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APPLICATION OF *MANGIFERA INDICA* (MANGO) AND *PHOENIX DACTYLIFERA* (DATES) SEEDS POWDERS AS COAGULANTS IN WASTEWATER TREATMENT

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ABSTRACT
This research was aimed at studying the possibility of efficiently combining Mangifera indica and Phoenix dactylifera seeds powders as coagulants in wastewater treatment. The seeds were characterized for their composition, active compounds and proximate constituents using atomic adsorption spectroscopy, Scanning Electron Microscopy, and X-ray Diffraction analysis. Jar test was carried out using the seeds extracts separately and in combination at varying dosages and the Electrical Conductivity, Total Dissolved Solids, Chemical Oxygen Demand, Biochemical Oxygen Demand, Turbidity, Temperature, pH, and Total coliform were measured with every varying dosage. Statistical tools were used to validate the results of the Jar test. The sludge from the Jar test experiment was analyzed for its total nitrogen, organic carbon, organic matter, total volatile solids, and carbon: nitrogen ratio. Mangifera indica removed 89% turbidity, 96% BOD, 84% COD and 99% total coliform of the wastewater. Phoenix dactylifera removed 75% turbidity, 85% BOD, 78% COD and 88% total coliform of the wastewater. Their combination removed 96% turbidity, 96% BOD, 87% COD and 98% total coliform of the wastewater.

Keywords: *Mangifera indica, Phoenix Dactylifera,* Coagulants, Wastewater

1. INTRODUCTION

The world population is growing at an alarming rate with water resources increasingly becoming scarce. The needs for water conservation and water management have never been more urgent. This is because our very existence as humans depends on the continued availability of this resource in good quantity and quality [25]. At the heart of this, is the ability to utilize water and wastewater effectively. To achieve this goal, wastewater must be treated in an economic manner [18]. In places where fresh water is in abundance, most often than not, people have to put on with the problem of heavy pollution. This makes it very expensive to treat water to meet the minimum standard required for domestic uses such as drinking and cooking and other industrial uses. Polluted water when not treated can be harmful to humans, animals and the environment.

According to WHO [27], 3,575,000 people die from water related diseases every year. Similarly, 884 million people lack access to safe drinking water and five die annually out of the 361,000 under children, mostly in low-income countries [27]. Dissolved and suspended particles form a major part of the impurities in most natural waters. These suspended materials mostly arise from erosion of the top soil, the dissolution of minerals and the decay of vegetation and from several domestic and industrial waste discharges.

Large solids can be removed by allowing them to settle (sedimentation) and then filtered (Filtration). Suspended particles and dissolved solids settle too slowly and may also pass through filters. Coagulation and flocculation processes are used to separate the dissolved and suspended particles from the water.

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The demand for water treatment chemicals has increased substantially from emerging economies such as Brazil, China, and India [23]. The conventional methods of assuring potable water in developing economies are unsustainable at the moment and may remain so for a long time. This necessitates the need to consider the application of sustainable technologies using locally available materials in water treatment. A prospective area is the Plant kingdom and the use of seeds in particular. Some seeds have the potentials of serving as alternative sources of coagulants owing to their advantages over the conventional organic and inorganic coagulants.

2. THE OBJECTIVE OF THE RESEARCH

This research was carried out to investigate the efficiency of combining *Mangifera indica* (Mango) and *Phoenix dactylifera* (Dates) seeds extract as coagulants in wastewater treatment. The mineral compositions and proximate analyses of *Mangifera indica* and *Phoenix dactylifera* seeds powders were determined. The active ingredients in *Mangifera indica* and *Phoenix dactylifera* seeds powders were also determined and treatability studies to determine the effect of graduated combined dosage of *Mangifera indica* and *Phoenix dactylifera* seeds powders on the physicochemical and bacteriological water quality parameters was performed

3. MATERIALS AND METHODS

3.1 Materials

The following materials were used in the experiment. Jar test flocculator (Peterson Candy), Soxhlet Extractor, pH meter (CRISON micro pH 2000), Muffle furnace (S30 2AU), weighing balance (Mettler H31), Atomic Absorption Spectrophotometer (Shimadzu AA6800), Mortar and Piston, Turbidy Meter (Hack Chemical Limited), Filter Paper, Stop watch, Magnetic Stirrer and Magnetic Oven. Others were burette, conical flask, pipette, funnel, spatula, flat-bottom flask.

