodia, Philippines, North and South America, Central America, and the Caribbean Islands.[46−48] Common names of *M. oleifera* are Sahjan, Moringa, Drumstick tree, Horseradish tree, Surajana, etc. In the Nile valley, Moringa is commonly called “Shagara al Rauwaq”, which means “tree for purifying”.[49] In Pakistan, this plant is grown and cultivated all over the country and is known by a common name “Sohanjna”.[50,51] It has widespread uses in agriculture, medicine, and industry. In India, cultivation of this tree occurs mainly in the southern region including the states of Karnataka, Tamil Nadu, Kerala, and Andhra Pradesh.[52] An average per hectare yield of *M. oleifera* seeds has been reported to be 3 tonnes against the average yield of sunflower and groundnut seeds, which are two tonnes and half tonnes, respectively. Every part of the *M. oleifera* tree has a use that is well documented in the literature.[53−57] Oil extracted from *M. oleifera* seed kernel (about 35%)[58] is slightly yellowish in color and usually known as “ben oil” or “behen oil”. In ancient periods, India used to export this oil to Europe, and it was used in various perfume industries, soap making, and as a lubricant in delicate machinery. The plant leaves are generally consumed as vegetables and are currently the focus of several development projects. *M. oleifera* is recognized as an ideal nutritional supplement because it contains several essential amino acids, minerals, and vitamins in high concentrations. It provides vitamins, minerals, and proteins to the mother and child during pregnancy, thus reducing the mortality in infants.[8,59] Regular intake of *M. oleifera* may also prove helpful to prevent anemia and most forms of malnutrition. Its seed contains a water-soluble substance[35] whose use has been recommended as a coagulant in developing countries.[4,13−15,19,23,60−65]

The toxicological assessments by Berger et al.[66] and Grabow et al.[67] have already indicated that there is no threat to human health in using *M. oleifera* as a primary coagulant. However, to get regulatory approval in a specific country, the safety of any proprietary seed product needs to be established by toxicological testings.[68] This natural material is probably the most studied coagulant among the environmental scientific community, and, encouraged by the result of these studies, various scientists have come up with treatment techniques of water and wastewater in small and pilot-scale trials.[32,39,68−75]

The main aim of this Review is to present an overview followed by critical analysis of the use of *M. oleifera* as a sustainable practice for water and wastewater treatment in varying conditions, its processing techniques and mechanism of coagulation, and applications as a coagulant and biosorbent. This Review has been divided into seven sections, which are concerned with details of (a) active component responsible for coagulation, (b) processing techniques, (c) comparison with conventional coagulants, (d) water and wastewater treatment using *M. oleifera* seeds, (e) sludge disposal, (f) effect of storage conditions and its effectiveness, and (g) cost.

2. *M. OLEIFERA* SEEDS AS COAGULANT

Coagulation−flocculation is an established technology having worldwide applications in water and wastewater treatment. This process is of prime importance for removing suspended and dissolved impurities. Because of the public’s enhanced awareness, more attention has been focused on environment friendly and eco-safety measures based on natural coagulants to combat the prevailing difficulties. *M. oleifera* seeds are one of the natural materials that can act as primary coagulant. In recent years, its use for water treatment has gained popularity, and ongoing research has attempted to characterize and purify the active component.[29,35,76,77] It has been reported that only ripe *M. oleifera* seeds (with brown seed coat) give high coagulation efficiency.[14,78] Most of the coagulant activity is present in the seed cotyledons. Bark around the seeds has no coagulation properties.[35] Seeds from different geographic locations exhibit variations in their coagulating properties, which may have to do with differences in the growing conditions of the seeds and their protein content.[79] Gassenschmidt et al.[76] reported the presence of more than one protein family with flocculating activity in *M. oleifera* seed. Indeed, various genomics projects have demonstrated that during different developmental stages of plants, many closely related proteins are expressed; therefore, it is not uncommon that a large number of sequence variants of *M. oleifera* seeds are produced, which exhibit coagulant activity.[80,81] Yet the complete array of proteins, possessing the coagulation and antimicrobial properties, has not yet been fully identified in the seeds. So to identify and characterize the whole range of proteins with their amino acid sequences and structure, extensive research is needed.[29] Although the nature and characteristics of active coagulating agent have been described by a number of researchers, the properties of the active component have been reported to be influenced by the extraction and purification method used.

2.1. Active Component

In the previous studies, the active ingredients of *M. oleifera* have been documented to be cationic proteins with molecular weight in the range of 6−16 kDa and with an isoelectric pH value of 10.[82,83] Further, the active components were suggested to be dimeric cationic peptides having isoelectric point (pI) above 10 and molecular mass of about 6.5 kDa[76] and 13 kDa.[35] Since then, inconsistent reports have been received about the precise nature of the active coagulating component of *M. oleifera* seed. Okuda et al.[77] argued that the active component extracted from an aqueous salt extraction is an organic polyelectrolyte of unknown structure with molecular weight of approximately 3.0 kDa. The isoelectric value of this nonproteinic organic component was found to be 8 and above. Further results of Ghebremichael et al.[81] revealed it to be a cationic protein with isoelectric point greater than 9.6 and molecular mass less than 6.5 kDa. This finding was not matching with the results of Okuda and co-workers.[77]

Although most of the researchers agree with the finding that the active component is a cationic protein, the results by Okuda et al.[77] cannot be ignored as there can be a myriad of unrevealed coagulating agents in *M. oleifera*.[84] It has also been reported that more than one coagulant peptide can be isolated from the seed, and the sequence of one of them (identified as $MO_{2.1}$) has been established.[76] The recombinant form of $MO_{2.1}$ was found to have good flocculation and antimicrobial properties capable of disinfecting heavily contaminated water.[41,42] Suarez et al.[85] justified the previous results by identifying a synthetic peptide that too mediated antibacterial activity against specific human pathogens.

2.2. Coagulation Mechanism

The coagulation mechanism of the active component of *M. oleifera* seed has been described as adsorption and neutralization of charges[35,76] and interparticle bridging by Muyibi and Evison.[33] Ndabigengesere et al.[35] explained that as the cationic proteins predominate, the zeta potential of *M. oleifera* solution is positive, and the zeta potential of synthetic water is negative; hence destabilization of negatively charged colloids of kaolin suspension takes place by cationic polyelectrolytes of *M. oleifera* solution. Although with over-dosages of coagulant the zeta potential of kaolin suspension is reversed to positive, it does not lead to restabilization of the colloids. Therefore, adsorption and charge neutralization or adsorption and bridging of destabilized particles appear to be the main coagulation mechanism. It was also stated that the two mechanisms may be taking place simultaneously.

Okuda et al.[86] stated that for coagulation by nonproteinic organic compound, the coagulation mechanisms such as compression of double layer, interparticle bridging, or charge neutralization were not responsible. The coagulation mechanism of nonproteinic organic component was described as the enmeshment by a net-like structure, that is, sweep coagulation. It was stated that at pH 9.0, the coagulation active component has negative charge. The zeta potential of MOC-SC-PC solution increased slightly with increase in coagulant dose, but even at optimum dose it remained negative. Therefore, coagulation by charge neutralization was not likely to occur. Being small in size, the active component (MOC-SC-PC) could also not induce interparticle bridging.

According to Ghebremichael,[29] due to high positive charge (pI above 9.6) and low molecular weight of MOCP (<6.5 kDa), the main destabilization mechanism could be adsorption and charge neutralization. Fahmi et al.[87] described the mechanism of simultaneous removal of hardness and turbidity in drinking water using *M. oleifera* by employing Langmuir and Freundlich isotherms. The mechanism for the removal of turbidity was supposed to be adsorption and charge neutralization with adsorption isotherm following the Freundlich adsorption model. It was assumed that the possible mechanism of removal of hardness was adsorption that could be modeled by both Langmuir and Freundlich models. MOC-SC led to removal of hardness by forming a net-like structure followed by the sweep coagulation mechanism for turbidity removal.

2.3. Processing Techniques

In the 1970s, Sudanese women had their own way to use the seeds of *M. oleifera* for water treatment. They had a technique that involved swirling of a cloth bag (containing crushed seeds) in a container filled with dirty water for a few minutes and allowing it to settle for an hour (Figure 1).

Figure 1. Earlier method of drinking water treatment using *M. oleifera* seed powder.

Nowadays, mainly three steps are involved in the preparation of *M. oleifera* coagulant. These are classified as primary, secondary, and tertiary (Figure 2). The primary processing stage is very straightforward and is generally used to clarify turbid water at household level. In this stage, the seedpods are allowed to mature on the tree before harvesting. The dried seeds are removed from the pods, and the husk covering each seed is taken off manually to get the kernel. Good quality seeds are then selected and crushed to the fine powder. The dried pods and husks can be subjected to pyrolysis for activated carbon production.[88] After that, the vegetable oils are extracted first from the powder with the suitable solvent such as petroleum ether, and then the press cake is used as a coagulant.

