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ENHANCING THE CLARIFICATION OF AZZABA LANDFILL LEACHATE USING BIOCOAGULANTS WITH OPTIMIZATION BY BOX BEHNKEN DESIGN

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Abstract. This study focuses on optimizing the coagulation-flocculation process for treating pollutants in the leachate from Azzaba landfill, utilizing natural coagulants: walnut shells, Moringa oleifera seeds, and Opuntia ficus-indica leaves. Walnut shells, at pH 4, a coagulant dosage of 12 g/L, 300 rpm stirring speed, and 25 minutes of treatment, demonstrated remarkable reductions in turbidity (83.92 %) and suspended solids (92 %). Opuntia ficus-indica, operating under pH 6, a coagulant dosage of 10 g/L, 300 rpm stirring speed, and 20 minutes of treatment, achieved significant reductions in turbidity (86 %) and suspended solids (90 %). Moringa oleifera, functioning at pH 3, a coagulant dosage of 6 g/L, 300 rpm stirring speed, and 35 minutes of treatment, exhibited substantial decreases in turbidity (91 %) and suspended solids (85 %). Adding lime and starch as flocculants further enhanced treatment efficiency, particularly in turbidity reduction. The Box Behnken design (BBD) optimization highlighted the outstanding effectiveness of coagulants, emphasizing their exceptional performance in turbidity and suspended solids removal. However, maintaining pH stability remains pivotal for optimal results. These findings underscore the efficiency of natural coagulants, especially walnut shells, in leachate treatment, showcasing a promising approach to environmental remediation.

Keywords: landfill leachate, coagulation, flocculation, natural coagulants, box behnken design.

1. Introduction

Landfill leachates, the liquid residues resulting from the percolation process through waste, represent one of the most alarming environmental issues globally. Loaded with toxic substances and pollutants, untreated landfill leachates seriously threaten human health and ecosystem balance[1]. In Algeria, as elsewhere, effective management of leachates has become a major environmental priority, demanding innovative and sustainable approaches. These leachates, saturated with heavy metals, organic compounds, and other pollutants, pose a significant ecological hazard, causing devastating harm to aquatic ecosystems and potable water reserves, as documented in various studies[2]. To address this challenge, various methods for leachate treatment have been developed, ranging from biological approaches to sophisticated physico-chemical treatment technologies. Among these methods, the increasing utilization of coagulation processes is justified by their proven efficiency in eliminating pollutants present in leachates[3]. Coagulation, whether conducted with chemical coagulants or biocoagulants, proves to be an effective method for precipitating pollutants, providing a promising solution in treating these contaminating liquids[4]. Bio-coagulants, derived from natural sources like common walnut shells, Moringa oleifera seeds, and Opuntia ficus-indica leaves, are gaining significant interest due to their availability, reduced cost, and lesser environmental impact[5].

Coagulation, recognized as an essential method in leachates' pollutant elimination, stands out not only for its efficiency in reducing turbidity and suspended solids but also for the advantages offered by natural coagulants. Their easy availability, low cost, environmental friendliness, biodegradability, and harmlessness to human health make them preferred solutions in lixiviate treatment[6]. However, the use of bio-coagulants is not without challenges, notably the variability in their quality based on their sources of origin. Understanding these pros and cons is crucial for the effective implementation of coagulation technologies[7]. The coagulation process involves the introduction of a coagulant to destabilize colloidal particles. Subsequently, flocculation is employed to further increase the particle size, forming larger floccules that settle more readily. This technique typically includes pH adjustment and the addition of ferric/alum

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salts as the coagulant, aiming to counteract the repulsive forces between the particles[8].