3.2 Methods

3.2.1 Wastewater sampling

Wastewater was sampled from the influent of the wastewater stabilization pond of A.B.U, Zaria. This was done by using two 25 litres plastic containers at a point where the velocity of the water was low enough to allow for getting a representative sample. The samples collected were grab samples.

3.2.2. Mangifera Indica and Phoenix Dactylifera Seeds Preparation

The seeds (Mango and Dates) were sourced locally from Sabon Tasha and Kawo markets in Chikun and Kaduna North local government areas in Kaduna state. They were washed, cleaned and dried under the sun for several days until they were completely dried, and any foreign material noticed was removed from the seeds. The dried clean seeds (after the removal of the husk or seed coating and separation of the seeds from chaff) were then crushed to reduce their particle sizes to between 2mm – 70um. Solvent extraction was used to remove oil from in the seeds powder. 200g of the pre-processed seeds was treated in a multistage counter current process with hexane (500ml) as the solvent in soxhlet extractor until the oil content was reduced to the lowest possible level. The mixture of oil and solvent were then separated by distillation with the cake washed with distilled water, dried in an oven to constant weight and then sieved using a size 75 microns Microplate sieve with the fine particles to be used as the coagulants.

3.2.3. Determination of Mineral Composition

The mineral contents of the samples were analyzed using the Atomic Adsorption Spectro-phometer (AAS instrument). The minerals analyzed using AAS were Ca, Fe, Mn, Mg and Pb. Flame Photometer was used to analyze for K and Na. Samples of *Mangifera indica* and *Phoenix dactylifera* seeds to be digested were prepared in concentrated HNO_3. Working standards and blank samples were prepared for each of the mineral elements. Readings were then taken and the concentration of each of the elements were computed in mg/L according to Orijajogun *et al* [17].

3.2.4. Proximate Analysis

The analyses included in this group, also known as Weende proximate analyses, are applied firstly to materials to be used in formulating a diet as a protein or energy source and to finished feedstuffs, as a control to check that they meet the specifications or requirements established during formulation. These analyses showed the moisture, crude protein (total nitrogen), crude fibre, crude lipids, ash and nitrogen-free extract content of the sample [6].

3.2.5. Determination of the Active Ingredient

The active ingredients in *Mangifera indica* and *Phoenix dactylifera* seeds were determined using the scanning electron microscope (SEM) combined with X-ray diffractometer. Specimens of the samples were prepared and a beam of electrons was focused on the sample surface which then gave information on the composition and surface topography of the samples. The samples were analyzed before they were used in the coagulation process.

3.2.6. Bacteriological Examination

The standard Plate Count method was used to test for coliform units in the wastewater sample, which relies on bacteria growing in a colony on a nutrient medium so that the colony becomes visible to the naked eye and the number of colonies on a plate can be counted. To ensure that an appropriate number of colonies were generated, several dilutions were cultured.

3.2.7. Jar Test

The ASTM D2035 – 13, standard practice for coagulation-flocculation Jar test of water was used to carry out the test. *Mangifera indica* (MI) and *Phoenix dactylifera* (PD) were used separately and in combination in different proportions to carry out the Jar test. Jar test was carried out with rapid mixing of about 100 rpm for 1 minute and slow mixing about 30 rpm for 30 minutes. Residual turbidity for different combinations of coagulant dosages was then measured in the interval of 60, 120 and 720 minutes [3].

3.2.8. Data Analysis

Regression and ANOVA analyses were used to ascertain the proportion of the variance in the dependent variable (turbidity) that is predictable from the independent variable (dosage) and to know the statistical significance of the overall model. Microsoft excel 2010 was used to carry out the data analyses.

3.2.9. Determination of Sludge Characteristics

Total Nitrogen Determination: DANI 89.00 CHN elemental analyzer was used to determine the nitrogen content of the sludge. This instrument automatically determines C-H-N by combustion of the sample, separation of the combustion products by means of a programmed temperature desorption system, and measurement by thermal conductivity.

