Journal of Aquatic and Terrestrial Ecosystems
1(1)
Received: August 10, 2023
Accepted: September 14, 2023
Published: September 18, 2023

Efficacy of Treating Wastewater from Wastepaper Recycling Mill by Blending *Moringa Oleifera* with Synthetic Coagulants

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Abstract

Wastepaper recycling is a growing global industry with worrying concerns in water pollution that emanate from defibering, deinking and paper making processes. This study sought to determine the efficacy of treating wastewater from wastepaper recycling mill using effective doses of individual and blended *Moringa oleifera* and synthetic coagulants. Wastewater samples were obtained from Maz International Paper Mill located in Kajiado County, Kenya. A randomized experimental design was applied in the study. Treatment efficiency was determined using standard jar test procedures and APHA standard methods. The data obtained was subjected to one-way analysis of variance using Stratigraphics version 16. The study revealed that alum was the most efficient individual coagulant whereas blend of deffated *Moringa oleifera* seeds and alum (DMos/Alum) was most efficient coagulant in treating the wastepaper recycling mill wastewater. The blend of DMos/Alum efficiently BOD, color, TDS, EC, and TSS to 28.7 mg/L, 14.4 PCU, 267.8 mg/L, 495.6 µs/cm, and 5.8 mg/L, respectively. These final DMos/Alum treated wastewater parameters were within WHO, NEMA, and USEPA permissible drinking water thresholds. Additionally, the DMos/Alum blend resulted in the highest microbial load removal of 99.2%. In conclusion, the wastewater from wastepaper recycling mill was effectively treated using the blend of DMos and alum. The study recommends that a blend of DMos and alum can be applied in treating wastewater from wastepaper recycling mills.

Keywords: Efficacy, wastewater, *moringa oleifera*, synthetic coagulants

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Competing interests: The authors have declared that no competing interests exist

Introduction

Wastepaper recycling technique is one of the effective technologies being applied in solid waste management to produce secondary fibres and subsequently in paper production (Li *et al*., 2020). Moreover, wastepaper recycling has been on upward trend in production of secondary fibers to supplement virgin fibers use (Abd El-Sayed, El-Sakhawy, & El-Sakhawy, 2020; Caltagirone *et al*., 2021). Wastepaper recycling mills alike other industries worldwide have steadily increased due to the demand for paper and paper products culminated by the increased human population (Sharma, 2022). Globally, annual wastepaper recovery has been reported to be more than 58%, with developed economies recovering more than 70% of the wastepaper (de Oliveira *et al*., 2023). Utilization of secondary fibers through recycling wastepaper plays a significant role in environment conservation. Wastewater from wastepaper recycling mills often contain high concentrations of

BOD, pH, COD, TDS, EC, TSS, AOX (adsorbable organic halides), phenolic compounds, heavy metals, plant materials. These pollutants, when discharged improperly from the industries, they pose threats to aquatic and terrestrial lives (Haq & Raj, 2020). High BOD, COD and heavy metals in wastewater from wastepaper recycling mills have detrimental effects to aquatic and terrestrial lives. The high levels of BOD, TDS, TSS, and COD, deplete the oxygen levels for aquatic life and microbial organisms. Globally, paper mills are regarded as intensive freshwater consumers, culminating in the discharge of massive volumes of wastewater into receiving ecosystems. Maz International Wastepaper Recycling Mill (MIWPRM) in Kenya processes wastepaper into paper products, resulting to wastewater that is highly colored, turbid and with lots of fibers. This resultant wastewater doesn't meet required wastewater regulations by NEMA despite the use of stabilization ponds for wastewater treatment by MIWPRM.

Materials and methods

Study area

Wastewater samples were obtained from Maz International Wastepaper Recycling Mill (MIWPM) located at Kisaju, Kajiado County along the Nairobi-Namanga Road (latitude 1°38'28.75"S; longitude 36°52'38.90"E). Figure 1 shows the location map of Maz International wastepaper recycling mill.

