EFFICIENCY OF MORINGA OLEIFERA AS A NATURAL COAGULATION AND FILTRATION AGENT FOR REMOVAL OF MINERALS FROM HARD WATER: AN IN-DEPTH ANALYSIS OF MOLECULAR MECHANISMS, ADSORPTION KINETICS, AND TECHNO-ECONOMIC FEASIBILITY

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Abstract: The increasing demand for drinking water and the environmental impacts associated with conventional chemical treatments have driven research into clean and sustainable technologies. Water hardness, caused primarily by calcium ions (Ca^{2+}) and magnesium (Mg^{2+}), poses a significant challenge in domestic and industrial contexts. This study evaluates, through an in-depth literature review and a detailed proposal of an experimental model, the efficiency of Moringa oleifera (MO) as a natural coagulant and filtration agent for the removal of mineral ions (Ca^{2+}, Mg^{2+}, Fe^{2+}/Fe^{3+}, Zn^{2+}) from hard water. The proposed methodology involves the development of a hybrid gravitational filtration system composed of activated carbon beds with controlled particle size and the application of coagulant based on OM seeds. The analysis of previous studies indicates that the cationic proteins present in OM seeds, with molecular weights between 6.5 and 48 kDa and isoelectric point above pH 10, they act effectively through adsorption mechanisms (Langmuir and Freundlich isotherms), neutralization of charge and precipitation, promoting agglutination and subsequent removal of dissolved particles and ions. The literature reports turbidity removal rates of more than 99%, pathogenic microorganisms (> log 6 for E. coli) and metals such as iron up to 100%. Kinetic studies demonstrate that the adsorption process follows pseudo-second-order models, indicating chemisorption as the predominant mechanism. Comparative economic analysis reveals that the costs of treatment with OM can be up to 50% lower than those of aluminum sulfate in rural contexts. Based on these data, it is projected that the proposed biotechnological system will achieve mineral removal efficiencies greater than 95%, consolidating

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itself as a low-cost, ecologically viable and high-performance alternative for hard water treatment, applicable at various scales and socioeconomic contexts.

Keywords: Moringa oleifera; water treatment; water hardness; natural coagulation; hybrid filtration; adsorption isotherms; pseudo-second-order kinetics; economic analysis; sustainable technologies.

INTRODUCTION

Contextualization and Relevance

Water quality is a fundamental pillar for public health, socioeconomic development, and environmental sustainability. According to the World Health Organization (WHO), approximately 2.2 billion people worldwide lack access to safely managed drinking water, and inadequate water quality is responsible for more than 500,000 deaths annually from diarrhea. One of the most recurrent problems related to water quality is its hardness, defined by the high concentration of multivalent cations, predominantly calcium ions (Ca^{2+}) and magnesium (Mg^{2+}), but also including iron (Fe^{2+}/Fe^{3+}), zinc (Zn^{2+}), manganese (Mn^{2+}) and other minerals (MUYIBI; EVISON, 1995).

Hard water entails a number of technical and economic problems. In domestic systems, it reduces the efficiency of soaps and detergents due to the formation of insoluble salts, increases energy consumption in heaters due to the formation of scale, and alters the organoleptic properties of water, affecting taste and consumer acceptance (FAHMI; NAJIB; PING, 2011). In industrial contexts, water hardness is even more critical: the formation of mineral deposits in boilers, heat exchangers, cooling towers, and pipelines can reduce energy efficiency by up to 30%, increase maintenance costs, and in extreme cases, cause catastrophic equipment failures (DE GISI et al., 2016).

Limitations of Conventional Methods

Conventional methods for water softening include chemical precipitation processes (using lime and soda), ion exchange with cationic resins, and membrane separation (such as reverse osmosis and nanofiltration). While effective, these technologies have significant limitations. Chemical precipitation requires large amounts of alkaline reagents (CaO, Na_2CO_3), generating calcium carbonate and magnesium hydroxide sludges that require proper disposal (BRATBY, 2016). Ion exchange, although efficient, requires periodic regeneration of the resin with concentrated solutions of sodium chloride (NaCl), generating saline effluents that are difficult to treat (HARLAND, 1994). Membrane technologies, in turn, have high capital and operating costs, high energy consumption, and fouling problems that reduce membrane life (BAKER, 2012).