Currently, in the realm of landfill leachate treatment, similar to practices in Algeria and many other countries worldwide, inorganic coagulants predominantly based on aluminum or iron find widespread application[9]. Their primary objective revolves around the removal of suspended and colloidal matter, reduction of turbidity, elimination of color, and, in certain scenarios, eradication of microorganisms. Introduction of these coagulants, characterized as trivalent metal salts (such as $Al_2(SO_4)_3$ or $FeCl_3$), into raw leachate triggers the formation of flocs ($Al(OH)_3$ or $Fe(OH)_3$) at neutral or acidic pH levels. These resultant flocs effectively entrap negative particles, facilitating their separation during the sedimentation phase[10]. Despite the established efficacy and cost-effectiveness of inorganic coagulants, their usage often necessitates adjustments in pH and alkalinity[11]. Furthermore, the utilization of aluminum sulfate as a coagulant may lead to issues like overdosing, inefficient utilization, or chemical imbalances in the treatment process, culminating in elevated concentrations of dissolved aluminum in the treated leachate. Additionally, it is imperative to acknowledge that the drawbacks associated with inorganic coagulants can manifest in the resultant sludge, laden with metals like aluminum and iron. Oftentimes, this sludge is stored untreated within landfill leachate treatment facilities, eventually posing environmental hazards after a limited storage period[12]. Given these inherent drawbacks of inorganic coagulants in landfill leachate treatment, recent endeavors have witnessed a surge in studies exploring sustainable and environmentally friendly alternatives. These studies focus on organic coagulants derived from various plant materials, tailored based on availability and treatment efficiency. Examples include Moringa oleifera[13, 14], Cactus[15], Citrus fruit peel[16], and walnut shells[17].

This research aims to assess and compare the performance in eliminating pollutants present in the leachates from the technical landfill center of Azzaba city using three types of coagulants based on common walnut shells, Moringa oleifera seeds, and Opuntia ficus-indica leaves. This assessment is conducted in the laboratory employing liquid stirring equipment such as magnetic stirrers, involving the use of chemicals to adjust pH. The study aims to measure coagulation efficiency concerning turbidity and suspended solids under controlled conditions, including pH, time, agitation speed, as well as coagulant and flocculant concentration.

In the pursuit of enhancing the efficiency of this coagulation process, a Box Behnken experimental design was employed to analyze the influence of three main factors: pH, coagulant dosage, and treatment time. Two primary responses were considered in this endeavor: the percentage of turbidity removal and the percentage reduction in suspended solids. This study aspires to shed essential light on the effectiveness of bio-coagulants in leachate treatment, emphasizing their viability and applicability in the specific context of Azzaba city, Algeria.

2. Materials and methods

2.1. Presentation of Azzaba technical center

Azzaba Technical Center serves as a pivotal hub for waste management in the region. Strategically located, it houses advanced facilities for the treatment and disposal of solid waste. The center's flagship component, the Azzaba Technical Landfill Site, commenced operations in May 2012 and has since played an important role in processing household waste and similar materials. Covering a vast expanse of 17 hectares, the landfill has accumulated leachate over a decade, showcasing its pivotal role in waste management. In 2022, it processed 17,415.5 tons of waste, followed by 17,295 tons in 2023, with an average annual leachate production of 5,776.3 cubic meters. Equipped with state-of-the-art infrastructure, the landfill features two cells, each spanning a hectare and boasting a 70,000 cubic meter capacity. Additionally, it comprises three collection and primary treatment basins, as well as a dedicated station for final treatment employing reverse osmosis technology. Moreover, an on-site laboratory equipped with advanced instruments ensures comprehensive analysis of pollution parameters. The administrative infrastructure further facilitates seamless operations. A weighbridge is utilized to precisely measure incoming waste quantities. The Azzaba Technical Landfill Site is depicted in Fig. 1, underscoring its significance in the region's waste management framework.

2.2. Preparation of coagulants

2.2.1. Walnut shell powder

The dry common walnut shells were processed by shelling and rinsing to remove impurities. After thorough drying at 100 °C for 10 hours, the shells were ground into a fine powder using a robust knife crusher. The resulting powder was sieved to ensure uniform consistency and eliminate large particles. Subsequently, 25 grams of this powder were mixed with 0.5 liters of demineralized water, creating a stock solution with a concentration of 50 g/L. The solution was stirred in a magnetic stirrer at low speed until homogeneous, followed by an increase in speed to 350 rpm for 1 hour to activate the coagulating agents (the preparation of walnut shells is shown in Fig. 2).