Because this crude *M. oleifera* seed extract not only contains just coagulating active agents but also plant tissues that lead to an increase in the organic load in treated water, hence further processing of crude extract is essential (secondary and tertiary treatment). Secondary processing stage involves the extraction of coagulant protein from crushed powder or press cake by water or salt solutions. The performance of MOC-DW for

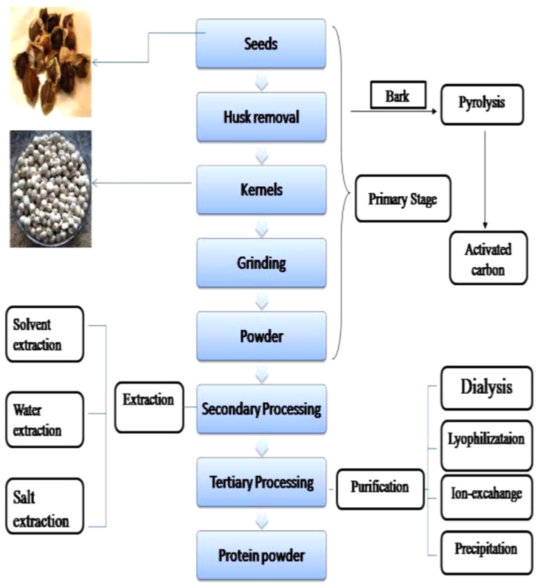

Figure 2. Processing steps for *M. oleifera* as a coagulant.

removing turbidity has been assessed by various research groups.[10,36,43] Extraction with water is commonly considered as the best method because of its easy availability and cost effectiveness and for the reason that its active component is a water-soluble protein. Several studies have been carried out using MOC-DW as an alternative coagulant or coagulant aid for water and wastewater treatments.[15,34−36,61,89] Yet the problem of residual DOC associated with the use of crude extract (MOC-DW) as coagulant renders its use in drinking water nonfeasible.[10,14] DOC is often considered to be a source of odor, taste, and color and a precursor of disinfection byproducts during water treatment.

The active component has also been extracted with salt water from *M. oleifera*, and its efficacy has been compared to that of the distilled water extracted coagulating component.[77,90] The amount and effectiveness of coagulant extracted by both methods varied significantly. The salt extract of *M. oleifera* exhibited better coagulating properties as compared to the water extract in its crude form. This may be attributed to the higher amounts of soluble protein present because of the salting-in mechanism; that is, the increased ionic strength due to salts results in increased solubility of active ingredients. Madrona et al.[91] compared the coagulation efficiency of *M. oleifera* extract prepared by using different concentrations of KCl solution and pure water for the color and turbidity removal of raw water. The best results were obtained when 1 M concentration of salt was used. In continuation to their previous study, Madrona et al.[92] further evaluated the efficacy of *M. oleifera* extracts prepared by using KCl, NaCl, and $MgCl_2$ salts with respect to removal of color and turbidity of raw water. The results showed no difference in the coagulation efficiency of different salt extracts, but it was higher than the water extract. Montakhab et al.[93] determined the effect of spray drying on the coagulation efficiency of *M. oleifera* seed extracted with $NaNO_3$ salt and achieved better coagulation performance and high turbidity removal.

However, the organic, nitrate, and phosphate contents of water have been reported to increase in case of crude MOC with either MOC-DW extracted or MOC-SC extracted, whereas it does not increase in case of purified MOC.[10,77] Therefore, in a view to overcome the limitations of crude extract, many researchers have worked on purification methods, for example, dialysis, delipidation, centrifugation, ion exchange, and lyophilization.[35,77] Okuda et al.[77] reported an increase of up to 34 times in coagulating activity of active component after ion exchange than the crude extract. Yet as these methods require a number of steps for MOCP purification, making the purification process cumbersome, costly, and difficult to use in large-scale treatment applications, a single step purification method (ion exchange method) was proposed by Ghebremichael and co-workers.[81,94,95] To further increase the performance and to minimize the concerns of residual DOC, Sánchez-Martín et al.[96] proposed a two-step procedure for purification of the coagulant from *M. oleifera* seed. They reported that relatively lower doses of two-step purified protein produce quality similar to that of single step purified protein.

Because the tertiary stage may enhance the processing costs, practically it may not be used as a POU water treatment technology. This stage is rarely implemented in case of plant-derived coagulants and is restricted currently for the purification of *M. oleifera* extracts for academic research purposes only.[36,81,86] Nonetheless, there can be a wide scope for the researchers to determine a cost-effective, sustainable way for the extraction and purification of active coagulating agents, so that this process can be commercialized at the industrial level.

2.4. Comparison with Conventional Coagulants

The most commonly used chemical-based coagulants in conventional water treatment processes are aluminum and iron(III) salts and synthetic polymers. All of these coagulants have the ability to produce positively charged ions when dissolved in water, which contributes to charge neutralization.[97] Aluminum salts are used throughout the world due to their excellent performance, easy handling, and availability.[98] Although the effectiveness of these chemical coagulants is well-recognized,[99,100] they do have numerous disadvantages associated with their usage such as they are ineffective at low temperature,[101,102] require high procurement cost, produce huge volumes of non biodegradable sludge, and affect human health adversely, and the fact that they alter the pH of treated water leading to significant changes in the chemistry of water.[84]

Moreover, when alum is used for drinking water treatment, its concentration increases in the finished water.[42] Also, the intake of treated water containing residual aluminum by human body leads to several neuropathological diseases, the most detrimental being Alzheimer's disease and other similar health-related problems.[103−108] Other chemical coagulants such as ferric salts and synthetic polymers/polyelectrolytes have also shown limited success as they too have disadvantages similar to those of aluminum salts.[98,109−114] In many underdeveloped countries, the use of synthetic chemicals is restricted due to the high costs of these chemicals for water treatment.[21,115,116] Hence, to counteract the aforementioned drawbacks, *M. oleifera* seed extract has been proposed as a substitute to conventional coagulants in less fortunate countries. Dorea[117] has described the benefits of coagulation with *Moringa* spp. as a sustainable option for water treatment in his review paper.

Ndabigengesere and Narasiah[10] compared the quality of water treated with seeds of *M. oleifera* to that of water treated with alum. It was observed that seeds of *M. oleifera* did not alter the pH value of water for any of the tested doses of *M. oleifera*, whereas the pH value decreased with alum, thereby making the addition of chemicals necessary for maintenance of pH of treated water. Change in the conductivity of finished water was also not significant in case of *M. oleifera*, whereas it increased considerably with increase in doses of alum, indicating the chemical coagulant to be relatively disadvantageous as compared to *M. oleifera*. Alkalinity of the treated water also remained nearly the same with *M. oleifera* seeds, while a rapid decrease in alkalinity was observed in case of alum. Because of this decrease in alkalinity and pH and increase in ionic strength, an imbalance is caused in the water chemistry that subsequently leads to corrosion problems in the distribution network.[35] Therefore, addition of lime is a common practice to add alkalinity in alum treated water, but it leads to increased treatment cost and high sludge volume. The sludge of alum is voluminous as compared to that of *M. oleifera* due to the production of aluminum hydroxide precipitates. Apart from being huge in volume, alum sludges are acidic, gelatinous, and difficult to dewater and dispose of in the environment;[106] while *M. oleifera* sludge is a biodegradable organic, it could also have an advantage of nutritional value when considering land application. Land application of alum sludge may not be desirable as alum sludges can present Al phytotoxicity, and it may absorb inorganic phosphorus from the soil, thus inhibiting phosphorus uptake of plants.[118]

In light of some disadvantages associated with the use of *M. oleifera* such as the seeds tend to increase the organic load considerably in the treated water, the use of its crude extract is not recommended for the removal of phosphates and nitrates. It may be used only after suitable purification of its cationic active proteins.

Many researchers reported that for a turbidity value higher than 100 NTU, similar turbidity reductions were achieved with *M. oleifera* seed extract and alum.[12,35,119] Figure 3 shows 90% reduction in turbidity with alum, SMOS, and NSMOS at optimum dosages. Yet, at low turbidity values (<50 NTU), *M. oleifera* seed extract achieved at the most 60% turbidity reductions, while alum achieved 75% reduction.[34] Ghebremichael and Hultman[120] compared *M. oleifera* to alum and synthetic polyelectrolytes for dewatering the chemical sludge of a water treatment plant and concluded that both *M. oleifera* and polyelectrolytes could improve the filterability of the sludge. Although alum showed slightly better results than *M. oleifera* alone, when *M. oleifera* and alum were used together, similar results were achieved as with other polyelectrolytes. Mandloi et al.[16] evaluated the effectiveness of natural coagulants and compared it to alum for removing turbidity and microorganism from lake water. It was observed that the turbidity of the filtered water was reduced to nearly 1 NTU with natural coagulants satisfying the WHO drinking water guidelines. Additionally, natural coagulants showed better performance for reduction of bacterial load than alum.