In addition to high operating costs, these technologies raise significant environmental concerns. The use of chemical coagulants such as aluminum sulfate ($Al_2(SO_4)_3$), widely employed in water treatment plants, has been associated with risks of aluminum toxicity, especially in vulnerable populations (NORDMARK et al., 2016). Epidemiological studies suggest a possible correlation between chronic exposure to aluminum in drinking water and the development of neurodegenerative diseases such as Alzheimer’s, although this relationship remains subject to scientific debate (XIONG et al., 2018). In addition, the sludge generated in the chemical coagulation process contains high concentrations of aluminum, representing an environmental liability that is difficult to manage (DUAN; GREGORY, 2003).

Natural coagulants: a sustainable alternative

In this context, the need to develop alternatives that are simultaneously efficient, low-cost and environmentally sustainable emerges. One of the most promising solutions lies in the use of natural coagulants of plant origin. Several plant species have been investigated for their coagulant properties, including Jatropha curcas, Opuntia ficus-indica (cactus), Cicer arietinum (chickpea), Plantago ovata (psyllium), and, notably, Moringa oleifera Lam. (CHOY et al., 2014; YIN, 2010).

Moringa oleifera Lam., a fast-growing tree native to the Indian subcontinent but widely cultivated in tropical and subtropical regions of Africa, Asia, and Latin America, has been noted for its remarkable water treatment properties. All parts of the plant (leaves, seeds, pods, roots) have nutritional and medicinal value (GOPALAKRISHNAN; DORIYA; KUMAR, 2016), but the seeds contain the active ingredient for coagulation. OM seeds contain a set of low molecular weight cationic proteins (6.5 to 48 kDa) that act as an effective coagulating agent, capable of destabilizing colloidal particles and promoting their agglutination (NDABIGENGESERE; NARASIAH, 1998; TAIWO; ADEKUNLE; ODENIYI, 2020).

Objectives and Structure of the Study

The main objective of this study is to evaluate, based on a robust review of the scientific literature and the detailed formulation of an experimental design, the efficiency of Moringa oleifera as a central component of a hybrid coagulation and filtration system for the removal of minerals from hard water. Specific objectives include:

1. To characterize the molecular coagulation mechanisms of BM cationic proteins;
2. Analyze the adsorption kinetics and equilibrium isotherms (Langmuir, Freundlich) for the removal of hardness ions;
3. Evaluate the removal efficiency of different physicochemical and microbiological parameters;
4. Propose an optimized hybrid gravitational filtration system;
5. Perform a comparative cost analysis between OM and conventional chemical coagulants;
6. Discuss the feasibility of scale-up and application in rural and industrial contexts.

The investigation seeks to demonstrate that a biotechnological system, combining the coagulant action of OM with the adsorptive capacity of activated carbon, can achieve removal rates higher than 95%, consolidating itself as a sustainable and low-cost alternative to conventional chemical methods, with applicability both in decentralized rural contexts and in industrial processes.

LITERATURE REVIEW

Molecular Characterization of Moringa oleifera Coagulant Proteins

The coagulant properties of Moringa oleifera are attributed to a set of cationic proteins present in the seeds. GASSENSCHMIDT et al. (1995) performed the first isolation and detailed characterization of an OM flocculant protein, determining a molecular mass of approximately 6.5 kDa by SDS-PAGE and an isoelectric point above pH 10. The amino acid analysis revealed a composition rich in basic amino acids (arginine, lysine, histidine), giving the protein a high positive charge density, an essential characteristic for the destabilization of negatively charged colloidal particles.

Subsequent studies have shown that MO seed extract contains multiple protein fractions with coagulant activity. NORDMARK et al. (2016) used high-performance liquid chromatography (HPLC) to isolate eight distinct cationic protein fractions, with native molecular weights ranging from 12 to 48 kDa and reduced molecular weights (after disulfide bond breakage) from 7 to 30 kDa. Dynamic light scattering (DLS) characterization revealed hydrodynamic radii of 1.2 to 1.5 nm for the strongly cationic fractions. Circular dichroism analysis indicated that these proteins predominantly have an alpha-helix secondary structure, a conformation that favors interaction with charged surfaces (NORDMARK et al., 2016).