Fig. 1. Azzaba technical landfill site

Fig. 2. Steps for preparing coagulant based on common walnut shells

2.2.2. Moringa oleifera seed powder

Ripe, high-quality Moringa oleifera seeds were harvested from a nursery in Oum Thiour, El M'ghair. After shelling, the seeds were dried in an oven at 50 °C for 24 hours and ground into a fine powder using a coffee grinder. 25 grams of this powder were mixed with 0.5 liters of demineralized water, creating a stock solution with a concentration of 50 g/L. The solution was filtered through cloth, stirred in a magnetic stirrer at 100 rpm for 10 minutes, and then agitated at 350 rpm for 50 minutes to activate the coagulating agents (the preparation of Moringa oleifera seed powder is shown in Fig. 3).

Fig. 3. Steps for preparing the coagulant based on Moringa oleifera grains

2.2.3. Opuntia ficus-indica leaf powder

Fresh mature Opuntia ficus-indica leaves were harvested from a farm near Skikda. After cleaning and removing visible spines, the leaves were sliced into small pieces (approximately 1 cm²) and dried in an oven at 40 °C for 3 hours, followed by further drying at 80 °C until completely dry to prevent burning. The dried leaves were ground into a fine powder using a household blender (steps for preparing Opuntia ficus-indica leaf powder are shown in Fig. 4). 25 grams of this powder were mixed with 0.5 liters of demineralized water, creating a stock solution with a concentration of 50 g/L. The solution was stirred in a magnetic stirrer at 100 rpm for 10 minutes and then agitated at 350 rpm for 50 minutes to activate the coagulating agents.



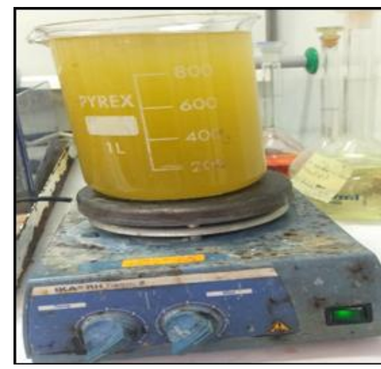

Fig. 4. Steps for preparing the coagulant based on Opuntia ficus-indica leaf powder

2.3. Raw leachate characterization

Leachate samples were collected from the Azzaba Technical Landfill Centre (TLC) site. These leachates exhibited a brown to black color and emitted an unpleasant odor upon initial observation. The physico-chemical analyses, detailed in Table 1, revealed the presence of various mineral and organic substances within the leachate composition. This variability primarily results from biological and physico-chemical alterations occurring in leachate from the TLC. The organic load is relatively high, as evidenced by the elevated values of COD (1630 mg/L) and BOD_5 (385 mg/L). Additionally, the low concentration of dissolved oxygen (0.23 mg/L) indicates microbial activity that might foster an increase in anaerobic populations. The high conductivity (18.41 ms/cm) is attributed to the presence of salts and heavy metals at low concentrations. The pH level is 8.3, suggesting a stable leachate environment, while turbidity and suspended solids are notably elevated due to the presence of undissolved materials. The COD/BOD_5 biodegradability ratio stands at 4.23, indicating a significant portion of non-biodegradable material within the leachate. These parameters pose challenges for conventional treatments, especially aerobic and anaerobic biological pathways, necessitating a physico-chemical treatment step to eliminate turbidity, suspended solids, and color.

Table 1. Physico-chemical analysis of the leachate from Azzaba TLC before treatment

Parameters	Units	Values
pH	–	8.3
Chemical oxygen demand (COD)	mg/L	1630
Biochemical oxygen demand (BOD_5)	mg/L	385
Turbidity	NTU	430
Suspended solids (SS)	mg/L	920
Dissolved oxygen (DO)	mg/L	0.23
Conductivity	ms/cm	18.41
Nitrate NO_3^-	mg/L	0.02
Color	Pt.Co	67.3

2.4. Measurement Equipment and Methods

Turbidity and Suspended Solids measurements are obtained by diluting the sample at ratios of 1:10 and 2:10, respectively. Color is measured by diluting the sample at a ratio of 1:18. COD (Chemical Oxygen Demand) is determined using the DR2800 spectrophotometer with a range of 0–50 ppm at a wavelength of 410 nm. COD is also measured using LANGE LCK 13 mm tubes, LCK714 version, with a range of 100–600 mg/L, usable with the HACH LT200 heating block. BOD5 (Biochemical Oxygen Demand) is measured using the OXITOP WTW apparatus. Dissolved oxygen, pH, and conductivity are measured using the HACH HQ430D oximeter and HACH

HQ440d pH-conductivity meter. Turbidity is measured using the Hach brand Turbidity meter, model 2100AN. Color is determined using the LICO690 color spectrometer with the iodine scale.