Organic Carbon: DANI 89.00 CHN elemental analyzer was used to determine the total organic carbon of the waste content. Determination of total organic carbon was carried out by running replicates at 500° and 1100°C with the Analyzer

Organic Matter: The organic matter in the sludge was determined by the oxidation of potassium permanganate using the gravimetric method of chemical analyses.

Total Volatile Solids: Total volatile solids were determined using gravimetric method as outlined by the USEPA Method 1684 for the determination of total solids and the fixed and volatile fractions in such solid and semisolid samples as soils, sediments, biosolids (municipal sewage sludge), sludge separated from water and wastewater treatment processes, and sludge cakes from vacuum filtration, centrifugation, or other sludge dewatering processes.

4. RESULTS AND DISCUSSION

Table 1 and 2 shows the results of the analyses of chemical composition and active compounds in *Mangifera indica* and *Phoenix dactylifera* seeds powders respectively.

Both seeds contain potassium and sodium as macro elements while calcium, iron, manganese, magnesium, lead and zinc are micro elements. All the elements, except Lead, fall within the acceptable limits for drinking water as contained in the Drinking Water Standards of Nigeria [14].

Table 2 gives the result of the SEM and XRD analyses carried out at the department of Physics, Umaru Musa Yar'adua University, Kastina. *Mangifera indica* contains mango starch, p-Carboxybenzaldehyde, which is a polyphenolic compound and o-Phthalic acid. *Phoenix dactylifera* contains Potassium Aluminum Silicate and Potassium Copper Chloride Hydrate as the active compounds. P-Carboxybenzaldehyde is used as Intermediate for Pharmaceuticals and as a metabolite in ampicillin. O-Phthalic acid is used in the production of chemicals such as dyes, perfume and saccharin. Potassium Aluminum Silicate is mostly used as an anti-caking agent.

4.1 Proximate Analysis

Table 3 shows the proximate analysis for *Mangifera indica* and *Phoenix dactylifera*. From the table, *Mangifera indica* and *Phoenix dactylifera* have protein values of 6.81% and 4.94% respectively, ash content of 2.44% and 1.61%, crude fiber of 8.01% and 19.33%, oil of 3.63% and 5.36%. This is within the range of the results of the similar studies on mangoes and dates reported by Mutua *et al* [12] and Harrasi *et al* [7] respectively. However, the results of the proximate analysis differ slightly from those of other seeds like *moringa* (most effective natural coagulant used in both water and wastewater treatment) which has been reported by Mikore and Mulugeta [10] to have a crude protein value of between 24 to 28%, ash content of between 14 to 16%, crude fiber of between 5 to 7% and oil of between 3 to 7%.

4.2 Treatability studies

Table 4 shows the parameters of the raw water before application of the treatment with the natural coagulants, while Tables 5, 6 and 7 show the results of the parameters after the application of graduated dosages of *Mangifera indica*, *Phoenix dactylifera*, and the combination of the two seeds extract on the raw water respectively. Finally, Table 8 shows the results of the parameters with the application of Alum as the coagulant.

Table 1: Chemical compositions of Mangifera indica and Phoenix dactilyfera

	Potassium %	Sodium %	Calcium ppm	Iron ppm	Manganese ppm	Magnesium ppm	Lead ppm	Zinc ppm
Mangifera I.	1.14	0.4	8.14	4.63	0.18	12.15	0.13	0.08
Phoenix D.	0.36	0.28	6.95	2.39	0.00	6.82	0.22	0.12

Table 2: Active compounds in Mangifera indica and Phoenix dactylifera

Sample	Active Compounds
Mangifera Indica	Carbon hydrogen
	Mango starch **$(C_6H_{10}O_5)n$**
	p-Carboxybenzaldehyde **$(C_8H_6O_3)$**
	o-Phthalic acid **$(C_8H_6O_4)$**
Phoenix Dactylifera	Potassium Aluminum Silicate **$(KAlSi_3O_8)$**
	Potassium Copper Chloride Hydrate **$(K_2CuCl_4 \cdot 2H_2O)$**

