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Research Article

THE USED OF CITRUS SPECIES FRUIT PEEL POWDER AS ECO-FRIENDLY WASTEWATER COAGULANTS

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ABSTRACT
Coagulation and flocculation have emerged as highly effective techniques for eliminating colloidal particles from water and wastewater. This study investigates the viability of dragon fruit peels as coagulants for reducing turbidity, total dissolved solids (TDS), and total suspended solids (TSS) in wastewater. Orange peel powder (OPP) and Lemon Peel powder (LPP) were applied to distillery wastewater at various dosages (ranging from 10 to 90 mg/l) using the standard jar test method, and sedimentation times were meticulously recorded. Turbidity, TDS, and TSS were quantified using appropriate meters. At an optimal dosage of 90 mg/L, OPP successfully reduced 67% of turbidity, 69% of TDS, and 36% of TSS. Meanwhile, LPP, when applied at an optimal dosage of 50 mg/L, reduced 60% of turbidity, 23% of TSS, and 65% of TDS. Although the removal percentages increased with longer sedimentation times, no significant difference was observed between the two peel powders. This study demonstrates that both OPP and LPP hold promise as environmentally friendly coagulants for wastewater treatment.

Keywords: *Distillery Wastewater, Turbidity, TDS, TSS, Coagulation, Fruit Peel*

INTRODUCTION

Coagulation and flocculation techniques are effective methods for the removal of colloidal particles in both water and wastewater. These methods have found widespread application in wastewater treatment processes. They consist of components that facilitate the aggregation and sedimentation of suspended particles within a solution (Nharingo *et al.*, 2015). The introduction of highly charged cations into the water serves to destabilize colloidal particles, resulting in the formation of larger aggregates. Subsequently, these larger aggregates can be efficiently separated through sedimentation, flotation, or filtration units (Mounir *et al.*, 2014).

Coagulants come in two primary categories: natural-based and chemical-based. Natural-based coagulants include materials derived from plants, such as Moringa oleifera seeds, Nirmali seeds, tannin, and Opuntia ficus indica cactus (Vijayaraghavan *et al.*, 2011; Yin, 2010). These natural coagulants are biodegradable, pose no risk to human health, are derived from abundant sources, cost-effective, highly biodegradable, and free from toxic elements (Yin, 2010; Asrafuzzaman *et al.*, 2011; Bratby, 2006).

In contrast, chemical-based coagulants are associated with toxicity and have been linked to certain diseases such as encephalopathy. Compelling evidence has established a connection between aluminum-based chemical coagulants and neurodegenerative conditions like senile dementia and Alzheimer's disease (Okuda *et al.*, 1999; Rondeau *et al.*, 2001; Flaten, 2001). Consequently, there is a growing interest among researchers in exploring the potential of natural plant-derived coagulants as a safer and more environmentally friendly alternative to chemical-based coagulants (Muralimohan *et al.*, 2014).

The Citrus species, a member of the Rutaceae family, thrives in various warm climates, particularly in the Mediterranean countries of the Middle East (Matthaus and Ozcan, 2012). Oranges are primarily consumed as fresh fruit, concentrated juice, or dehydrated slices, with the seeds being a by-product (FAOSTAT). Unfortunately, orange seeds are often considered economically insignificant, leading to their widespread disposal, resulting in environmental problems like greenhouse gas emissions and financial losses for the food industry (Papargyropoulou, *et al.*, 2014). Orange varieties encompass common oranges (Citrus sinensis), navel oranges, blood oranges, and acid less orange, each distinguished by their unique taste, color, texture, and juice yield (Sandhu and Minhas, 2006 and Etebu and Nwauzoma, 2014).

A recent review of published studies on orange seeds has shed light on their bioactive and nutritional properties. These seeds are rich in fatty acids, phytosterols, tocopherols, fibers, minerals, and vitamins (Zayed, *et al.*, 2021 and Malacrida, *et al.,* 2012). The oil content in orange seeds ranges from 34.92% to 41.66% (Samia, *et al.*, 2012 and Reazai, *et al.*, 2014) and is notably high in unsaturated and essential fatty acids, including oleic and linoleic acids. Moreover, orange seeds contain valuable bioactive compounds like carotenoids and flavonoids, which serve as antioxidants, effectively countering the activities of free radicals and potentially offering health benefits (Khokhar and Apenten, 2003 and Farag, *et al.,* 2020). However, orange seeds also contain antinutrients, which can interfere with the absorption of certain essential micro- and macronutrients in food when present in high concentrations (Al Juhaimi, *et al.*, 2018). These antinutrients include oxalate, phytate, saponins, nitrate, and cyanide (Omoruyi, *et al.,* 2007).

Analyzing the physical characteristics of orange seeds and peels evaluating their anti-nutritional and nutrient composition is vital for providing valuable insights to industry stakeholders. This information aids in determining crucial factors to incorporate into the development of equipment for processing orange seeds.