3. Results and discussion

3.1. Experimental results

The coagulation-flocculation experiments utilized three natural coagulants: walnut shells, Moringa oleifera seeds, and Opuntia ficus leaves. The experiments were conducted on Cimarec Immono-type stirrers, with each beaker containing 100 mL of crude leachate.

The first stage of the trials involved testing various doses of coagulants to determine the optimal dosage for each coagulant. In the second stage, agitation time was varied from 10 to 35 minutes. The third stage involved adjusting the stirring speed from 100 to 350 rpm. Additionally, pH adjustments were made using 1M sulfuric acid (H_2SO_4) or 1M sodium hydroxide (NaOH), with pH levels monitored after the addition of a few drops. All experiments were conducted within a temperature range of 17 to 20 °C.

To assess the impact of adding a flocculant, lime, and starch were introduced at different doses under varied agitation. The tested samples were transferred to separating ampoules and allowed to settle for 4 hours, facilitating the formation and settling of flocs. The supernatant was then filtered through filter paper and transferred to small glass bottles for subsequent analysis.

3.1.1. Effect of pH

Certainly, the effect of pH is highly significant in the coagulation process. pH plays a crucial role in determining the charge of particles and coagulants in a solution. Most coagulants function effectively within a specific pH range, where they carry the opposite charge to the particles they are trying to neutralize. This charge neutralization process is essential for the coagulants to bind with the particles and form larger flocs, aiding in their settling and removal from the solution[18]. The optimal pH for coagulation varies depending on the type of coagulant being used and the characteristics of the substances in the solution. Deviating from the appropriate pH range can lead to reduced coagulation efficiency, incomplete removal of contaminants, and increased operational costs[19]. Therefore, maintaining the right pH level is crucial to ensuring the success of coagulation processes in various applications, including water treatment, wastewater management, and leachate treatment. Researchers and practitioners carefully consider pH adjustments to achieve the best outcomes when employing coagulation techniques in these contexts. To assess the efficiency of the treatment process, an extensive analysis of coagulation-flocculation operating parameters was conducted, with a specific focus on the leachate's pH level, a critical determinant influencing treatment outcomes[20]. The pH value of the leachate was meticulously adjusted within a range from 2 to 9 through the addition of sulfuric acid for acidification and sodium hydroxide for alkalization. The samples were stirred at 300 rpm, employing various coagulants: walnut shells, Opuntia ficus, and Moringa oleifera, at concentrations of 12 g/L, 10 g/L, and 6 g/L, respectively. This study yielded profound insights into the intricate relationship between pH levels and the efficiency of the coagulation-flocculation process in leachate treatment. The systematic analysis of pH variation's impact on coagulant efficiency was conducted, revealing compelling results. The findings are now summarized in Table 2, illustrating the influence of pH changes on the removal rates of turbidity and suspended solids (SS) by different coagulants.

- *Regarding walnut shells*:

When evaluating the effectiveness of walnut shells in treating landfill leachate, noteworthy results were obtained. At pH 4, removal rates for turbidity and SS reached as high as 83 % and 96 %, respectively, underscoring the efficiency of the process. However, it is important to note that at pH levels exceeding 9, a notable decline in efficiency was observed, with removal rates dropping to 60 %. This highlights the sensitivity of the process to extreme pH values. To further support these findings, the study by Ghaffariraad and Ghanbarzadeh Lak (2021) provides valuable insights. They investigated landfill leachate treatment methods involving coagulation-flocculation with lime and bio-sorption by walnut-shell. Their findings align with the observations made in this study, providing additional context and validation to the presented results[5].

- *For Opuntia ficus:*

In the evaluation of Opuntia ficus performance, it was found that optimal efficiency was attained at pH level of 6. At this pH, significant removal rates were observed for turbidity and suspended solids (SS), with removal rates reaching 86 % and 90 % respectively. This observation emphasizes the critical role of maintaining specific pH levels to ensure effective contaminant removal, highlighting the pH pivotal role in the coagulation process[21].