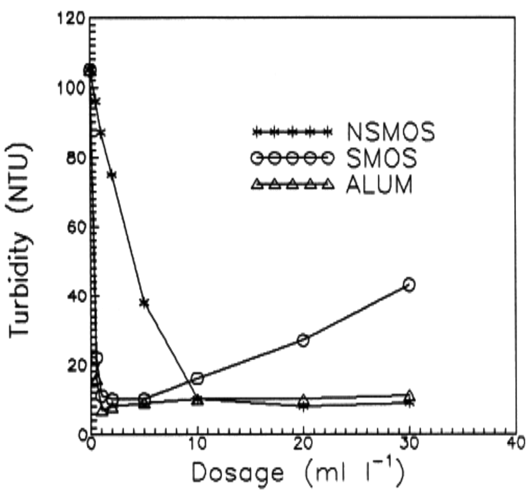

Figure 3. Turbidity removal by coagulation with nonshelled *Moringa oleifera* seeds (NSMOS), shelled *Moringa oleifera* seeds (SMOS), and alum (ALUM). Reprinted with permission from ref 10. Copyright 1998 Elsevier.

Ghebremichael et al.[81] compared the coagulation activity of *M. oleifera* extract with alum for high turbidity (250−300 NTU) and low turbidity (76−110 NTU) samples. Similar results were observed with both coagulants for high turbidity samples, but for low turbidity samples alum showed better coagulation activity. Gunaratna et al.[121] screened different plant seeds to find their usage as a primary coagulant in clarifying drinking water and suggested that the seeds from natural plants could replace chemical coagulants being used for water purification.

Arnoldsson et al.[43] in their piece of work investigated the use of *M. oleifera* combined with direct filtration. Their study showed that coagulation with alum led to more efficient treatment, but *M. oleifera* could also produce water of acceptable quality. *M. oleifera* did not alter the chemistry of water and was relatively more efficient for high initial turbidities. Abaliwano et al.[8] noticed similar findings with respect to change in pH value and conductivity as were reported by Ndabigengesere and Narasiah,[10] for alum and ferric chloride. Increase in DOC with increase in MOCP dose could be attributed to the presence of residual coagulant protein in the treated water.

It has been found that inorganic salts like alum and ferric chloride leave their residues in the treated water.[8,122,123] Yarahmadi et al.[124] compared the efficiency of *M. oleifera* to that of polyaluminium chloride. *M. oleifera* seeds showed higher efficiency for the removal of high turbidities in comparison to lower turbidities and had a minimal effect on pH. Reduction of pH decreased the efficiency of polyaluminium chloride in turbidity removal.

Pritchard et al.[75] commissioned a research project to evaluate the coagulation activity of *M. oleifera* in comparison to alum and ferric in terms of turbidity removal and reduction of total coliforms for different types of waters. Model water samples were created with desired coliform counts and turbidity levels by kaolin suspension. Raw water samples for testing were taken from three natural sources having different turbidity levels. Hybrid water samples were generated by mixing both types of water samples. Reduction in total coliforms was found to be directly linked to the removal of turbidity in treated water samples. Although *M. oleifera* could not perform as well as alum and ferric did for the samples tested under this project, it met the WHO guidelines, which encourages its use to treat water in many developing countries. Sarpong and Richardson[125] compared the efficiency of *M. oleifera* seed extract and conventional aluminum sulfate with respect to turbidity removal and total coliform reduction for a river water source. The entire estimated equivalent dose based on applied coagulant mass and removal of turbidity was comparable between the two coagulants.

Ndabigengesere and Narasiah[126] also proposed *M. oleifera* seeds as a viable substitute to alum worldwide. Sánchez-Martín et al.[127] tested and compared the turbidity removal efficacy of different synthetic flocculating agents, (i) Flocudex-AS/10 and AS/23, (ii) Flocudex-CS/41 and CS/49, and (iii) aluminum sulfate, with *M. oleifera* seed extract. Under similar conditions, *M. oleifera* showed coagulant ability comparable to that of CS/49 flocculant and alum coagulant. The ability of *M. oleifera* was observed to be higher than that of the other synthetic flocculants.

Some workers[36,70,128,129] have reported that *M. oleifera* can be used as a coagulant aid or in synergy with alum effectively, and its use with alum could bring savings in the quantities of alum required for coagulation. The use of *M. oleifera* seeds or its extract mixed together with alum has been termed as cocoagulation. In this mode, Sutherland et al.[119] reported reduction in alum usage in the range 50–80%, and others recorded alum savings of 40%.[34,36] Dalen et al.[128] showed characteristic synergies in water treatment by blending alum and *M. oleifera* seed powder rather than using alum or *M. oleifera* alone. The optimum dose reduced the alum requirement by 40–60%. It reduced the cost of treatment as well as the risk of alum-related diseases by about 40–60%.

3. WATER AND WASTEWATER TREATMENT WITH *M. OLEIFERA* SEEDS

3.1. Drinking Water Clarification and Disinfection

Water is a natural resource that is vital to the presence of life on earth. Although it exists in abundance, only a small fraction of it is fit for human consumption. It links human beings, poverty, health, and education.[130] Clarification of suspended particles, that is, turbidity and disinfection of water, is of paramount importance for making it fit for human use.

3.1.1. Turbidity Removal. Raw water drawn for human consumption becomes highly turbid due to the addition of solid particles (clay and silt), organic and inorganic compounds, and many other micro- and macroscopic organisms.[131] The increased turbidity levels of water reduce the process of photosynthesis, thereby affecting the aquatic ecosystems and food chain adversely. The consumption of highly turbid water may constitute a health risk. This can lead to waterborne disease outbreaks. The removal of turbidity in water would be extremely beneficial as it would alleviate the majority of problems associated with turbidity. Coagulation has been identified as one of the most effective techniques for removal of turbidity from water. In developing countries, water treatment companies utilize alum and other chemicals for coagulation, but as these chemicals are not locally available in all places, they have to be imported, which requires high foreign exchange. Hence, the use of locally available plant materials such as *M. oleifera* either alone or in combination with other coagulant materials may prove highly beneficial. Turbid water-clarifying properties of *M. oleifera* were first reported by Jahn after observing the use of its seeds by Sudan women for clarifying turbid Nile water. Since then, *M. oleifera* has gained the sudden interest of researchers and has been significantly studied as a coagulant and disinfectant.[32,90,132−134] On average, 92–99% turbidity reductions have been reported using *M. oleifera* seeds as coagulant.[135] Figure 4 shows the efficacy of this plant-derived coagulant for turbidity removal.[136] Being cationic in nature, increased dosages of *M. oleifera* beyond optimum result in saturation of polymer bridge sites. This will ultimately lead to reversal of charges, and subsequently destabilized particles will be restabilized resulting in increased residual turbidity in water samples. So optimum dose is required for maximum turbidity removal.

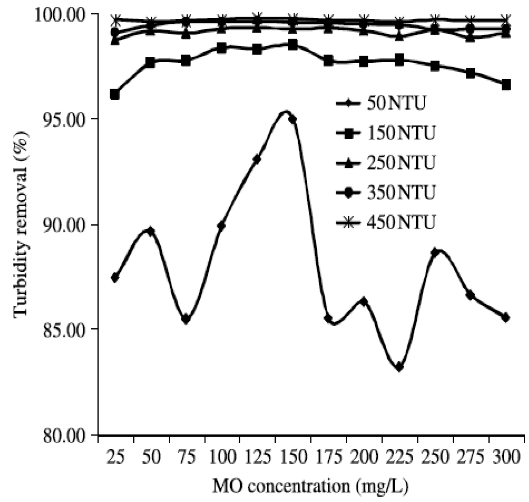

Figure 4. Turbidity removal (%). Reprinted with permission from ref 136. Copyright 2009 IWA Publishing.

3.1.2. Water Disinfection. Water disinfection is highly beneficial. The two most commonly used chemical additives for water disinfection are chlorine and chloramine.

M. oleifera seeds have been reported as an ecofriendly substitute to widely used disinfectants. Eilert et al.[30] isolated an active antimicrobial agent from the seeds of *M. oleifera*, α L-rhamnosyloxy-benzyl isothiocyanate. Madsen et al.[31] observed 80–99.5% turbidity removal and 90–99.99% bacterial reduction within the first 1–2 h of treatment with *M. oleifera* seed material as coagulant while treating Sudanese water by employing the water purification method traditionally used in Sudan.

Suarez et al.[42] identified and characterized one of the cationic *M. oleifera* seed polypeptides, which could coagulate suspended particles and facilitate sedimentation more efficiently for water cleaning. In addition, it was also found to possess an antimicrobial activity and was able to clear water from several waterborne human pathogens. Broin et al.[41] tested the potential of a flocculant protein $MO_{2.1}$ purified from *M. oleifera* seeds toward the aggregation of bacteria and clays for water treatment. Ghebremichael et al.[81] discussed the flocculating and antibacterial effects of the coagulant protein purified by ion exchange technique from *M. oleifera* extract and stated that plants are prolific sources of coagulating peptides for water and wastewater treatment applications. Further, Ghebremichael et al.[137] tested the antiflocculating and antimicrobial properties of the purified protein for the treatment of surface water, using it as a primary coagulant and as a coagulating aid with metal ions. It was observed that when used in combination with metal ions, its addition before the metal ions showed better results in terms of removal of turbidity and reduction of organic load than its addition after the metal ions. Poumaye et al.[138] used *M. oleifera* dried powder for clarifying the river M'Poko surface water. They employed a factorial design to determine the importance of various factors and their interaction effects with a great precision. The optimization confirmed the coagulant properties of *M. oleifera* for water clarification. A significant reduction in turbidity was observed after treatment with *M. oleifera*, which was further lowered by following with a filtration step after treatment. Clarification by *M. oleifera* also resulted in 62%, 95%, and 47% elimination of *streptococci*, *clostridium*, and *E. coli*, respectively.