The aim of this research was to investigate characteristics of both Orange peel powder (OPP) and Lemon Peel powder (LPP), focusing on their effective treatment performance. This study also gives a new insight of the potential of OPP and LPP as an environmental friendly coagulant for wastewater treatment.

MATERIALS AND METHODS

Wastewater Collection: The wastewater samples were gathered from Samson's Distilleries private limited, Duggavati Village, Viajayanagara District. This distillery waste water diluted for experimental study.

In-Situ and Ex Situ measurement:

The pH of the wastewater samples was determined in-situ using a portable pH meter. Measurement of Turbidity, TDS, and TSS, Total Dissolved Solids (TDS), and Total Suspended Solids (TSS) were quantified using standard operating procedure. Turbidity was assessed using turbid meter, TDS levels were measured using a TDS meter, and TSS was determined through a vacuum filtration apparatus by referring APHA, 1999 guidelines.

Preparation of OPP and LPP peel as coagulant:

Two types of citrus species fruit peels involved in this study. They are the Orange peel (OP) and Lemon peel (LP). The peels were washed with tap water and subsequently cut into small pieces to facilitate the drying. It was dried in the oven and blended through 250 μm sieved (Romero-cano *et al.*, 2016). Then, 1.0 g dried powder was suspended in 100 mL distilled water and filtered using Whatman 42 filter paper (Prasad, 2009; Vega-rodriguez and Perezcasas, 2003; Shafad *et al.*, 2013). The extract was collected to be used as coagulant.

Coagulation tests:

Coagulation tests were carried out utilizing the Jar test apparatus, maintaining a neutral pH throughout each experiment (Idris *et al.*, 2013), which aligns with the optimal pH for river water (Shafad *et al.*, 2013). Various dosages of the coagulant (ranging from 10 mg/L to 90 mg/L) were introduced into separate beakers to assess the efficacy of dragon fruit peels across a broad spectrum (Shafad *et al.*, 2013). The Jar test apparatus involved rapid mixing at 100 rpm for 4 minutes, followed by slow mixing at 40 rpm for 25 minutes, and concluded with a 30-minute sedimentation step, in accordance with the methodology. Additionally, total dissolved solids (TDS) were measured using a TDS meter and total suspended solids (TSS) were determined using a vacuum filter apparatus, (APHA, 1999).

Data and Statistical analysis

Descriptive statistics were employed to elucidate the data, encompassing metrics like the mean, standard deviation, t-test, and p-value. Subsequently, the results underwent rigorous statistical scrutiny through the utilization of both Paired t-test and Independent t-test methodologies. The Paired t-test facilitated a comparative evaluation of the removal percentages for all parameters before and after the introduction of coagulants. Meanwhile, the Independent t-test was harnessed to assess the removal percentages of parameters in distillery waste water between the two distinct coagulants, OPP and LPP.

Quality Control & Quality Assurance

Procedures were diligently implemented throughout the study. Duplicate measurements were conducted (Shafad *et al.*, 2013). Instrument calibration was carried out prior to measurements, adhering to the manufacturer's recommendations and manual instructions. Maintenance of instruments was performed in strict accordance with specified guidelines. Furthermore, to ensure the integrity of the data, at least one reagent blank was included with each sample set, in alignment with the protocols outlined in APHA, (1999).

RESULTS AND DISCUSSION

In Figure 1, the graph illustrates the effectiveness of OPP and LPP in reducing turbidity, TDS (Total Dissolved Solids), and TSS (Total Suspended Solids) at varying dosage levels (mg/L). The optimal dosage for OPP was determined to be 90 mg/L, resulting in the removal of 67% of turbidity, 69% of TDS, and 36% of TSS. Meanwhile, LPP exhibited its best performance at a dosage of 50 mg/L, achieving a 60% reduction in turbidity and a 23% decrease in TSS. The highest TDS removal by LPP was observed at 90 mg/L, with a 65% removal rate.