- *For Moringa oleifera*:

The performance of Moringa oleifera was notably exceptional at pH 3, demonstrating robust efficiency. Under these conditions, remarkable removal rates were achieved for turbidity, suspended solids (SS), and color, reaching 91.86 %, 85.83 % and 68 %, respectively. This underscores the importance of pH optimization in maximizing the effectiveness of natural coagulants in leachate treatment processes. It emphasizes the critical need for precise pH control to fully harness the potential of such natural coagulants[22].

Table 2. Influence of pH on removal rates of turbidity and suspended solids

pH	Treatment effects, %					
	Walnut shells		Opuntia ficus		Moringa oleifera	
	Turbidity	Suspended solids	Turbidity	Suspended solids	Turbidity	Suspended solids
2	79.04	80.03	79.22	88.31	82.22	40
3	58.64	83.20	72.41	70.50	91.86	85.83
4	83	96	55.13	76.31	83.02	62.05
6	73.5	72.5	86	90.3	83.65	80.22
8	64.31	65	36.47	65.02	77.14	75.64
9	55.17	51.5	30.02	62.3	71.27	70.72

3.1.2. Effect of stirring speed

The significance of stirring speed in the coagulation process cannot be overstated. Stirring speed directly influences the formation and characteristics of flocs, pivotal in eliminating impurities from the solution. It affects how coagulant particles collide and adhere to contaminants. The right stirring speed guarantees the even dispersion of coagulants, enhancing their interaction with particles and resulting in the creation of larger, denser flocs. The size of these flocs is critical for efficient settling and subsequent removal from the solution. Excessive stirring speed can prematurely break down flocs, diminishing their ability to capture impurities. Conversely, inadequate stirring might lead to insufficient mixing, causing incomplete coagulation and reduced removal efficiency[23]. To investigate the influence of stirring speed on the removal of turbidity and suspended solids (SS) in landfill leachate, various stirring speeds were tested. Initial experiments at slower speeds yielded unsatisfactory results. Subsequently, a range of fast mixing speeds from 100 to 350 rpm was explored. Remarkably, significant improvements were noted, particularly when using the three selected coagulants. Based on the results in Table 3, the optimal stirring speed was found to be 300 rpm. At this speed, the removal rates for turbidity and SS were notably enhanced. Specifically, the walnut shell coagulant achieved a 69 % turbidity removal and a 58 % SS removal, the opuntia Ficus coagulant demonstrated a 53 % turbidity removal and a 71 % SS removal, and the Moringa oleifera coagulant exhibited a 42 % turbidity removal and an impressive 90 % SS removal. These compelling results can be attributed to the role of agitation in ensuring the uniform dispersion of the coagulant throughout the leachate. Adequate stirring promotes the formation of well-defined flocs, which are crucial for the effective removal of turbidity and SS. The enhanced performance at 300 rpm underscores the importance of appropriate mixing conditions in leachate treatment processes. Moreover, these findings align with previous research on coagulation processes in wastewater treatment, emphasizing the critical nature of optimal stirring speeds for achieving desirable treatment outcomes. Studies by researchers[24] have demonstrated similar trends in the impact of stirring speed on the efficiency of coagulation-based treatment methods, further validating the results obtained in this study.

Additionally, it is worth noting that the effect of stirring speed in the coagulation treatment of leachate has been the subject of numerous studies in the literature. This is attributed to its significant impact on the efficiency of the coagulation process. Many researchers have investigated this aspect extensively, recognizing its importance in achieving optimal treatment outcomes. Consequently, understanding the influence of stirring speed has become a crucial factor in designing effective leachate treatment systems. The varied approaches and findings in these studies underscore the complexity of this parameter and highlight the need for further research to refine our understanding and optimize treatment strategies[3].