Sánchez-Martín et al.[127] optimized different parameters to improve the efficacy of flocculation process using *M. oleifera* seed extract for clarification of water. The coagulation process was slightly enhanced in acidic pH, and the optimum stirring rate was identified as 80 rpm. Significant reductions in microbial colonies were also observed. Even up to 72 h of treatment, no significant growth of microorganism was detected in the treated water (Figure 5).

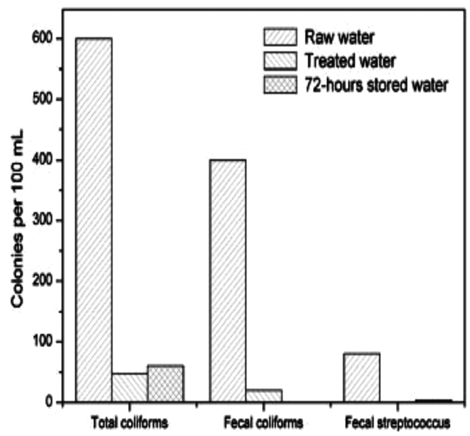

Figure 5. Evolution of the microorganism population (16 mg L^{-1}, pH 7, 20 °C, standard jar test). Reprinted with permission from ref 127. Copyright 2012 ABEQ – Brazilian Society of Chemical Engineering.

Sengupta et al.[139] conducted field-based and laboratory-based trials to investigate the effect of *M. oleifera* seed extract in reducing Helminth egg numbers and turbidity in irrigation water, wastewater, turbid water, and tap water. *M. oleifera* was found to be able to reduce Helminth eggs by 94–99.5% and turbidity by 80–96% in different types of water samples. A strong correlation was seen between turbidity removal and Helminth egg reduction in turbid water and wastewater.

Various other studies related to the removal of turbidity and water disinfection using *M. oleifera* have been presented in tabular form (Table 1).

3.2. Wastewater Treatment

3.2.1. Removal of Metal Ions.

Toxic metals cause environmental pollution through metal smelters, industrial operations, agricultural practices, and improper disposal of solid waste.[143] Most of the heavy metals have long biological life. Being non biodegradable, they accumulate and human beings get their exposure through food and water. Heavy metals, which affect the environment most severely, include arsenic, copper, cadmium, lead, mercury, nickel, and zinc. Their presence in the environment, even in low concentrations, may prove toxic and lead to dermatological problems, respiratory diseases, and sometimes cancer.[144−146] Depending upon the concentration of metal ions in water, various conventional methods have been used to remove these metals from wastewater such as precipitation, oxidation/reduction, coagulation–flocculation, electro-coagulation, ultrafiltration, solvent extraction, adsorption, biosorption, and ion-exchange, etc.[147,148] Recent studies have described the use of *M. oleifera* as a potential alternative natural biosorbent to remove toxic metals.[149−154] Different mechanisms have been proposed by different co-workers for the sorption properties of SMOSP for metal ion removal.

Kumari et al.[143] explored the sorption property of SMOS for decontaminating arsenic (As III and As V) from water. They found protein/amino acid–arsenic interactions to be responsible for biosorption process.[150]

Sajidu et al.[155,156] explored the potential of *M. oleifera* for the removal of lead, iron, and cadmium metal ions. They reported that *M. stenopetala* and *M. oleifera* could be used successfully to remove Cd^{2+}, Cr^{3+}, Zn^{2+}, and Cu^{2+} from water. Further, they stated about the removal of metals to be pH dependent as each metal was removed within a specific range of pH. It was suggested that the hydrogen ion–metal cation exchange mechanism and metal hydroxide precipitation were involved for metal removal process using extended X-ray absorption fine structures (EXAFS).[156] Mataka et al.[157] used *M. stenopetala* and *M. oleifera* seed powder for lead detoxification of water and found *M. stenopetala* to be more effective for removing lead from water. Sharma et al.[158] explored the potential of *M. oleifera* for decontamination of Cd and found amino acid–Cd interactions to be responsible for the sorption process through FTIR spectrophotometry. Later, Sharma et al.[151] investigated the competitive biosorption of Cd(II), Cr(III), and Ni(II) on SMOSP present in a ternary mixture. The selectivity order of metal ion removal was seen to be Cd(II) > Cr(III) > Ni(II). They also attempted the regeneration of used biomass for its effective reuse. Bhatti et al.[159] investigated the removal of Zn(II) ions from water using native and chemically modified *M. oleifera* biomass and observed that the biosorption capacity of *M. oleifera* biomass increased after its pretreatment with alkalis, acids, and surfactants.

Sajidu et al.[160] in continuation of their previous studies explained different uptake mechanisms for sorption of heavy metals by *M. oleifera* water extracts. Cr(III) cations were shown to be sorbed as hydrolyzed octahedral trimeric complexes as indefinite chains. Cu(II) sorption was through complexation at neutral pH value by coordination to carboxylate groups, whereas Hg(II) was sorbed through the nitrogen as a donor atom. Beltrán-Heredia and Sánchez-Martín[161] explored the capability of *M. oleifera* seed extract as flocculant for removing Cu^{2+}, Zn^{2+}, and Ni^{2+} metal ion from surface water and stated that metal removal is linked to turbidity removal; turbidity and metal ion contact lead to a higher process effectiveness. Boucher et al.[153] investigated the use of *Brassica napus*, *M. oleifera*, and *Glycine max* press cake as a biosorbent to remove metal ions from wastewater. The findings showed that the use of plant-derived byproducts represents a domestic, inexpensive, and environmentally friendly technique for the removal of toxic metal ions. Jamal et al.[162] explored the sorption properties of *M. oleifera* active ingredient to remove cadmium from contaminated water. They carried out the statistical optimization for evaluating the polynomial regression model through the effect of linear, quadratic, and interaction of the factors. Kalibbala et al.[163] assessed the efficacy of *M. oleifera* as a coagulant aid for the removal of humic materials and iron content from drinking water. It was found that the use of *M. oleifera* as a coagulant aid with alum shows fruitful results as an initial treatment.

Mataka et al.[164] investigated the ability of *M. oleifera* and *M. stenopetala* to remove cadmium. The removal capacity of *M. stenopetala* was found to be more than that of *M. oleifera* for the removal of cadmium from water. Araujo et al.[165] described the sorption capacity of *M. oleifera* seeds for the removal of Ag(I) from water. Further, the applicability of *M. oleifera* was investigated for removing various metal ions Cd(II), Pb(II),

Table 1. Treatment of Water Using *M. oleifera* as Coagulant

water sample	coagulant dose	removal (%)/residual turbidity	refs
raw water from Hafir of El Qerabin	200 mg/L of extra fine seed powder	98%	12
	fine seed powder	97%	
kaolin suspension in tap water (105 NTU)	SMOSP — 50 mg/L	80–90%	35
	NSMOSP — 500 mg/L		
synthetic water, surface water, and groundwater from two tube wells (300–900 mg/L as $CaCO_3$)	MOC-DW	softened water (zero residual hardness)	33
	950 mg/L for synthetic water		
	1800 mg/L for other water samples		
kaolin suspension+tap water (50–750 NTU)	MOC-DW	92–99%	135
	50–100 mg/L		
kaolin suspension (105 NTU)	SMOSP—1 mL/L	90%	10
	NSMOSP—10 mL/L		
	purified protein—(0.5–1 mg/L)		
kaolin suspension (50 NTU)	MOC-SC—4 mL/L	>95%	90
	MOC-DW—32 mL/L	78%	
5–50 mg of kaolin/L	MOC-SC-PC	residual less than 0.5 mg kaolin/L	77
river water–wet season (400 NTU)	100 mg/L	90%	39
Sungai Semenyih WTP			58
(a) initial turbidity	SOECD-200 mg/L	98%	
451 NTU	SBCD-250 mg/L	96.9%	
(b) initial turbidity	SOECD-250 mg/L	87%	
56 NTU	SBCD-250 mg/L	80.7%	
Sungai Selangor WTP			
(a) initial turbidity	SOECD-250 mg/L	97.9%	
321 NTU	SBCD-250 mg/L	96.9%	
(b) initial turbidity	SOECD-250 mg/L	90.4%	
65.8 NTU	SBCD-250 mg/L	91%	
surface water from a stream	SOECD		72
<50–100 NTU	20–30 mg/L	96%	
>100 NTU	50–80 mg/L	96%	
turbid surface water	*M. oleifera* water extract		23
turbidity	100–200 mg/L	0.3–1.5 NTU	
15–25 NTU			
heterotrophic bacteria—280–500 cfu/mL		5–0 cfu/mL	
fecal coliforms—280–500 MPN 100/mL		5–10 MPN 100/mL	
Maiduguri raw water	ESC-180 mg/L	82.35%	140
	USC-300 mg/L	76.47%	
drinking water	*M. oleifera* water extract	residual turbidity 1.85–1.3 NTU	43
15–50 NTU	33 mg/L		
shallow well water from five sources			4
Kumponda-49 NTU	MOC-DW		
Mtembo-24 NTU	250 mg/L	100%	
Nlukla-7 NTU, 219 NTU	250 mg/L	99%	
Chelewani-3 NTU, 39	100 mg/L, 250 mg/L	75%, 100%	
NTU	100 mg/L, 250 mg/L	93%, 72–93%	
Kumazale-1 NTU	50 mg/L	100%	
		also no. of fecal coliforms reduced by 80% but reduction not met by WHO guidelines, that is, 0 cfu/100 mL	
raw water (129 NTU)	dried *M. oleifera* seed powder alum to *M. oleifera* dose		128
	60:40 mg/L	<5 NTU	
	80:20 mg/L	<5 NTU	
water samples from shallow wells	*M. oleifera* powder, 1 g/L		62
total coliform per 100 mL = 9		total coliforms—50–100% improvement	
E. coli per 100 mL = 9		*E. coli*—100% improvement	
kaolin suspension in drinking water	MOC-SC		124
50 NTU	20 mg/L	91%	
500 NTU	10 mg/L	98.7%	
1000 NTU	10 mg/L	99.4%	
turbid colored water	MOC-DW	residual filtrate	129
47–48 NTU	500 mg/L	turbidity—4 NTU	
43–46 TCU		color—15 TCU	
drinking water (river)	MOC-SC		136