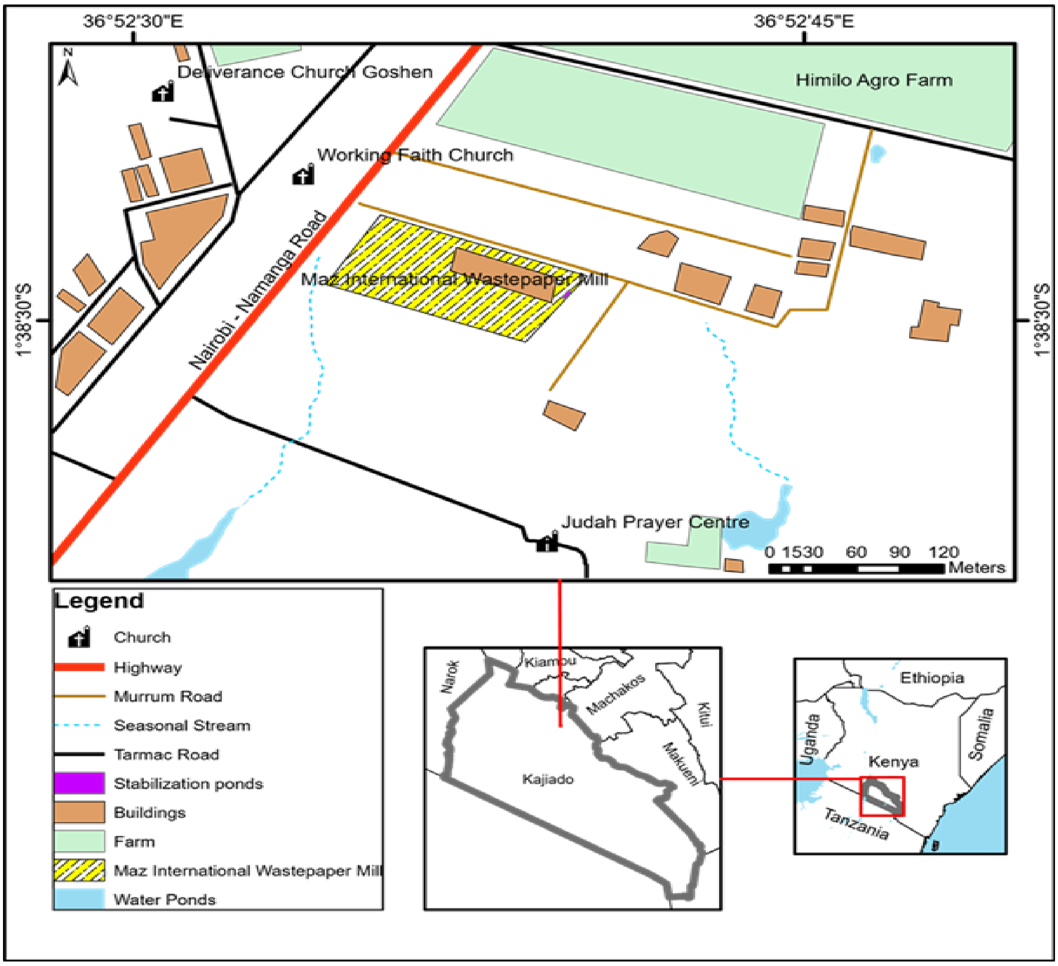

Figure 1: Location map of Maz international wastepaper recycling mill

Methods

A completely randomized design was adopted to determine the efficacy treatment of wastewater from MIWPRM by individual and blended coagulants using batch jar test experiments.

The study adopted grab sampling method in collection of wastewater samples. The pH, temperature and electrical conductivity were measured using HANNA pH multimeter. The composite samples collected were transported to the laboratory in conducive conditions that ensured biological degradation did not take place. Ten kilos of *Moringa oleifera* bark and seed kernels were collected from Kirinyaga County and transported to the laboratory for further processing.

Moringa oleifera seeds were sun-dried, mechanically dehusked, ground, sieved and stored for DMos and FMos stock solutions preparations. Oil from crude powder was extracted using the Soxhlet technique and prepared for experiments following Ojewumi et al. (2019) recommendations and the obtained DMos stored for experiments.

A stock solution of polyaluminium chloride was prepared by diluting 12g of PAC with 500ml wastewater while 5g of alum were diluted with 500ml wastewater to achieve 1%wt solution. Bio-coagulants stock solutions, 100g of DMos and 100g of FMos were diluted with 500ml wastepaper wastewater each and stirred using an electric shaker for 24 hours and their supernatants used for jar tests batch treatments. Whereas 200g of BMo were diluted with 500ml wastewater and stirred for 24 hours.