Table 3: Proximate Analysis of Mangifera indica and Phoenix Dactylifera seeds

	Moisture Content %	Dry Matter %	Crude Protein %	Crude Fiber %	Oil %	Ash %	Nitrogen FE %
Mangifera I	9.33	90.67	6.81	8.01	3.63	2.44	69.78
Phoenix D	16.18	83.82	4.94	19.33	5.36	1.61	52.58

Table 4: Natural Turbid Wastewater Parameters

SAMPLE	EC µS/cm	BOD mg/L	COD mg/L	TURB NTU	TDS mg/L	pH	Temp °C	Coliforms CFU/100ml
RAW	4000	140	1000	796	1728	6.9	26.6	>350x10^4

Table 5: Jar Test results with Mangifera indica

SAMPLE	DOSAGE mg/L	EC µS/cm	BOD mg/L	COD mg/L	TURB NTU	TDS mg/L	pH	Temp °C	Coliforms CFU/100ml (X 10^4)
MI	5	2736	55	380	185	1449	7.8	28.4	75
MI	10	2803	35	200	185	1503	7.1	28.2	28
MI	12.5	2994	15	190	155	1510	7.1	27.5	21
MI	15	2846	10	180	118	1469	7.5	27.5	7
MI	20	2996	5	170	99.3	1773	7.0	28.1	2
MI	25	2955	5	160	79.9	1396	6.9	27.7	9

Table 6: Jar Test results with PD

SAMPLE	DOSAGE mg/L	EC µS/cm	BOD mg/L	COD mg/L	TURB NTU	TDS mg/L	pH	Temp °C	Coliforms CFU/100ml
PD	5	3187	90	330	198	1584	6.9	26.9	75
PD	10	3069	80	320	226	1532	6.8	28.1	68
PD	12.5	2861	25	310	238	1504	6.9	28.2	65
PD	15	2986	20	270	225	1480	7.6	28.6	36
PD	20	2940	5	220	237	1466	6.8	27.0	3
PD	25	2912	5	190	241	1452	6.8	28.3	7

Table 7: Jar Test result with combination of MI and PD

SAMPLE	DOSAGE mg/L	EC µS/cm	BOD mg/L	COD mg/L	TURB NTU	TDS mg/L	pH	Temp °C	Coliforms CFU/100ml
MI: PD	20:5	2811	50	350	74.9	1418	7.1	25.4	235
MI: PD	15:10	2849	40	280	107	1437	7.3	24.9	250
MI: PD	12.5:12.5	2860	35	270	125	1405	6.9	24.7	60
MI: PD	10:15	2852	35	220	132	1426	7.1	25.0	46
MI: PD	5:20	2800	20	220	151	1416	6.8	25.4	42

Table 8: Jar test results with Alum

SAMPLE	DOSAGE mg/L	EC µS/cm	BOD mg/L	COD mg/L	TURB NTU	TDS mg/L	pH	Temp °C	Coliforms CFU/100ml
Al	5	3100	45	340	54.8	1555	6.9	26.1	125
Al	10	3127	30	240	53.3	1572	6.7	25.9	85
Al	12.5	3026	25	210	44.2	1527	7.5	26.2	29
Al	15	3186	15	200	27.0	1578	7.2	24.5	250
Al	20	3046	10	180	21.6	1560	6.8	27.7	87
Al	25	3153	5	130	24.8	1575	6.5	26.8	5

Table 5 shows that 90% of the Turbidity was removed at a dosage of 25mg/L. 96% of BOD and 99% of Coliforms were removed with a dosage of 25mg/L at a pH of 6.9. Similarly, the electrical conductivity was reduced by 32% at a dosage of 5mg/L, COD by 84% at a dosage of 25mg/L, and TDS by 19% at a dosage of 25mg/L. This result was observed to be consistent with results from similar experiments with other plants as coagulants. Birima *et al* [1] reported 92% turbidity removal efficiency with peanut seeds, Thakur and Choubey [24] reported 91%. The 99% total Coliform removal efficiency is higher than the 75% reported by Nnaji [15] using Garcinia kola. Kalibbala [9] reported an increase in conductivity with the use of moringa as coagulant and as coagulant aid.