ARREOLA, CANEPA and BARAJAS (2016) investigated the influence of the extraction method on the molecular characterization of MO seed extract. When extracted in distilled water, the extract showed protein fractions with molecular weights between 12 and 14 kDa. However, when extracted in saline solution (NaCl 1.0 M), the active ingredient presented a molecular weight of approximately 3 kDa, suggesting that the ionic strength of the medium can affect protein conformation and aggregation (ARREOLA; CANEPA; BARAJAS, 2016). This observation has important practical implications for the optimization of the extraction process.

More recently, PATIL et al. (2022) performed a comparative characterization of OM leaf and seed proteins, demonstrating that seed proteins have greater emulsifying and coagulating activity, attributed to their unique amino acid composition and three-dimensional structure. JAIN et al. (2019) identified three main protein bands in MO seed meal under reducing conditions, corresponding to molecular weights of 29, 14.2 and 6.5 kDa, corroborating the molecular heterogeneity of the extract (JAIN et al., 2019).

Coagulation and Flocculation Mechanisms

The mechanism of action of OM proteins involves multiple physicochemical processes. The most widely accepted model proposes that cationic proteins act primarily through adsorption and charge neutralization. Colloidal particles suspended in water (clays, organic matter, bacteria) usually have a negative surface charge due to the ionization of functional groups (carboxylics, phenolics, phosphates) or the adsorption of negative ions (HUNTER, 2001). This negative charge creates an electrostatic barrier that prevents the particles from aggregating, keeping them stable in suspension.

When the cationic proteins of MO are added to water, they adsorb on the surfaces of the colloidal particles, neutralizing their negative charges. NORDMARK et al. (2016) have experimentally demonstrated that the mechanism of kaolin flocculation by MO cationic proteins is dominated by adsorption and charge neutralization, with evidence that the effective concentration range of the coagulant expands with increasing water hardness (NORDMARK et al., 2016). This suggests that the ions Ca^{2+} and Mg^{2+} can act as cationic bridges, facilitating the interaction between proteins and particles.

Once the charges are neutralized, Van der Waals forces and hydrophobic interactions promote the aggregation of the particles into larger and denser flakes, which can be easily removed by sedimentation or filtration (GREGORY; DUAN, 2001). XIONG et al. (2018) quantified this effect through the adhesion coefficient (α), which measures the probability of one particle colliding and adhering to another. In MO functionalized sand filters (f-sand), the α coefficient was 0.8, compared to only 0.01 for untreated sand, demonstrating an 80-fold increase in capture efficiency (XIONG et al., 2018).

In addition to charge neutralization, MUYIBI and EVISON (1995) proposed that OM also acts through chemical precipitation, converting soluble hardness ions into insoluble products (MUYIBI; EVISON, 1995). This mechanism is particularly relevant for the removal of Ca^{2+} and Mg^{2+}. Proteins can complexify these ions, forming precipitates that are subsequently removed by sedimentation. The hardness ion adsorption isotherm is close to the Langmuir type, indicating a monolayer adsorption at specific protein sites (MUYIBI; EVISON, 1995).

Adsorption Kinetics and Equilibrium Isotherms

Understanding adsorption kinetics and equilibrium isotherms is critical to the design and optimization of treatment systems. Several studies have investigated these aspects for OM.

Kinetic Models: Adsorption kinetics describes the rate at which the adsorbate (ions, particles) is removed from solution and transferred to the adsorbent (MO proteins, activated carbon). The most commonly applied models are the pseudo-first-order (Lagergren) and the pseudo-second-order (Ho and McKay).

The pseudo-first-order model assumes that the adsorption rate is proportional to the difference between the adsorption capacity at equilibrium (q_e) and the capacity at time t (q_t):

$$dq\_t/dt = k_1(q\_e - q\_t)$$

The pseudo-second-order model assumes that the adsorption rate is proportional to the square of the difference:

$$dq\_t/dt = k_2(q\_e - q\_t)^2$$

VARSANI et al. (2022) applied both kinetic models to the biocoagulation process with functionalized OM seed powder in municipal wastewater. The results showed that the pseudo-second-order model presented a better fit to the experimental data ($R^2 > 0.99$), indicating that the adsorption process is controlled by chemisorption, involving the sharing or transfer of electrons between the adsorbate and the adsorbent (VARSANI et al., 2022). RASHEED, HADI, and HAMZA (2023) confirmed this finding, demonstrating that the pseudo-first-order model fits well with the nature of colloidal particle removal with MO seed extract (MOSE) (RASHEED; HADI; HAMZA, 2023).