Batch jar test experiments were conducted in treatment of the wastewater using individual and blended coagulants. Efficacy of wastewater treatments were carried out using standard methods of wastewater pollution according to (Rice *et al.*, 2012).

Results

Efficacy of treating wastewater using blended of *Moringa oleifera* and chemical coagulants easily degradable materials

The highest BOD reduction among individual coagulants was of alum with efficiency of 90.93% followed by PAC with efficiency of 81.78%. The lowest BOD reduction was by BMo coagulant. There was a significant difference in BOD reduction among all individual coagulants ($F_{0.05\ (4,\ 40)}$ =141.59, $p<0.0001$) as portrayed in table 1. Among the blended coagulants the highest BOD reduction efficiency of 92.05% was established by the blend of DMos/Alum followed by FMos/Alum and DMos/PAC. The lowest BOD reduction was by the blend of FMos/PAC with efficiency of 84.59% as portrayed in table 1. Despite the achieved reductions by individual and blended coagulants, only the blend of DMos/Alum reduced BOD to levels within (20-30 mg/L) WHO, NEMA and USEPA drinking water standards.

Table 1: Biological Oxygen Demand of treated wastewater by various coagulant

Coagulant	Initial (mg/L)	Final (mg/L)	Reduction(mg/L)	Efficiency (%)
Alum	360.4±7.3	32.7±4.7	327.8	90.9
PAC	360.0±7.0	65.6±6.7	294.4	81.8
DMos	360.9±7.7	78.4±6.1	282.4	78.3
FMos	361.6±9.3	88.7±4.7	272.9	75.5
BMo	360.7±6.3	140.0±5.5	220.7	61.2
DMos/Alum	360.7±5.0	28.7±3.2	332.0	92.1
FMos/Alum	360.0±5.5	31.3±4.0	328.7	91.3
DMos/PAC	361.3±6.9	34.0±3.7	327.3	90.6
FMos/PAC	360.9±7.4	55.6±3.1	305.3	84.6

Complete degradability of wastewater

Complete degradability of wastewater was achieved by measuring Chemical Oxygen Demand (COD). Alum, as an individual coagulant, portrayed a high Chemical oxygen demand (COD) reduction efficiency of 78.32% followed by PAC. The least COD reduction was by BMo with an efficiency of 50.45%. COD reductions among all individual coagulants were significantly different ($p<0.0001$) as shown in table 2.

Table 2: Chemical Oxygen Demand of treated wastewater by various coagulant

Coagulant	Initial (mg/L)	Final (mg/L)	Reduction(mg/L)	Efficacy (%)
Alum	598.7±8.8	129.8±5.9	468.9	78.3
PAC	602.2±8.8	180.9±6.4	421.3	70.0
DMos	599.3±9.1	214.4±6.9	384.9	64.2
FMos	596.4±9.2	232.0±7.1	364.4	61.1
BMo	597.1±13.8	295.8±9.5	301.3	50.5
DMos/Alum	599.8±9.4	113.8±4.1	486.0	81.0
FMos/Alum	598.2±8.4	127.6±6.0	470.7	78.7
DMos/PAC	598.2±12.5	118.0±5.5	480.2	80.3
FMos/PAC	600.0±7.9	168.0±4.9	432.0	72.0

Among the blended coagulants COD reductions efficiencies were high in DMos/Alum, DMos/PAC and FMos/Alum at 81.02%, 80.27% and 78.68% respectively. The blend of FMos/PAC had low COD reduction as portrayed in table 2. However, the reduced COD levels exceeded the WHO, NEMA and USEPA drinking water limits of less than 90 mg/L COD.

Transparency of wastewater

Alum, as an individual coagulant had the color reduction efficiency of 96.61%. DMos followed with a reduction efficiency of 94.98%. BMo had the lowest color reduction efficiency and was significantly different from other coagulants ($p<0.0001$) as portrayed in table 3.