Table 3. Effect of stirring speed on removal rates of turbidity and suspended solids

Effect of stirring speed, rpm	Treatment effects, %					
	Walnut Shells		Opuntia ficus		Moringa oleifera	
	Turbidity, %	Suspended solids, %	Turbidity, %	Suspended solids, %	Turbidity, %	Suspended solids, %
100	60	55	32.58	68.27	36.17	87.41
150	50	50.71	33.15	68.65	37.15	87.61
200	61.16	50.66	34.08	69.22	37.80	88.86
250	62.41	52.22	35.11	69.82	40.11	89.14
300	69	58	53	71	42	90
350	61.28	48.16	49.02	68.01	41.20	87.24

3.1.3. Effect of stirring time

Indeed, the effect of stirring time is crucial in coagulation treatment processes. The duration of stirring directly influences the formation and characteristics of flocs, which are essential in removing impurities from the solution. Adequate stirring time allows coagulants to interact effectively with particles, facilitating the formation of larger and denser flocs. Properly formed flocs settle more efficiently and can be readily removed from the solution, leading to clearer and purified water. Insufficient stirring time may result in inadequate mixing, leading to incomplete coagulation and reduced removal efficiency[25]. On the other hand, excessively long stirring can break down flocs prematurely, diminishing their effectiveness in capturing impurities. Finding the right stirring time is essential to achieve efficient coagulation. Careful consideration of stirring time, along with other operational parameters such as coagulant dosage, pH, and flocculant addition, is necessary to optimize the coagulation process. Researchers and practitioners meticulously adjust stirring times to strike the right balance and enhance the effectiveness of coagulation processes in water treatment, wastewater management, and leachate treatment applications[26].

Coagulation-flocculation tests were conducted with three distinct coagulants, and the stirring time was varied from 10 to 35 minutes to assess its influence on treatment outcomes. The objective was to understand how the duration of mixing affects the removal of turbidity and suspended solids (SS). The obtained results, as depicted in Table 4, revealed intriguing insights into the process.

Table 4. Effect of stirring time on removal rates of turbidity and suspended solids

Effect of stirring time, min	Treatment effects, %					
	Walnut Shells		Opuntia Ficus		Moringa Oleifera	
	Turbidity, %	Suspended solids, %	Turbidity, %	Suspended solids, %	Turbidity, %	Suspended solids, %
10	65.17	50	30.24	69.41	38.41	87.80
15	66.21	50.27	51.18	69.88	38.50	88.54
20	67.18	51.17	59.30	70.65	39.02	88.56
25	69.77	58.15	40.28	69.14	40.15	88.70
30	66.42	56.44	35.17	68.55	41.33	89.01
35	65.41	54.12	25.41	68.11	45.22	89.57

- *Walnut shells coagulant*

Similarly, the coagulation process employing walnut shells showcased its highest efficiency at 25 minutes of stirring. During this period, a significant reduction in turbidity by 69.77 % and a notable decrease in suspended solids by 58.15 % were recorded. Beyond the 25-minute threshold, diminishing returns were observed due to disturbed mixing.

- *Opuntia ficus coagulant*

For the Opuntia ficus coagulant, the removal rates for turbidity and SS displayed a consistent increase from 10 to 30 minutes of stirring. Remarkably, optimal results were achieved at 20 minutes, indicating a substantial reduction in turbidity by 59.30 % and a remarkable 70.65 % decrease in suspended solids. Beyond this optimal mixing time, disturbances in the mixing process became evident, leading to an escalation in both turbidity values and SS content.

- *Moringa oleifera coagulant*

The coagulation-flocculation process utilizing Moringa oleifera demonstrated its optimal performance at 35 minutes of stirring. At this duration, a considerable reduction in turbidity by 45.22 % and an impressive 89.57 % decrease in mixed liquor suspended solids (SS) were achieved. However, extending the stirring time further adversely affected the process, leading to an increase in both turbidity values and SS content.

3.1.4. Effect of coagulants dose

Indeed, the dosage of coagulants plays a pivotal role in the coagulation process. The right dosage ensures the efficient removal of contaminants, including turbidity, suspended solids, and other impurities, from water or wastewater. It's essential to find the balance: too little coagulant might not effectively neutralize the charged particles, whereas an excess can lead to wastage and increased chemical residues in the treated water. Consequently, finding the optimal dosage is critical for achieving effective and economical coagulation[27].