Adsorption Isotherms: Equilibrium isotherms describe the relationship between the amount of adsorbate adsorbed and its concentration in the solution at equilibrium at a constant temperature. The most commonly used isotherms are Langmuir and Freundlich.

The Langmuir isotherm assumes monolayer adsorption at homogeneous sites, with no interaction between the adsorbed molecules:

$$q\_e = (Q\_max \times K\_L \times C\_e) / (1 + K\_L \times C\_e)$$

Where Q_max is the maximum adsorption capacity in monolayer, K_L is the Langmuir constant related to the adsorption energy, and C_e is the equilibrium concentration.

The Freundlich isotherm assumes multilayer adsorption on heterogeneous surfaces:

$$q\_e = K\_F \times C\_e\hat{}(1/n)$$

Where K_F is Freundlich's constant indicating the adsorption capacity and n is a parameter related to the adsorption intensity.

ÇELEKLİ, AL-NUAIMI and BOZKURT (2019) studied the adsorption of the dye Red Reactive 120 in MO seeds by applying the Freundlich, Langmuir and Dubinin-Radushkevich isotherms to the equilibrium data. The Freundlich model presented the best fit, suggesting multilayer adsorption on a heterogeneous surface (ÇELEKLİ; AL-NUAIMI; BOZKURT, 2019). MATOUQ et al. (2015) investigated the removal of heavy metals (Pb, Cd, Cr) by MO pods, concluding that Freundlich and Temkin's models fit well to the Pb and Cd data, while Langmuir's model better described the adsorption of Cr (MATOUQ et al., 2015).

For the removal of hardness ions specifically, MUYIBI and EVISON (1995) reported that the adsorption isotherm is close to the Langmuir type, with the maximum adsorption capacity dependent on the OM dosage and the initial hardness of the water (MUYIBI; EVISON, 1995). REDDY et al. (2010) determined that the adsorption capacity of Pb^{2+} adsorption capacity (Q_m) calculated from the Langmuir isotherm was 34.6 Pb^{2+}/g at initial pH 5.0, demonstrating the high affinity of MO seeds for metal cations (REDDY et al., 2010).

Efficiency of Removal of Physicochemical Parameters

The scientific literature presents robust evidence of the efficiency of OM in the removal of several water quality parameters.

Turbidity: Turbidity removal is the most extensively studied parameter. DESTA and BOTE (2021) reported turbidity removal efficiencies of 98% in acidic wastewater and 99.5% in basic wastewater, using an optimal dosage of 0.4 g/500 mL (800 mg/L) of MO seed powder (DESTA; BOTE, 2021). KENEA, TSEGAYE, and LETA (2023) achieved 88.5% turbidity removal using a blend of MO and Aloe vera (KENEA; TSEGAYE; LETA, 2023). ARNOLDSSON et al. (2008) demonstrated, in a pilot water treatment plant, an average turbidity removal of up to 96% with dosages of 20-30 $\

mg/L for low turbidity waters (<50 NTU) and 50-80 mg/L for high turbidity waters (>100 NTU) (ARNOLDSSON et al., 2008).

Color: Color removal is equally efficient. DESTA and BOTE (2021) reported removals of 90.76% in acidic waters and 97.7% in basic waters (DESTA; BOTE, 2021). KENEA, TSEGAYE, and LETA (2023) achieved 87.1% color removal (KENEA; TSEGAYE; LETA, 2023).

Total Dissolved Solids (STD) and Electrical Conductivity: STD removal is particularly relevant for hard water softening. KENEA, TSEGAYE, and LETA (2023) reported a remarkable 92.1% STD removal utilizing the combined MO + Aloe vera system (KENEA; TSEGAYE; LETA, 2023). OGUNSHINA et al. (2019) documented significant reductions in electrical conductivity, from 54.4 $\mu\text{S}/\text{cm}$ to 53.4 $\mu\text{S}/\text{cm}$ (OGUNSHINA et al., 2019). MARZOUGUI et al. (2021) observed that the Egyptian variety of MO was the most effective in reducing electrical conductivity, with removal efficiency of up to 29.7% (MARZOUGUI et al., 2021).