Table 3: Transparency of treated wastewater by various coagulant

Coagulant	Initial (PCU)	Final (PCU))	Reduction (PCU))	Efficacy (%)
Alum	900.0±50.2	30.3±3.9	869.7	96.6
PAC	881.7±27.4	235.0±13.7	646.7	73.3
DMos	877.2±22.2	44.1±2.9	833.2	95.0
FMos	859.4±37.0	170.0±12.5	689.4	80.2
BMo	863.9±30.7	730.6±35.8	133.3	15.4
DMos/Alum	876.4±36.3	14.4±2.9	634.5	98.3
FMos/Alum	891.7±17.7	27.7±3.9	864.0	96.9
DMos/PAC	873.3±47.0	40.2±4.6	833.2	95.4
FMos/PAC	881.1±19.5	167.8±13.5	713.3	81.0

Among the blended coagulants, the blend of DMos/Alum had the highest color reduction efficiency of 98.29%, followed by FMos/Alum and DMos/PAC as shown in table 3, while the blend of FMos/PAC had the lowest color reduction with an efficiency of 80.95% which was significantly different from other blends ($p<0.0001$). DMos/Alum reduced color to less than 15 PCU which are allowable drinking water limits as per WHO, NEMA and USEPA.

Dissolved Solids in the wastewater

DMos coagulant had the highest TDS at a reduction efficiency of 86.07% followed by FMos with reduction efficiency of 78.51%. BMo and PAC individual coagulants resulted in the lowest TDS reductions as portrayed in table 4. There was a significant difference in TDS reductions among all individual coagulants.

Table 4: Total Dissolved Solids of treated wastewater by various coagulant

Coagulant	Initial (mg/L)	Final (mg/L)	Reduction(mg/L)	Efficacy (%)
Alum	1820.0 ± 51.5	770.0 ± 39.69	1050.0	57.7
PAC	1781.1 ± 35.9	1281.1 ± 41.7	500.0	28.1
DMos	1817.8 ± 41.2	253.3 ± 31.6	1564.4	86.1
FMos	1775.6 ± 32.1	381.1 ± 60.7	1394.4	78.5
BMo	1795.6 ± 34.3	831.1 ± 32.6	964.4	53.7
DMos/Alum	1784.4 ± 39.1	267.8 ± 28.6	1516.7	85.0
FMos/Alum	1778.9 ± 29.3	355.6 ± 41.0	1423.3	80.0
DMos/PAC	1815.6 ± 39.1	286.7 ± 32.8	1528.9	84.2
FMos/PAC	1803.3 ± 38.1	447.8 ± 41.5	1355.6	75.2

For the blended coagulants, DMos/Alum and DMos/PAC had the highest reduction efficiencies of 84.98% and 84.21% respectively with no significant difference ($p>0.05$). The blends of FMos/Alum and FMos/PAC were significantly different with TDS reductions as shown in table 4. Despite the efficient TDS reductions obtained, PAC reduced levels exceeded the TDS drinking water limits of less than 1000 mg/L as per WHO and NEMA.

Wastewater Conductivity

The highest EC reduction achieved by DMos coagulant at an efficiency of 82.36%. BMo and PAC coagulants had the lowest EC reductions efficiencies of 48.11% and 23.01% respectively.

There was a significant difference in EC reductions among the individual coagulants ($p<0.0001$). Albeit, the EC reduced levels by PAC coagulant exceeded WHO, NEMA and USEPA drinking water EC allowable limits. For the blended coagulants DMos/Alum had the highest EC reduction efficiency followed by the blend of DMos/PAC at efficiencies of 79.98% and 79.32% respectively, which were significantly different from EC reductions other blends as shown in table 5.

Table 5: Electrical conductivity of treated wastewater by various coagulant

Coagulant	Initial (µs/cm)	Final (µs/cm)	Reduction(µs/cm)	Efficacy (%)
Alum	2477.8 ± 42.4	885.6 ± 30.9	1776.7	64.3
PAC	2486.7 ± 32.0	1914.4 ± 47.2	572.2	23.0
DMos	2467.8 ± 38.0	435.6 ± 34.7	2032.2	82.4
FMos	2497.8 ± 38.0	724.4 ± 28.8	1773.3	71.0
BMo	2476.7 ± 41.8	1285.6 ± 51.0	1191.1	48.1
DMos/Alum	2476.7 ± 44.7	495.6 ± 39.1	1981.1	80.0
FMos/Alum	2467.8 ± 38.0	691.1 ± 39.5	1776.7	72.0
DMos/PAC	2476.7 ± 45.6	512.2 ± 33.5	1964.4	79.3
FMos/PAC	2497.8 ± 38.0	650.0 ± 48.7	1847.8	74.0