Table 1. continued

water sample	coagulant dose	removal (%)/residual turbidity	refs
50 NTU	150 mg/L (50–150 NTU)	83.2%	
450 NTU	125 mg/L (>150 NTU)	99.8%	
		also *E. coli* is almost fully removed	
synthetic water (surface water+bottom sediments diluted with lab grade sediments)	dried shelled *M. oleifera* seed powder		63
10 NTU	100 mg/L	14.2%	
	200 mg/L	22.8%	
300 NTU	100 mg/L	95.9%	
	200 mg/L	96.2%	
Bilaoli lake water		≤1 NTU in both cases	16
15 NTU	MO-DW–30 mg/L		
25NTU	MO-DW–60 mg/L	also bacteria removal efficiency is better than alum	
kaolin suspension	MOC-DW		75
(a) 146 NTU	25–75 mg/L	84%	
E. coli at 10^4 per 100 mL		88%	
(b) <5 NTU		82%	
605 cfu/100 mL		82%	
(c) 45 NTU		76%	
2650 cfu/100 mL		93%	
(d) 160 NTU		97%	
E. coli at 10^4 cfu/100 mL		66%	
(e) color		88%	
deionized water with kaolin stock suspension	SMOS		64
	30–55 mg/L	50–90%	
40–200 NTU			
river water	MOC-DiW	of order 95% or greater	125
88–195 NTU	50–200 mg/L		
Nigeria pond water		after 24 h	141
<50 NTU		96.34%	
50–150 NTU		82.5%	
raw water from Pirapo River-Maringa city, Parana state (450 NTU)	100–300 mg/L		142
	MOC–1 M NaCl extracted	99.8%	
	MOC–0.1 M NaCl extracted	55–97%	
	MOC–0.01 M NaCl extracted	50%	
	MOC–water extracted	52%	
surface water from River Mues in Rotterdam (20–45 NTU)	7 mg of DOC/L (purified)	97%	137
	58 mg of DOC/L (crude)		
canal water in Delft, The Netherlands (24–60 NTU)			

Co(II), Cu(II), and Ag(I) from aqueous solutions. The evaluation of morphological characteristics and chemical composition of *M. oleifera* seeds was done using FTIR spectroscopy, thermogravimitric analysis (TGA), XRD, and SEM.[166]

Acheampong et al.[167] evaluated the performance of coconut shell and *M. oleifera* seeds as biosorbents based on kinetic models for Cu(II) removal. It was found that interparticle diffusion was the rate-limiting step, which controlled the sorption process. However, at the initial stage, film diffusion could not be ignored.

Vardhan and Karthikeyan[168] employed physicochemical processes of adsorption and coagulation to investigate the potential of low-cost materials like rice husk, *M. oleifera*, and chemicals like manganese sulfate and manganese chloride for removal of fluoride from water. They obtained comparable results with different chemicals and cost-effective agricultural materials to remove fluoride from water.

Obuseng et al.[169] investigated the uptake of different metal ions such as lead, copper, cadmium, nickel, and manganese from the single and multimetal solutions using MOSB and ABL. The work revealed that MOSB performed well in multisolute systems and followed the decreasing orders as Pb(II) > Cu(II) > Cd(II) > Ni(II) > Mn(II) and Zn(II) > Cu(II) > Ni(II), suggesting the presence of sufficient active binding sites for all metal ions studied, whereas ABL showed the reverse trend as compared to that of MOSB.

Recent studies on *M. oleifera* for treatment of water with heavy metal ion concentrations in tabular form have been presented in Table 2.

3.2.2. Removal of Dyes. Textile effluent contains large volumes of hazardous substances, 60–70% of which are azo compounds.[171] These substances damage aquatic and vegetable life when released into the environment. Several conventional and traditional methods have been applied for treating dye containing effluent, but these methods are either expensive or cannot cope with the high concentration of contaminants. On the other hand, advanced technologies may be economically unacceptable. Therefore, in the past few years, the use of natural biological adsorbents (biomaterials) from agricultural materials has been focused on as an alternative and cost-effective technology to remove pollutants. In this sense, researchers have been searching the use of *M. oleifera* as an interesting natural coagulant for the removal of dyes.

Beltran Heredia's group researched the ability of *M. oleifera* seed extract for removing several dyes.[172−174] Figure 6

Table 2. Optimal Metal Ion Reductions Using *M. oleifera* Seeds

metal ion	metal ion concentration	general preparation procedure	coagulant/sorbent dose	removal rate/ efficiency	conditions	refs
As(III)	25 mg/L	DPP or SMOSP	2.0 g/200 mL	60.21%	pH 7.5, contact time 60 min	143
As(V)	25 mg/L	-do-	2.0 g/200 mL	85.6%	pH 2.5, contact time 60 min	
Pb	7 mg/L	*M. oleifera* seed kernals and ram press cakes	120 mg/L	70−89%		155
Fe	7 mg/L		120 mg/L	66−92%		
Cd	7 mg/L		120 mg/L	44−47%		
Pb	7 mg/L	dried defatted *M. stenopetala* and *M. oleifera* seed powder	2.5 g/100 mL	98%	pH 10	157
Cd(II)	4 mg/L	shelled *M. stenopetala* and *M. oleifera* water and NaCl seed extract	1 mL of sorbent in 9.50 mL of metal solution	100%	pH 7.8	156
Zn(II)	4 mg/L			100%	pH 4.0	
Cu(II)	4 mg/L			60%	pH 6.0−8.0	
Cr(III)	4 mg/L			100%	pH 4.0	
Cd(II)	25 μg/mL	DPP or SMOSP	4 g/200 mL	85.10%	pH 6.5, contact time 40 min	158
ternary mixture of [Cd(II), Cr(III), and Ni(II)]	1−100 mg/L	DPP or SMOSP	4 g/200 mL	Cd(II)-76.59%		151
				Cr(III)-68.85%	pH 6.5	
				Ni(II)- 60.52%	pH 7.5 for Ni(II)	
Zn(II)	50 mg/L	DDP or SMOSP	0.5 g/L	74.76% with treated *M. oleifera* biomass	pH 7, contact time 50 min	159
Cr(III)	25 mg/L	SMOSP	4.0 g	81.02%	pH 6.5, contact time 40 min	152
Cr(VI)	50 mg/L	SMOSP	4.0 g	88.15%	pH 2.5	
Ag(I)	25 mg/L	SMOSP	2 g/100 mL	23.13 mg of Ag(I)/g of *M. oleifera* seeds	pH 6.5, extraction time 20 min	165
Cd(II)	7 mg/L	*M. oleifera* and *M. stenopetala* seed powder	2.5 g/100 mL	*M. oleifera*-70.7%	pH 5, stirring time 2 h	164
				M. stenopetala-82.7%		
Ni(II)	25 mg/L	DPP or SMOSP	4.0 g/200 mL	75.64%	pH 6.5, contact time 40 min	154
Cu(II)	10 mg/L	DPP or SMOSP	2 g/100 mL	85%	pH 7.0, contact time 30 min	167
fluoride	5 mg/L	*M. oleifera* seed extract	1 g/L	89%	pH 6.0	168
Ni(II)	4 mg/L	DPP treated with 0.1 mol/L NaOH	2 g/50 mL	90%	pH 4.0−6.0, agitation time 5 min	170