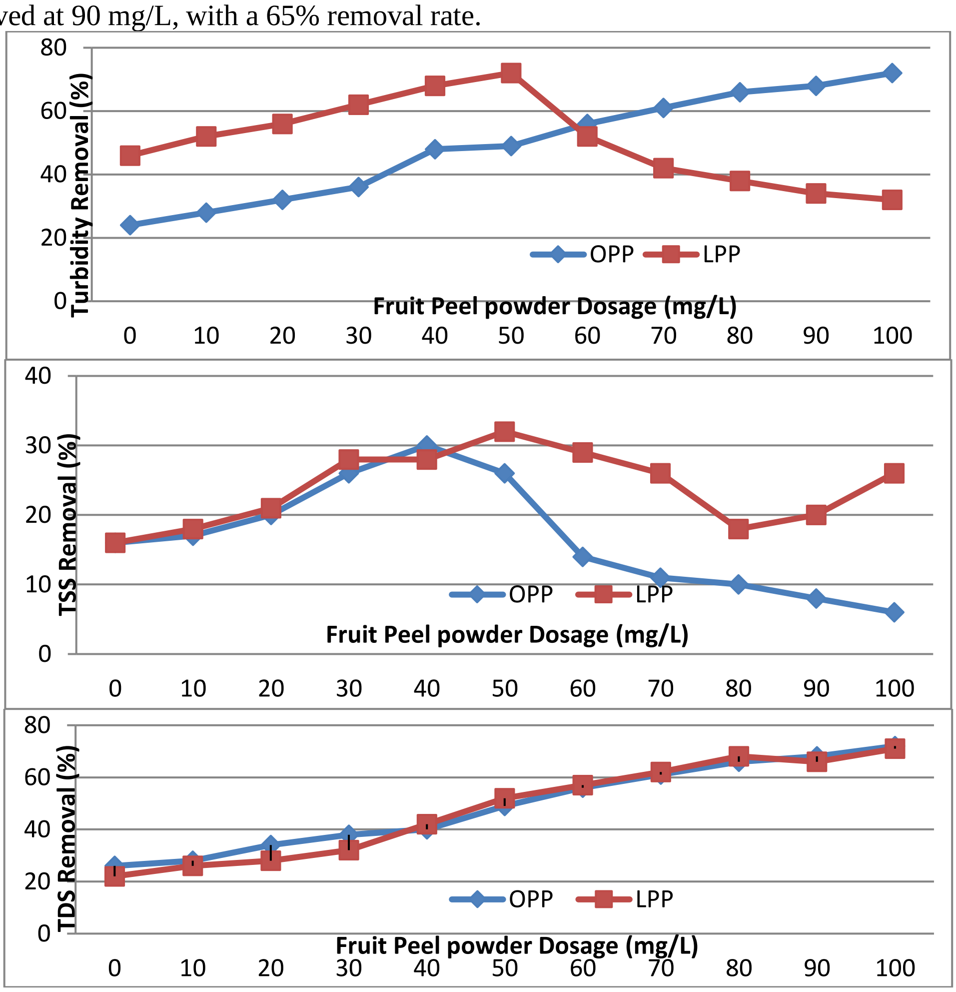

Figure 1: % of removal efficiency of turbidity, TDS and TSS by OPP and LPP dosage (mg/L)

As sedimentation time increased, the removal percentages for all parameters also improved. For instance, the turbidity removal by OPP increased from 38% within 5 minutes to 68% within 60

minutes. A similar trend was noted for LPP, where turbidity removal gradually increased from 35% in 5 minutes to 63% in 60 minutes.
It is worth noting that there was no significant difference in removal percentages between OPP and LPP. On average, both types of peels removed approximately 48.8% (OPP) and 45.2% (LPP) of turbidity. Similarly, the average removal of TDS was 46.2% for OPP and 45.8% for LPP, while the average TSS removal was 14.6% for OPP and 22.6% for LPP. Statistical analysis revealed that the differences in removal percentages between LPP and OPP were not statistically significant for turbidity ($t = -0.420$, $p = 0.686$) and TDS ($t = -0.40$, $p = 0.969$). However, there was a statistically significant difference in TSS removal ($t = 1.678$, $p = 0.132$).