Wastewater suspended solids

Total suspended solids reduction by individual coagulants was highest in alum, DMos, FMos and PAC with reduction efficiencies of 96.64%, 95.07%, 93.78% and 91.47% respectively. BMo had the lowest TSS reduction with a significant difference ($p<0.0001$) as shown in table 6. However, the reduced TSS levels by PAC, FMos and BMo exceeded the allowable drinking water thresholds of 30mg/L in accordance to WHO, NEMA and USEPA. For the blended coagulants; DMos/Alum blend achieved the highest TSS reductions at an efficiency of 99%. All the blends were significantly different ($p<0.0001$) in total suspended solids reduction as illustrated in table 6.

Table 6: Total suspended solids of treated wastewater by various coagulant

Coagulant	Initial (mg/L)	Final (mg/L)	Reduction(mg/L)	Efficacy (%)
Alum	577.8 ± 30.3	19.4 ± 3.7	558.3	96.6
PAC	571.6 ± 22.4	48.8 ± 4.9	522.8	91.5
DMos	550.7 ± 19.6	27.1 ± 3.1	523.6	95.1
FMos	564.4 ± 43.9	34.9 ± 3.7	529.6	93.8
BMo	556.1 ± 16.7	256.1 ± 12.2	300.0	53.9
DMos/Alum	564.4 ± 43.9	5.8 ± 2.1	558.7	99.0
FMos/Alum	566.0 ± 23.5	17.0 ± 4.6	549.0	97.0
DMos/PAC	557.2 ± 17.0	11.1 ± 3.2	546.1	98.0
FMos/PAC	573.3 ± 27.8	28.8 ± 6.1	544.6	95.0

Microbial load reduction

DMos individual coagulant had the highest microbial load reduction efficiency of 97.51%, followed by FMos with a reduction efficiency of 93.99%, while PAC had the least reduction efficiency of 77.19%. There was a significant difference ($p=0.0000$) in microbial load reduction among all individual coagulants treatments as shown in figure 2

For the blends, DMos/Alum had the lowest microbial load with a reduction efficiency of 99.16% followed by DMos/PAC with a reduction efficiency of 96.99%. There was a significant difference ($p=0.0000$) in all the final microbial load for all blends of coagulants. However, the reduction efficiencies of the individuals and blends exceeded the WHO, NEMA and USEPA drinking water limits of Nil CFU/ml.