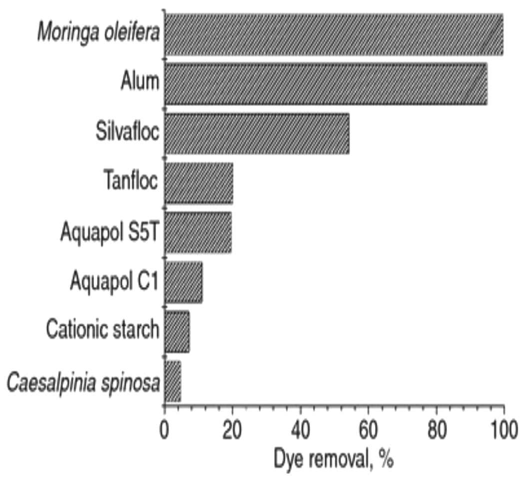

Figure 6. Preliminary screening on dye removal at 25 °C and pH 7. Reprinted with permission from ref 172. Copyright 2008 John Wiley and Sons.

demonstrates the ability of *M. oleifera* for the removal of anionic dyes.[172] Later, they studied the influence of various parameters, pH, temperature, and initial concentration of dye on the coagulation/flocculation process for the removal of anthraquinonic, indigoid, and azoic dyes using RSM.[175]

Marandi and Sepehr[176] investigated the removal of Orange 7 dye from wastewater using *M. oleifera* seeds as a natural adsorbent and concluded that shelled *M. oleifera* seeds could be successfully used as a viable, cost-effective, and pretreatment approach in advanced technologies for wastewater pretreatment. Srivastava et al.[177] treated water containing six different concentrations of Methylene Blue and Methylene Green to study the use of *M. oleifera* seeds as a sorbent in the treatment of dye contaminated water for the reduction of basic dyes concentration in effluent water. It was assessed that *M. oleifera* seeds could be applied for the industrial wastewater treatment under optimum conditions in a cost-effective manner, replacing the existent technology involving chemical-based procedures. Recently, Kumar et al.[178] investigated the biosorption characteristics of SMOS for the removal of Methylene Blue and Congo Red dyes from aqueous system and reported the applicability of a single layer ANN model for the simulation of the process and for the prediction of removal efficiency of SMOS for removing dyes. Studies on the removal of dyes with *M. oleifera* have been summarized in Table 3.

3.2.3. Removal of Surfactants. Surfactants are considered as one of the most dangerous and noxious contaminants of all emerging water pollutants. Their magnitude of contamination being high, they may prove fatal for aqueous flora and fauna. The development of methods for the removal of surfactants that are cheaper and easy to apply is still a challenge and has

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Table 3. Removal of Dyes under Standard Conditions

dye	initial dye concentration (mg/L)	general preparation procedure	*M. oleifera* dose	optimum pH	% removal	refs
Chicago Sky Blue 6B (CSB)	100	MOC-SC	125.7 mg/L	7	up to 99%	172
Alizarin Violet 3R	100	MOC-SC	62.86 mg/L	7	95%	173
Carmine Indigo	100	MOC-SC	315 mg/L	7	80%	174
Orange 7 dye	20	SMOSP	0.4 g	6	60% within first 5 min	176
Congo Red		SSP	25 mg/L	4	98%	179
Methylene Blue (MB)	25	SMOSP	1−4 g	6.5	90.27%	178
Congo Red	25	SMOSP	1−4 g	2.5	98.52%	

attracted the interest of various researchers. The ability of *M. oleifera* for the remediation of different anionic surfactants from wastewater has also been investigated, and interesting results have been reported.

Beltrán-Heredia and Sánchez-Martín[180] evaluated the potential of *M. oleifera* as an anionic surfactant removal agent in aqueous solutions. 80% removal of sodium lauryl sulfate was observed, indicating that it works well for the surfactant elimination. It was also observed that the efficiency of the process decreased with increase in pH. It was explained to be due to the cationic nature of *M. oleifera* extracted protein. On the other hand, the effect of temperature was found to be non significant for the removal of surfactant.

Thereafter, Sánchez-Martín and Beltrán-Heredia[181] investigated the ability of *M. oleifera* seed extract for the removal of sodium lauryl sulfate from polluted surface water. Jar tests revealed the high efficacy of *M. oleifera* for surfactant removal. Surfactant was removed rapidly with relatively lower dosages of coagulant, and the efficiency of the process increased with increase in initial surfactant concentration. The coagulation−flocculation process was modeled with different adsorption models, and the Gu−Zhu model was found to present the best fit (Figure 7).

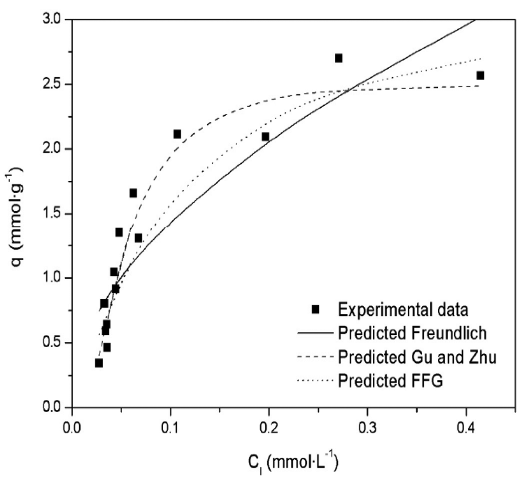

Figure 7. General equilibrium and adjustment data. Reprinted with permission from ref 181. Copyright 2010 IWA Publishing.

Further, Beltrán-Heredia et al.[182] selected Polyoxyethelene (3.5) SLES, a long-chain anionic detergent as a model compound to assess the performance of *M. oleifera* in removing surfactants from industrial effluents. Wastewater from laundry industry was selected for this purpose. The influence of various process parameters on the coagulation process was investigated. The statistical design of the experiments then was performed to identify optimum combinations of different process parameters. Thereafter, the coagulation phenomenon was explained by theoretical models prior to pilot plant implementation.

3.2.4. Industrial Effluent Treatment. *M. oleifera* has been reported as an eco-friendly and inexpensive alternative solution for the treatment of industrial effluents too by various researchers.

Bhuptawat et al.[68] incorporated MOC-DW in a sequential wastewater treatment process. It was seen that the overall percentage of COD removal increased from 50% to 58% and 64% with 50 and 100 mg/L dosages of *M. oleifera*, respectively, when it was used along with alum (10 mg/L) rather than using it alone. Prasad[183] discussed the scope of introduction of MOC in a sequential treatment of distillery spent wash for its decolorization. RSM was used to optimize different process parameters of color removal, dosage, pH, and salt concentrations. Maximum color removal with NaCl (53%) and KCl (64%) salts, respectively, with optimum dose of 20 and 60 mL and a concentration of 0.25 M was achieved at pH 7 and 8.5. Vieira et al.[184] investigated the sorption properties of *M. oleifera* seeds for the removal of organic components of DIW and demonstrated the ability of this natural material in the preconcentration of organic pollutants in DIW samples. Maximum efficiency in terms of color and turbidity removal was achieved in the pH range of 5−8 with 0.2 g of *M. oleifera* and 0.2 L of 1.0 g/L sorbate solution (DIW). Menkiti et al.[185] investigated the application of MOC in the treatment of highly turbid coal washery effluent. RSM was used to optimize different process parameters and to demonstrate their interactions with one another. Babatunde et al.[186] incorporated *M. oleifera* seeds as a coagulant in a sequence of wastewater treatment processes in their studies conducted at pilot scale. *M. oleifera* was found to be most efficient for the removal of coliforms (98%) and total bacterial count (84%) in comparison to sand filtration and alum for the treatment of pond effluents. Considering its economic benefits and effectiveness, they encouraged its use as a central component in the water treatment procedures at domestic level as well as at industrial scale. Bhatia et al.[187,188] used this well-recognized and eco-friendly natural coagulant for the pretreatment of POME in an attempt to obtain the maximum water recovery with the lowest content of suspended particles and organic components in it. They used different biodegradable flocculants in a series of laboratory and pilot plant studies. Initially, KP 9650 flocculant was used. The comparison of different effluent characteristics was carried out before and after the treatment. A substantial reduction in COD, BOD, and suspended solids concentration showed the effectiveness of this process. The additional advantage of the procedure was that water recovered after treatment could be recycled and reused in palm oil mills for extracting palm oil. Moreover, the disposal of sludge did not

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require any sort of further treatment. In proceeding years, Bhatia et al. combined MOAE with flocculant (NALCO 7751) and evaluated the improvement in removal of suspended particles and reduction in COD. The best performance was observed at a temperature of 30 °C. Thereafter, estimation of sludge volume index and percentage recovery of sludge and water was also done. The extraction of edible oil commercially known as Ben oil and use of sludge as an organic fertilizer further made this procedure highly economical.[188,189]

Figure 8 shows the comparative performance of MOAE and alum for the removal of suspended solids. To obtain the optimum values of parameters, affecting the treatment process for palm oil mill effluent, Bhatia et al.[190] applied RSM involving experimental design to find the effect of pH, settling time, MOAE dosage, and flocculant dosage. The results showed 87% recovery of sludge at pH 5.