Table 6 shows the result of the Jar test using *Phoenix dactylifera* as the coagulant. It had 99% and 96% efficiency in removing total coliforms and BOD respectively at a dosage of 25mg/L but had only 75% efficiency in the removal of Turbidity of the wastewater at a dosage of 5mg/L. 81% of the COD was removed at a dosage of 25mg/L, EC was reduced by 28%, and TDS by 16%. The BOD level was lowered to 5mg/L and the Coliform to 3 CFU/100ml. These values are within the range set by the Drinking Water Quality standards of Nigeria [14].

Table 7 shows the result of the Jar test with *Mangifera indica* and *Phoenix dactylifera* combined in varying dosages as coagulants. Turbidity removal efficiency of 91% was observed at a combined dosage of 20mg/L and 5mg/L of *Mangifera indica* and *Phoenix dactylifera* respectively. 86% BOD removal and 88% total coliforms removal was observed at a combined dosage of 5mg/L *Mangifera indica* and 20mg/L *Phoenix dactylifera.* COD was reduced by 78%, EC by 30%, and TDS by 19%. The reduction in efficiency of the combined seeds is consistent with the observation by Neeraj [13] when he combined chitosan and moringa as coagulants to treat wastewater. The reduced efficiency of the combined seeds can be attributed to coagulant extract deterioration. This occurred possibly due the extract been stored for more than 24 hours at room temperature before it was used [19]. Freshly prepared extracts have been shown to lose their potency when stored at room temperature for more than 24 hours [5, 19]. Another possible explanation for the reduction in their combined efficiency is due to inter particle interaction of the two seeds. When extract of crushed seeds are added to raw water, the proteins produce positive charges acting like magnets and attracting the predominantly negatively charged particles such as clay, silt, bacteria, and other toxic particles in water [5], but in this case, instead of attracting the negatively charged particles from the raw water, the acid–base equilibria was dominated by preferential solvation of the ions by water molecules in the mixtures, forming new compounds [2] and furthering the loss of the potency of the extracts

Table 8 shows the result of the Jar test with Alum as the coagulant. No substantial difference was observed when compared with the biocoagulants with respect to BOD, COD, and total coliforms removal respectively.

4.3 Regression analyses

Tables 9 to 11 show the regression analysis for the dosage with *Mangifera indica* together with the analysis of variance, and Tables 12 to 14 show the regression analysis for the dosage with *Phoenix dactylifera* together with the analysis of variance.

Table 9 shows the regression analysis of the Jar test results with *Mangifera indica* as the coagulant. From the analysis, 98.3% change in dependent variable (Turbidity) of the wastewater can be predicted by the independent variables (Dosage and pH). The model has a strong correlation of 99.14%. Table 10 shows the ANOVA result of the Jar test with *Mangifera indica* as the coagulant. With F statistics of 86.76 and P-value of 0.0022, it shows that the model is statistically significant. Table 11 gives the individual P-values and standard errors of the independent variables.

Table 12 shows the regression analysis of the Jar test results with *Phoenix dactylifera* as the coagulant. From the analysis, 95.6% change in dependent variable (Turbidity) of the wastewater can be predicted by the independent variables (Dosage and pH). The model has a strong correlation of 97.77%. Table 13 shows the ANOVA result of the Jar test with *Phoenix dactylifera* as the coagulant. With F statistics of 14.5 and P-value of 0.06, it shows that the model is statistically significant. Table 14 gives the coefficients of the regression model.