DISCUSSION

The findings of this study underscore the remarkable efficacy of Orange Peel Powder (OPP) in reducing turbidity and Total Dissolved Solids (TDS) to half of their initial concentrations, while Total Suspended Solids (TSS) were reduced by a third. Notably, a previous study (Idris *et al.,* 2013) achieved a higher turbidity removal rate of 95% by using a higher dosage of OPP, ranging from 200-800 mg/L, compared to the results obtained in our research.
The removal of TSS reached 60% with an OPP dosage of 500 mg/L. Furthermore, the mucilage extract of OPP significantly improved particulate-settling rates, surpassing the performance of aluminum sulfate by a remarkable 330% when applied at a concentration of 3 mg/L. It's worth noting that the effectiveness of mucilage extract was equivalent at just 0.3% of the $Al_2(SO_4)^3$ concentration.
In a separate investigation involving Moringa oleifera, commonly known as the horseradish or drumstick tree, a 94% turbidity removal rate was reported with a dosage of 400 mg/L (Sarpong and Richardson, 2010). Moringa oleifera, a member of the Moringaceae family, is native to the sub-Himalayan region of northwest India, Sudan, and other parts of country.
Citrus species fruit peels belonging to the same family as cacti, are rich in pectin a heterogeneous structural polysaccharide (Ridley *et al.,* 2001). The mucilage of Citrus species fruit peels contains galacturonic acids, which serve as the primary active coagulation agent, irrespective of the species involved (Choy *et al.,* 2014). The high presence of natural polymers, such as polysaccharides (particularly galacturonic acid) and proteins in Citrus species fruit peels provides numerous active sites along the polymeric chain for particle adsorption and facilitates the coagulation process (Ridley *et al.,* 2001 and Choy *et al.,* 2014).
Coagulation is the process of reducing or neutralizing the electric charge of suspended particles (Idris *et al.,* 2013). This phenomenon takes place within the colloidal particles in wastewater, where they carry negative electrical charges on their surfaces. To destabilize this stable colloidal system, coagulants with an opposite charge are introduced (Mahvi and Razavi, 2005). Meanwhile, it's worth noting that the colloids in DFPs (Dissolved and Fine Particles) are identified as cationic. Consequently, adsorption and charge neutralization are proposed as potential mechanisms for coagulation, leading to the formation of flocs (Choy *et al.,* 2014; Idris *et al.,* 2013).
In situations where small particles in water carry similar electric charges, they naturally repel each other, thus preventing the aggregation of colloidal particles and maintaining their suspension. Therefore, the coagulation/flocculation process serves to neutralize or diminish the negative charge on these particles (Ebeling *et al.,* 2003), enabling the initial aggregation of colloidal and finely suspended materials. Flocculation, on the other hand, involves the process of bringing together these micro particles to form larger agglomerations, achieved through physical mixing or facilitated by flocculants, such as long-chain polymers (Ebeling *et al.,* 2003). In summary, the formation of flocs is primarily driven by the mechanisms of adsorption and charge neutralization (Choy *et al.,* 2014; Idris *et al.,* 2013), where charge neutralization can result in charge reversal in the event of an overdose of coagulants or flocculants (Mahvi and Razavi, 2005).
The entire treatment process of coagulation-flocculation can be divided into two distinct sequential procedures. The first stage, known as coagulation, involves destabilizing a colloidal suspension or solution. Its purpose is to counteract the factors that maintain the stability of the system. This is achieved through the use of specific chemicals, typically aluminum or iron salts, and referred to as

coagulant agents. The second step, termed flocculation, involves inducing the destabilized particles to come together, make contact, and form larger agglomerates, which can be more easily separated, often through gravity settling (Bratby, 2006). In wastewater, the material carries a negative charge, resulting in electrostatic repulsive forces that hinder particle aggregation, keeping the solution stable and requiring a longer time to sediment. To expedite sedimentation, destabilization is necessary, primarily facilitated by coagulants through mechanisms like adsorption and charge neutralization. Subsequently, flocculation encourages aggregation and floc formation.

Our study also observed a decrease in removal percentages (except for TDS) when the dosage of OPP exceeded the optimum level. This is possibly due to overdosing, which reverses the coagulation process and increases the charge of suspended solids, leading to destabilization (López-Maldonado *et al.,* 2014). Overdosing results in the saturation of polymer bridge sites and destabilizes the particles due to an insufficient number of particles to form inter-particle bridges (Muyibi and Evison, 1995). Studies have been conducted to examine coagulation-flocculation for treating distillery waste water, aiming to optimize the process, including the selection of the most suitable coagulant. Despite the effectiveness of chemical coagulants in treating turbid water, they fall short in terms of green chemistry (Choy *et al.,* 2014).

In addition to the findings of this research, there are several limitations that could be addressed in future studies. For instance, this study did not investigate the exact composition of the coagulant agent in both Citrus species fruit peels (OPP and LPP). As indicated throughout the article, the coagulant agent in fruit peel is likely derived from pectin, a heterogeneous structural polysaccharide residues and natural polymers (Ridley *et al.,* 2001). Future research is recommended to identify these coagulant agents and understand their mechanisms. The study did not account for pH variability in removal efficiency. Future studies should consider pH variation and optimization to determine its relationship with removal efficiency. This study only assessed turbidity, TDS, and TSS, while many other constituents in wastewater require testing. Future research can evaluate the efficiency of citrus species fruit peels (OPP and LPP) in removing other constituents, such as oil and grease.

CONCLUSION

In summary, the OPP demonstrated effective removal rates, eliminating 67% of turbidity, 69% of TDS, and 36% of TSS when applied at the optimal dosage of 90 mg/L. Meanwhile, the LPP exhibited its efficacy by removing 60% of turbidity and 23% of TSS at the recommended dosage of 50 mg/L, and achieving a 65% reduction in turbidity at the ideal dosage of 90 mg/L. Notably, as sedimentation time increased from 5 to 60 minutes, both Citrus species fruit peels (OPP and LPP) exhibited a significant improvement in removal percentages. Significantly, it's worth noting that the removal efficiency for both Citrus species fruit peels was quite similar throughout the study.

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