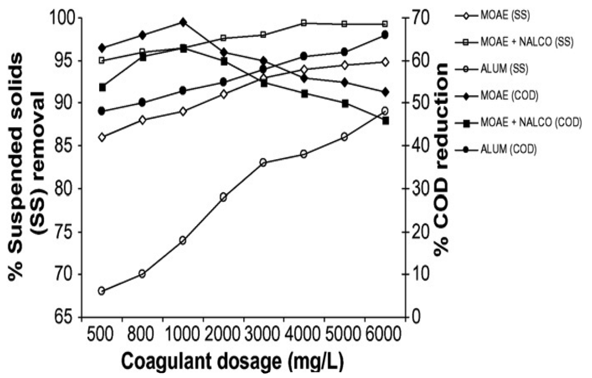

Figure 8. Effect of *M. oleifera* and alum dosage on the removal of suspended solids and COD reduction in POME pretreatment. Experimental conditions: pH 5, flocculant (NALCO 7751) dosage = 7000 mg/L, 90 min of sedimentation time, and 30 °C temperature. Reprinted with permission from ref 188. Copyright 2007 Elsevier.

4. SLUDGE DEWATERING USING *M. OLEIFERA* AS A CONDITIONER

Drinking water[191,192] and wastewater treatment methods generate large volumes of sludge containing over 90−99.9% water. Sludge is a settled suspension of small solid particles in a liquid/water, which are very difficult to separate from liquid phase. Direct disposal of untreated sludge to the environment is believed to adversely affect the recipient as a result of solids deposition and chemical composition of sludge.[193] Therefore, the most important thing in sludge management is to reduce the sludge volume by water separation before it is disposed off. It will reduce the costs associated with transportation, handling, and waste disposal and the environmental impacts.

Sludge dewatering has been pointed out as one of the most expensive and least understood process. Organic compounds as well as colloidal particles make the sludge dewatering difficult. Sludge dewatering can be highly improved by its conditioning.[194] Conditioners facilitate the agglomeration of small sludge particles into bigger particles prior to their separation from liquid phase, thus making the separation easier. Widely used chemicals include aluminum sulfate, lime, ferric chloride, and synthetic organic polyelectrolytes. Being expensive, these chemicals may pose an unfavorable impact on sludge handling and the ecosystem. Conversely, the high cost and environmental impact of the commonly used chemical conditioners may be counteracted, if locally available alternative materials are used. In the search of cheaper materials, *M. oleifera* has been studied as one of the most effective conditioners for the treatment of sludge. Sludge produced by *M. oleifera* is 4−5 times less than other chemical conditioners. Moreover, when *M. oleifera* is used, biodegradable byproducts are generated, favoring its use as a biosolid for agricultural and land applications seeing that it does not contain heavy metals.[35]

Ademiluyi[195] investigated the utility of *M. oleifera* for sewage sludge conditioning. The study showed that powdered seeds of *M. oleifera* improved sludge filterability. It was comparable with the traditional ferric chloride conditioner with respect to cake solids concentration. The conditioning potential of oil-free seed of *M. oleifera* was found to be more than ordinary *M. oleifera* seed.[196] Ghebrimichael and Hultman[120] compared *M. oleifera*, alum, and synthetic polyelectrolytes for conditioning the sludge from the drinking water treatment plant, and it was reported that MOCP showed sludge conditioning capabilities similar to those of alum. *M. oleifera* and alum formed relatively stronger flocs than other polyelectrolytes. It was concluded from the results that *M. oleifera* could be used effectively either alone or linked with other polyelectrolytes as an alternative material for dewatering of sludge. Improvement in the dewatering characteristics was assessed by reductions of CST and SRF values. Figures 9 and 10 show the results of SRF and CST reduction, respectively, for alum, *M. oleifera*, and polyelectrolyte conditioned sludge.

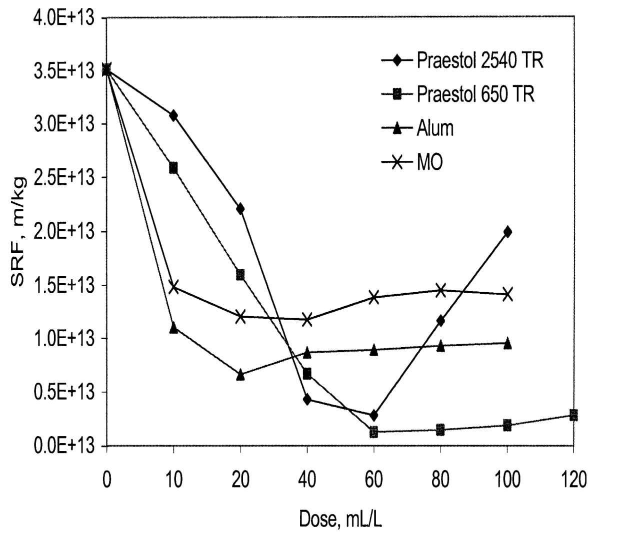

Figure 9. SRF values at different doses of alum, MO, and polyelectrolytes. Reprinted with permission from ref 120. Copyright 2004 Springer.

Tat et al.[197] evaluated the effect of *M. oleifera* in sewage sludge conditioning by employing *M. oleifera* seeds in the form of dry powder, distilled water extract, and salt extract. The distilled water extracted form of *M. oleifera* was found to be most effective, and the effectiveness of its dried form was found to be comparable to that of the chemical polymer, Zetag 7653. Later, Tat et al.[198] optimized significant factors, that is, mixing speed, mixing duration, and dosage of *M. oleifera* using the design of experiment technique.

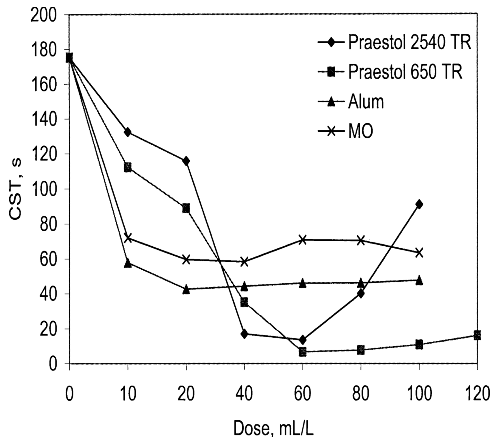

Figure 10. CST values at different doses of alum, MO, and polyelectrolytes. Reprinted with permission from ref 120. Copyright 2004 Springer.

5. EFFECT OF STORAGE/PRESERVATION CONDITIONS OF *M. OLEIFERA* ON ITS COAGULATION EFFICIENCY

Like all other biological materials, *M. oleifera* being a biomaterial, its efficacy depends upon the environmental conditions such as humidity, pH, temperature, etc. The active agents of seeds are degraded when exposed to the environment. Therefore, it is necessary to study the effect of storage duration and storage conditions on the coagulation efficiency of *M. oleifera* seeds. Controlling the seed moisture and temperature has been found to be very essential for the proper preservation of seeds to increase their shelf life, which in turn assures the minimum degradation of active ingredients and promises the completion of coagulation mechanism for clarifying turbid water. Also, the market acceptability of *M. oleifera* with longer shelf life is enhanced.[199]

Katayon et al.[132] investigated the effects of storage duration and temperature on the coagulation efficiency of *M. oleifera*. Its stock solutions were kept for 7 days at two different temperatures, that is, at room temperature (28 °C) and at refrigerator temperature (3 °C). Their study revealed that *M. oleifera* stock solutions, which were kept at 28 °C, were effective for 3 days for water samples with medium, high, and very high turbidity levels, whereas the stock solutions kept at 3 °C showed effective coagulation for up to 5 days for the samples with the same range of turbidity. The efficiency decreased with increase in storage duration. The percentage of turbidity removal was found to be more for high initial turbidity with optimum dosage.

Further, Katayon et al.[200] selected open and closed containers, room temperature and refrigeration temperature, and storage periods of 3 and 5 months as variables in their study while investigating the effect of storage conditions on the coagulation efficiency of *M. oleifera* seeds. They recorded the better performance of *M. oleifera* solutions, which were kept in the refrigerator and at room temperature for 1 month, than those that were kept for 3 and 5 months in both types of containers. The reason for the same was not discussed, but it was assumed to be due to the microbial degradation of proteins.

In light of the above observations, Katayon et al.[199] further extended their work to explore more. *M. oleifera* seed powders were kept for 12 months under different storage temperature conditions, and the effects of storage temperature, packaging methods, and freeze-drying technique were investigated on preservation of *M. oleifera*. It was observed that in the refrigerator, the coagulation efficiency of non freeze-dried seeds was preserved, but at room temperature it deteriorated rapidly. Similarly, an open container was not found to be that appropriate when compared to a closed container and vacuum packing. On the other hand, the coagulation efficiency of freeze-dried *M. oleifera* seeds was retained irrespective of the storage temperature and packaging method for up to 11 months.

Yet in a recent study, Golestanbagh et al.[201] did not find any significant difference in the coagulation efficiency of SMOS for turbidity removal stored in open and closed containers at room temperature for different durations, which varied from fresh seeds taken from the tree to storage for 2, 4, 6, and 8 weeks.