Table 9: Regression Analysis of dosage with Mangifera indica

Multiple R (Correlation)	0.991465857
R Square	0.983004545
Adjusted R Square	0.971674242
Standard Error	7.517190294
Observations	6

Table 10: Analysis of variance for dosage with Mangifera indica

Analysis of Variance					
	Df	*SS*	*MS*	*F*	*Significance F*
Regression	2	9805.168884	4902.584	86.75889	0.00221564
Residual	3	169.5244498	56.50815		
Total	5	9974.693333			

Table 11: Regression Coefficients

	Coefficients	Standard Error	t Stat	P-value
Intercept	623.3719474	113.7205999	5.48161	0.01194
DOSAGE mg/L	-7.821799073	0.705323566	-11.0897	0.001571
pH	-51.46599691	14.62805034	-3.51831	0.038965

Table 12: Regression Analysis of dosage with Phoenix dactylifera

Regression Statistics	
Multiple R	0.977773526
R Square	0.956041069
Adjusted R Square	0.890102672
Standard Error	5.265652456
Observations	6

Table 13: Analysis of variance for dosage with Phoenix dactylifera

ANOVA with Phoenix dactylifera					
	Df	*SS*	*MS*	*F*	*Significance F*
Regression	3	1206.045808	402.0152695	14.49900388	0.065208353
Residual	2	55.45419157	27.72709578		
Total	5	1261.5			

Table 14: Regression coefficients with Phoenix dactylifera

	Coefficients	*Standard Error*	*t Stat*	*P-value*
Intercept	1552.758575	364.6003305	4.258796401	0.050957298
DOSAGE mg/L	-2.978520574	1.344913477	-2.214655906	0.157181951
TDS mg/L	-0.723537873	0.198158586	-3.651307208	0.067501072
pH	-27.89632455	9.807916593	-2.844266087	0.104577759

To treat and dispose of the solids produced from wastewater treatment plants in the most effective manner, it is important to know the characteristics of the solids that will be processed. Some of the characteristics of the wastewater sludge from this research are given in Table 15.

Table 15: Sludge Parameters

Parameters	Results (Mg/L)
Total Nitrogen (N)	8070
Organic Carbon (O.G)	10400
Organic Matter (O.M)	17900
Total Volatile Solids (TVS)	570
Carbon-Nitrogen Ratio	1:29

Nitrogen and phosphorus are the most abundant major plant nutrients in sludge [11, 22]. Sludge that is treated typically contains 1 – 6 percent nitrogen by dry weight [4]. Sludge can be a reliable source of nitrogen and phosphorus which are major nutrients required by plants for proper growth. The nitrogen content of this sludge was observed to be less than 1% (0.8%) which is typical of treated waste [11, 16]. The result of the sludge sample shows low content of organic carbon and organic matter (1.04% organic carbon, 1.7% organic matter). The knowledge of carbon content in wastewater samples is an important element in water monitoring programs. Using Total organic carbon measurements, several compounds with carbon content can be determined. Organic matter present in wastewater can pose a challenge for efficient treatment, as it may cause low coagulation efficiency [8, 28]. Typical content of organic carbon and organic matter in sludge treated with conventional chemicals have been reported to be in the range 10 - 50% [11, 22].

Total volatile solid (TVS) is a water quality measurement obtained from the loss on ignition of total suspended solids. It has great importance in water and wastewater treatment. The greater the concentration of organic or volatile solids, the stronger the wastewater. A test of TVS in sludge is very useful in the design and operation of sludge digesters, vacuum filters and incineration plants [21]. The TVS value obtained for the sludge was found to be lower compared to sludge that has been treated [26].

The amount of nitrogen mineralized is inversely proportional to the carbon to nitrogen ratio (C/N ratio). Soils with large C/N ratios result in low quantities of mineralized nitrogen [11]. A high C/N ratio of sludge ensures that there is limited mobilization of nitrogen by incorporation into cell mass. This in turn makes this nitrogen available at a later period when nitrogen is needed most for plants during the growing period [11, 20]. The C/N ratio is within the range reported in the studies by Mtshadi *et al* [11].

5. CONCLUSION

The research shows the efficacy of combining *Mangifera indica* and *Phoenix dactylifera* seeds powders as coagulants in wastewater treatment at the effective combined ratio (of 20mg/L of *Mangifera indica* with 5mg/L *Phoenix dactylifera*) for turbidity and total coliforms removal. The statistical analyses showed that over 85% of the Turbidity and Total coliforms removed can be attributed to the change in dosage administered to the wastewater. Thus, the use of *Mangifera indica* and *Phoenix dactylifera* seeds powders have great potentials as coagulants and to some extent disinfectants in wastewater treatment.

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