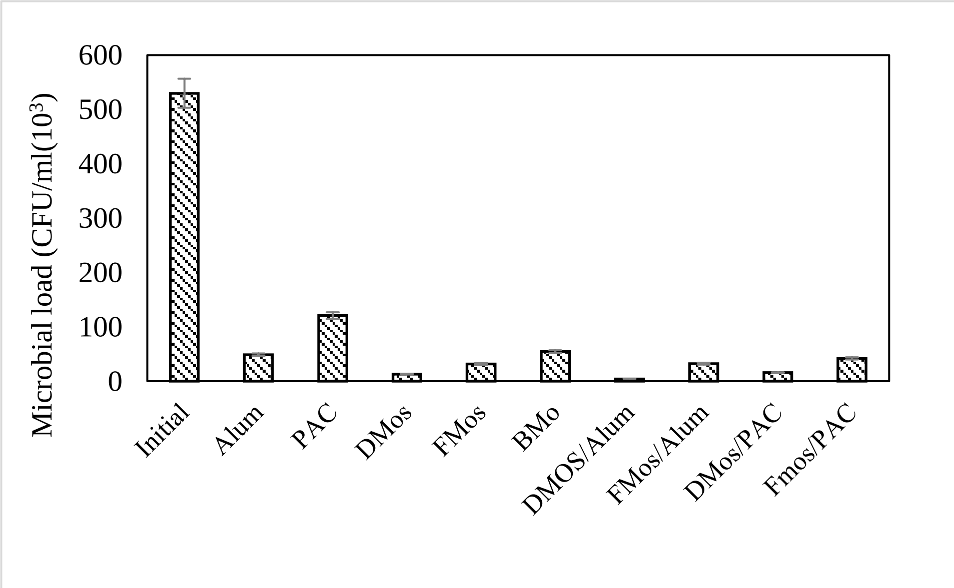

Figure 2: Microbial load reduction by individual and blended coagulants

Discussions

The blend of DMos and alum coagulation mechanisms; particle adsorption, interparticle bridging, and charge neutralization contributed to its effectiveness in BOD reduction (Mehmood *et al*., 2019). The lowest BOD reduction by BMo was due to inadequate coagulation resulting from low polypeptide concentration, facilitating colloidal particle destabilization. However, despite presence of polyelectrolytes in *Moringa oleifera* seeds, the oil content in fatted seeds hindered effective coagulation and contributed to biodegradable matter, resulting in lower BOD reduction compared to defatted seeds (Boulaadjoul *et al*., 2018). The findings were in accordance with those of Al-Jadabi *et al*. (2021)who described significantly higher BOD reductions with aluminium sulphate as likened to fatted *Moringa oleifera* seeds at 75.5% and 72% respectively from domestic wastewater treatment through coagulation process. Additionally, Municipal wastewater treated with defatted Moringa oleifera seeds reduced BOD levels by 91.81%, slightly higher than the study's findings due to wastewater nature (Adelodun *et al*., 2019).

The combination of DMos/Alum and DMos/PAC showed high chemical oxygen demand removal efficiencies due to polymerized metal ionic species and positively charged polypeptides in defatted *Moringa oleifera* seeds. This led to effective coagulation, eliminating both inorganic and organic pollutants by embedding particulate adsorption and forming flocs, achieving high COD reduction efficacy. (Desta & Bote, 2021; Naceradska *et al*., 2019). These study findings aligned with those of Al-Jadabi *et al*, (2021) who reported that, *Moringa oleifera* defatted seeds were observed to have a lesser COD decrement of 72 % than aluminium sulphate, which whittled down COD by 75.5 % at a dosage of 150 mg/L in treating domestic wastewater. In contrast with the findings, Rifi *et al*. (2022) reported 88% COD removal using *Moringa oleifera* seeds in the treatment of wastewater from an olive oil mill which contrasted with the findings of the current study.

The efficient color removal by the blend of DMos/Alum was attributed to positively charged proteins in defatted *Moringa oleifera* seeds which enabled adsorption and interparticle bridging coagulation mechanisms, destabilizing colloidal particles via charge neutralization (Wagh *et al*., 2022). Furthermore, the monomeric aluminium solubilized species in aluminium sulphate figured prominently in destabilization of negatively charged particles via electrostatic interactions. Precipitation of colloidal particles reduced the color levels in the treated wastewater (Mehmood *et al*., 2019). Through the clustering of the precipitated particles, the efficacious coagulation from defatted Moringa seeds and the amorphous hydrolysate ions attributed to reduced color levels (Boulaadjoul *et al*., 2018). Consistent with current study findings, the use of defatted *Moringa oleifera* in synthetic dairy wastewater treatment effectively reduced color levels by 94% as reported by Wagh *et al*, (2022). Additionally, 95% color removal was reported for aluminium sulphate coagulation on paper industry wastewater (Öztürk & Özcan, 2021). In contrast, polyaluminium chloride was reported to remove 95% of color from textile dyeing effluents under optimal conditions (Islam & Mostafa, 2020).

The organic polypeptides in DMos adsorbed the dissolved solids in the wastewater through particle adsorption and bridging mechanisms hence neutralizing the charges this led to their reduction as they clumped together forming flocs (Shan *et al*., 2017). For the synthetic coagulants, the existence of multivalent, solubilized, and absorbable aluminium ions made up a significant portion of the dissolved solids. However, during coagulation mechanisms some of these neutralized ions, were not completely eliminated as flocs clustered. These study results aligned with those of Panhwar *et al*. (2020) who reported TDS reduction from 2630 mg/L to 1640. Contrary to study findings, the treatment of textile dyeing effluent with polyaluminium chloride was reported to yield 85.7% TDS reduction efficiency (Islam & Mostafa, 2020). However, in alignment with the study findings, treating raw water with a 60:40 dosage ratio mixture of defatted *Moringa oleifera* seeds and polyaluminium chloride reduced TDS levels from 171 mg/L to 43 mg/L(Cardoso Valverde *et al*., 2018).