6. COST OF *M. OLEIFERA* SEEDS AS COAGULANT

Water purification using *M. oleifera* is a method that can be considered as a good, sustainable, and cheap solution, if the supply of the *M. oleifera* seeds is guaranteed because the amount of seeds required for production of *M. oleifera* extract is quite large.

According to Jahn,[202] an uncultivated *M. oleifera* tree is able to produce approximately 2000 seeds per annum, and nearly 6000 L of water can be treated with these seeds with a dose of 50 mg/L. However, if cultivated fully, the yield of seeds can be increased by 4−5 times, that is, 10 000−20 000 seeds, that would be sufficient for treating 60 000 L of water per annum. As a rough estimate, a single tree cultivated in a rural village is capable of providing enough seeds that will be sufficient to use for the whole year for four families (assuming 20 L consumption per person per day).

According to the cost analysis carried out by Sutherland et al.,[32] in 1993, the purchase price of *M. oleifera* seeds was MK 75 per 1000 m^3 water treated (£1 = MK 10.07 in March 1993), whereas the cost of alum and soda ash was MK 501 per 1000 m^3 water treated. According to WELL,[203] assuming that a mature tree yields an average of 3 kg seed kernel, the harvest of a single tree will be able to treat 30 000 L of water with a 100 mg/L dose. As per these assumptions, keeping the tree spacing near 3 m, mature trees harvested from about 1 ha (nearly 3000 kg) would be able to treat 30 000 m^3 water.

Sutherland et al.[32] stated that the cost benefits of *M. oleifera* in African countries are employed to the net zero cost of its press cake in those countries. It is obtained easily as a byproduct of oil extraction. However, the situation is not the same in all places. In countries like Malaysia, the cultivation cost of 1 kg of *M. oleifera* was estimated to be approximately U.S. $2, which was double the cost of alum. Similarly, in Botswana, the cost of 1 kg of seeds has been reported to be approximately U.S. $27. Yarahmadi et al.[124] estimated that in south of Iran, the cultivation cost of every kilogram of *M. oleifera* seed was U.S. $2, which was higher in comparison to the production cost of alum and polyaluminium chloride.

According to Arnoldsson et al.,[43] the area required for production of *M. oleifera* seed is not entirely unrealistic particularly for small- to medium-sized water supplies located in rural areas. Further, there may be significant reductions in the transportation costs of imported chemicals if aluminum sulfate is replaced by *M. oleifera*. Yet additional investments will be required for storage, grinding, and mixing of seeds. Therefore, it was suggested to prepare the *M. oleifera* extract onsite to minimize transport and storage costs. Also, because extract can be stored for 1 day without losing coagulation properties, batch processes designed for the demand of one full production day may be more suitable than a continuous process.

7. FUTURE PERSPECTIVES OF *M. OLEIFERA*

The development of any "new product" in the scope of alternative products hosts many interlinked constraints and limitations. *M. oleifera* too mediates some issues, such as the problem of residual DOC associated with the use of its crude extract (MOC-DW and MOC-SC) as coagulant may facilitate bacterial growth in the purified water. The seeds also tend to increase the organic nitrate and phosphate content of treated water. Although the use of purified protein (coagulant) overcomes these drawbacks of crude extract, it will increase the processing cost of water treatment technology and thus hinder the usage of this plant-based coagulant at the industrial level. Currently, the application of *M. oleifera* is restricted to small-scale usage and academic research. Not even a single effort has been made to commercialize its application for water/wastewater treatment. The reasons behind this are shortage of R&D data, market awareness, and regulatory approval. In the light of market awareness, its acceptance may further be enhanced by fervent promotion and endorsements from real stake holders particularly from the regulatory authorities for commercialization of plant on a wide scale. Another huge shortcoming is the supply of its seeds. If the *M. oleifera* seeds are not available in abundance, its application will remain limited to lower markets only. Remedial measure to this problem could be the abundant growth of this tree in the tropics similar to coffee and tea plants, which are cultivated intensively only in tropical regions but are consumed worldwide. This is the only way by which producing countries may obtain the maximum economical benefits of this product. Apart from growing this plant intensively, proper management with regard to the pruning and pollarding of cultivated plant is of utmost importance as excessive growth of this plant is undesired.

Despite these constraints associated with their application on a large scale, the significant benefits such as safety issues and eco-friendly nature of *M. oleifera* seeds address the bright future of this plant in water purification and treatment processes. Its application is highly recommended for domestic water purification in underdeveloped countries where people are compelled to consume polluted water. Although it does not ensure 100% purification of water, it might reduce the risk of waterborne diseases significantly.

8. CONCLUSION

M. oleifera is a unique plant that demands less and offers much. It holds a unique and consistent position in international market due to its easy adaptation to various ecosystems and farm systems. A critical evaluation of *M. oleifera* usage as a sustainable material for water and wastewater treatment in this Review reveals that it can be successfully used for the removal of turbidity, metal ions, organic, and biological species from water. This biomaterial is capable of removing pollutants even at lower doses, which makes its application economical. As its sludge is not hazardous, there will be no need to develop any eco-friendly waste management method. Further, the chemistry of water too remains unaltered with respect to its pH, alkalinity, and ionic strength. These characteristics render this plant a potential candidate for quick and inexpensive water treatment technology. Replacement of chemical coagulants by *M. oleifera* may bring about significant reductions in the transportation cost of imported chemicals as well as the health risk associated with their usage.

The coagulation effectiveness of this plant varies depending upon the initial turbidity of the water to be treated. Its coagulation efficacy has been observed to be similar to other chemical coagulants for high turbidity waters; however, its effectiveness is reduced for low turbidity waters. Apart from its uses as such, *M. oleifera* seed extract has also been assessed as a primary coagulant, cocoagulant, or secondary coagulant aid (flocculant) for the purification of drinking water and treatment of wastewater. In our opinion, scientists must explore the synergy of this process with other existing water treatment technologies to further reduce the problems associated with the usage of this plant and find a viable solution for its commercialization. Yet it needs the collaborative efforts of farmers/suppliers of seeds, R&D institutions, and industry to fulfill this dream of cost-effective and feasible water/wastewater treatment technology.

AUTHOR INFORMATION

Corresponding Author

*Phone: +91-172-2534920. Fax: +91-172-2779173. E-mail: sushilkk1@yahoo.co.in, sushilkk1@pu.ac.in.

Notes

The authors declare no competing financial interest.

Biographies

Dr. Sushil Kumar Kansal is an academician and researcher of international recognition at the Dr. S. S. Bhatnagar University Institute of Chemical Engineering & Technology, Panjab University, Chandigarh, India. He obtained his B.Tech., M.E., and Ph.D. degrees in 1998, 2001, and 2007. His research areas are environmental science and engineering, biomaterials, advanced oxidation processes, nanotechnology, air pollution, mass transfer, water pollution, and material science. He has published more than 30 research papers in international and national journals of repute. One of the research publications that was published in 2007 has more than 220 citations. He is on the editorial boards of 5 scientific journals. Dr. Kansal has visited many countries, including China as a visiting scientist, Germany under the DAAD fellowship, Canada under the Shastri Indo-Canada faculty mobility program, and was also nominated by UGC, Government of India, to visit Mauritius. Dr. Kansal is also a life member of six international scientific societies and has international collaborations in four countries. He is a reviewer of more than 30 international journals.

Amit Kumari is a postgraduate student at Dr. S. S. Bhatnagar University Institute of Chemical Engineering & Technology, Panjab University, Chandigarh, India. She has been doing research in Dr. S. K. Kansal's group for the last 3 years and is working exhaustively on the exploration of *M. oleifera* for the treatment of water and wastewater. She has presented one paper at an international conference at Panjab University. She has availed scholarships at Higher and Senior Secondary level on a merit basis.

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ABBREVIATIONS

ABL	amine-based ligand
ANN	artificial neural network
COD	chemical oxygen demand
CST	capillary suction time
DIW	dairy industry wastewater
DOC	dissolved organic carbon
DPP	dried and pulverized seed powder
ESC	extracted seed coagulant
MO	*M. oleifera*
MOAE	*M. oleifera* seeds after oil extraction
MOC	*M. oleifera* coagulant
MOC-DW	*M. oleifera* coagulant extracted using distilled water
MOC-DiW	*M. oleifera* coagulant extracted using deionized water
MOCP	*M. oleifera* coagulant protein
MOC-SC-PC	*M. oleifera* coagulant-1 M NaCl extracted-purified coagulant
MOC-SC	*M. oleifera* coagulant-1 M NaCl extracted
MOSB	*M. oleifera* seed biomass
NSMOS	nonshelled *M. oleifera* seed
NSMOSP	nonshelled *M. oleifera* seed powder
POME	palm oil mill effluent
POU	point-of-use
RSM	response surface methodology
SBCD	shelled blended coagulant dose
SDP	suspended and dissolved particle
SLES	sodium lauryl ether sulfate
SMOS	shelled *M. oleifera* seed
SMOSP	shelled *M. oleifera* seed powder
SOECD	shelled oil extracted coagulant dose
SRF	specific resistance to filtration
SSP	shelled seed powder
USC	unextracted seed coagulant

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