Chemical Oxygen Demand (COD): COD removal indicates the ability to remove dissolved organic matter. DESTA and BOTE (2021) reported removals of 65.8% in acidic waters and 65.82% in basic waters (DESTA; BOTE, 2021). MARZOUGUI et al. (2021) achieved 83.3% COD removal with OM (MARZOUGUI et al., 2021).

Heavy Metals: MO demonstrates high efficiency in heavy metal removal. Ali et al. (2020) reported complete removal (100%) of iron (Fe), and removals exceeding 98% for copper (Cu) and cadmium (Cd) using MO seed cake residue (ALI; MUYIBI; SALLEH, 2020). Dandesa et al. (2023) observed Fe^{3+} reduction from 7 mg/L to 2.93 mg/L (58.1% removal) (DANDESA; GEBREYOHANNES; SIME, 2023).

pH and Alkalinity: A significant advantage of MO is that, unlike aluminum sulfate, it does not significantly alter the pH and alkalinity of the treated water. MUYIBI and EVISON (1995) demonstrated that the removal of hardness is independent of the pH of the raw water (MUYIBI; EVISON, 1995). ARNOLDSSON et al. (2008) confirmed that MO seed extract has no significant effect on the pH or alkalinity of water (ARNOLDSSON et al., 2008).

Removal of Microorganisms and Organic Compounds

OM also has antimicrobial and organic compound removal properties.

Antimicrobial Activity: MO has antimicrobial peptides that act in water disinfection. NTIBREY, BAYOR, and DZOMEKU (2020) reported the removal of 99.9999% (>log 6) of E. coli and Salmonella typhi (NTIBREY; BAYOR; DZOMEKU, 2020). EILERT, WOLTERS and NAHRSTEDT (1981) isolated the antibiotic principle from OM (EILERT; WOLTERS; NAHRSTEDT, 1981). SUAREZ et al. (2005) characterized the function and optimized a plant-derived antibacterial peptide (SUAREZ et al., 2005).

Organic Compounds: OM can remove precursors of disinfection byproducts (SPD). SANTOS et al. (2015) studied the minimization of trihalomethanes (THMs) by combining OM with activated charcoal (SANTOS et al., 2015). RICHARDSON and POSTIGO (2012) discuss NTS in drinking water (RICHARDSON; POSTIGO, 2012).

HYBRID SYSTEM PROPOSAL AND FEASIBILITY

Optimization of the Hybrid Filtration System

The proposal for a hybrid gravitational filtration system combines the natural coagulation of OM with the adsorption of activated carbon. AHMED (2019) evaluated filtration systems with charcoal, OM seed, and sand (AHMED, 2019). The combination optimizes the removal of particles (coagulation) and dissolved contaminants (adsorption).

Technical-Economic Feasibility Analysis

The economic viability of the OM is a crucial factor. YUSOFF et al. (2017) and DELFÍN-PORTELA et al. (2024) analyzed the economic viability of the OM bioflocculant (YUSOFF et al., 2017; DELFÍN-PORTELA et al., 2024).

Economic Advantages:

- Raw Material Cost: In growing regions, the cost of OM seed can be up to 50% lower than that of aluminum sulfate (SALAZAR GÁMEZ et al., 2015).
- Waiver of pH Correction: OM does not change the pH, eliminating the cost of adding lime or soda, necessary with aluminum sulfate (ARNOLDSSON et al., 2008).
- Sludge Reduction: The sludge generated by OM is biodegradable and non-toxic, reducing disposal costs (GRABOW et al., 1985).

Challenges:

- Dosage: The dosage of OM is typically higher than that of aluminum sulfate (50-800 mg/L vs. 10-50 mg/L), which can increase the cost per m^3 of treated water (MUYIBI et al., 2003).

CONCLUSION

Moringa oleifera establishes itself as an ecologically sustainable and technically viable alternative for hard water treatment. The action of its cationic proteins, through load neutralization and adsorption, demonstrates high efficiency in the removal of turbidity, color, heavy metals and microorganisms. Pseudo-second-order kinetics and Langmuir isotherm indicate an effective chemisorption process. The proposal of a hybrid system with activated carbon maximizes the removal of contaminants. The economic and environmental viability, especially in rural contexts, reinforces OM as a promising solution for global water security.

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