The removal of dissolved electrolytes was credited to the cationic proteins functional group in the DMos hence the highest EC reduction (Desta & Bote, 2021). These proteins adsorbed and neutralized the colloidal particles through interparticle bridging, facilitating their agglomeration and removal from the treated effluent. Similarly to these study findings, 86.28% electrical conductivity reduction was reported after using defatted *Moringa oleifera* in phytoremediation of commercial laundry wastewater (Hakeem *et al*., 2019). The existence of residual aluminium ions dissolved in water from hydrolyzed multivalent aluminium species resulted to inefficient electrical conductivity reduction using polyaluminium chloride. These research findings were in contrast with those of Islam & Mostafa, (2020) who reported 83.66% reduction efficiency of electrical conductivity in treatment of textile dyeing effluent using PAC. The research findings were in line with those of Gandiwa *et al*. (2020), who reported decrease in electrical conductivity to a final value of 308.2 using combination of crude *Moringa oleifera* seeds and alum in raw water treatment.

The blended coagulants achieved high reduction efficiencies due to the presence of polyelectrolytes in *Moringa oleifera* seeds, hydrolyzed ions in polyaluminium chloride, and amorphous ions in aluminium sulphate, facilitating coagulation. The study findings were in consistent with Jagaba *et al*. (2018), who reported 97.19% TSS removal efficiency in treatment of palm oil mill wastewater using combined dosage of 4g/L of alum and 2g/L of *Moringa oleifera* seeds. Moreover, the comparative coagulation efficacy using defatted *Moringa oleifera* seeds and aluminium sulphate for domestic wastewater achieved 95.5% and 96.8% TSS reduction (Al-Jadabi *et al*., 2021). Additionally, defatted *Moringa oleifera* seeds removed 97.4% total suspended solids from treatment of coal beneficiation plant effluent (Kapse & Samadder, 2021).

Pathogen removal in wastepaper recycling mill wastewater was achieved through coagulation. *Moringa oleifera* seeds' bioactive components, including isothiocyanate complexes and fatty acids, provided antimicrobial effects by destroying cell membranes. This resulted in an increase in the discharges of solutes from the microbial cells, thus their death (Prajapati *et al*., 2022). Additionally, the influence of minerals, ketones, esters, and aromatic amines in the seeds precluded the early stages of bacterial cell wall synthesis and buildup onto cellular membrane that made them impermeable, this impeded the metabolism of the bacteria and caused cell death (Taiwo *et al*., 2020). *Moringa oleifera* seeds effectively coagulated wastewater through adsorption, interparticle bridging, and charge neutralization techniques, removing bacterial load as agglomerated flocs. Sterol glycoside compounds inhibit gram-positive bacteria growth, resulting in effective microbial load reduction (Azad & Hassan, 2020). The presence of aluminium ions in the blend of DMos/Alum attributed to slightly lowered pH levels thus microbial growth inhabitation (Taiwo *et al*., 2020). In accordance with study results, utilizing defatted *Moringa oleifera* seeds and aluminium sulphate in tertiary treatment of domestic wastewater resulted in total coliforms reduction efficiencies of 99.6% and 99.5 %, respectively (Andrade *et al*., 2021). Moreso, the use of aqueous *Moringa oleifera* seeds highly reduced bacterial load compared to aluminium sulphate reduction efficiency in the treatment of domestic wastewater (Vunain *et al*., 2019). In contrast with the study findings, Njewa *et al*. (2021) reported increase of microbial load while using *Moringa oleifera* seeds in clarification of sewage wastewater as compared to *Jatropha curcas* and rice husks ashes.

Conclusion and recommendation

The most efficient coagulant among the bio-coagulants was DMos while among synthetic coagulants was alum. The study revealed among the blended coagulants DMos/alum was the most efficient in most wastewater parameters reductions. In microbial load reduction from the wastewater from wastepaper recycling mill, the study divulged that the blend of DMos and alum was the best coagulant. It was also found out *Moringa oleifera* reduced the microbial load from the wastewater from the wastepaper recycling mill at higher rates than synthetic coagulants.

The study recommends the use of DMos and Alum blend for wastewater treatment to remove microbial loads from wastepaper recycling mills. Further research should be done to evaluate effects of extraction of oil and/or extraction of active components using saline solution for efficient coagulation of Moringa oleifera seeds in treatment of industrial wastewater treatment. Further research should be conducted on cost effectiveness of blending Moringa oleifera and/or other plant-based coagulants and synthetic coagulants in industrial wastewater.

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