To evaluate the effectiveness of three natural coagulants: walnut shells, Opuntia ficus, and Moringa oleifera in treating Azzaba TLC, extensive coagulation trials were conducted. The objective was to determine the optimal dosage for each coagulant, a critical parameter influenced by various factors, including the coagulant type and its chemical composition, encompassing suspended solids and ions. Precise dosage tests for Opuntia ficus were conducted, ranging from 6 g/L to 14 g/L. The significant impact of these dosages on the removal of key parameters such as turbidity, suspended solids, and color is illustrated in Table 5. The findings revealed that optimal turbidity and suspended solids removal were achieved at specific dosages:

- Walnut shells (12 g/L): This dosage resulted in remarkable reductions, with a 56.52 % decrease in turbidity and a 66.97 % decline in suspended solids. Furthermore, there was a noticeable increase in dissolved oxygen concentration, accompanied by a decrease in conductivity.
- Opuntia ficus (10 g/L): This dosage yielded significant results, leading to a 42.09 % reduction in turbidity and an impressive 89.13 % removal of suspended solids. The treatment also resulted in an increase in dissolved oxygen concentration and a decrease in conductivity.
- Moringa oleifera (6 g/L): At this dosage, turbidity was reduced by 49 %, and suspended solids decreased by 70 %. Moreover, there was a notable increase in dissolved oxygen concentration, along with a decrease in conductivity. These outcomes can be explained through the action of cations found in walnut shells, Moringa oleifera, and Opuntia ficus, including potassium, calcium, magnesium, sodium, and iron[28]. When introduced into the leachate, these coagulants neutralize negatively charged colloids, leading to their destabilization. Additionally, Opuntia ficus contains crude proteins and vitamin K, known for their blood clotting properties. Moringa oleifera seeds contain a soluble cationic protein that efficiently reduces turbidity in treated water[13]. These inherent properties of natural coagulants underscore their effectiveness in eliminating turbidity and suspended solids from leachates.

Table 5. Effect of coagulants dose on removal rates of turbidity, suspended solids and color by the three coagulants

Coagulants dose, g/L	Treatment effects, %								
	Walnut Shells			Opuntia Ficus			Moringa Oleifera		
	Turbidity, %	Suspended solids, %	Color, %	Turbidity, %	Suspended solids, %	Color, %	Turbidity, %	Suspended solids, %	Color, %
6	30	30.20	17.25	14	25	30.12	49	70	73.21
8	35	33	25.64	10	21	45.01	45	68.22	70.14
10	38	35.21	45.21	42.09	89.13	78.41	30.21	54.01	66.21
12	56.52	66.97	62	17	30	78	28.15	47.12	64.12
14	26.17	30.17	60	12	27	76.25	25.12	28.02	60.15

3.1.5. The role of flocculants in the treatment

Certainly, the pivotal role of flocculants cannot be overstated in the coagulation process. These specialized agents play a fundamental role in clumping together the destabilized particles or colloids, neutralized earlier by coagulants, into larger, denser flocs. Once these flocs are formed, they settle more efficiently and can be easily removed from the solution. This essential process leads to water that is not only clearer but also purified[4]. Flocculants act as catalysts, ensuring the creation of well-structured flocs, vital for effectively removing suspended solids, turbidity, and other impurities. The careful selection of the appropriate flocculant, combined with precise dosing, is crucial for achieving optimal coagulation and flocculation results. Furthermore, flocculants play a pivotal role in enhancing the clarity and overall quality of treated water. Their presence prevents the settled impurities from re-dispersing in the treated water, thus maintaining the desired water quality standards. In summary, the meticulous choice and application of flocculants significantly elevate the efficiency of the coagulation process. Their indispensable role is evident in a wide array of applications, including water treatment, wastewater management, and leachate treatment[29].

In this study, the efficacy of landfill leachate treatment was explored using three natural coagulants: walnut shells, Opuntia Ficus, and Moringa oleifera. Additionally, the impact of pH variation was investigated to optimize the treatment process. Specifically, the study focused on the role of flocculants in the coagulation-flocculation treatment method. Two types of natural flocculants were utilized: lime (a mineral-based flocculant) and starch (an organic-based flocculant).

Lime (Calcium Hydroxide, $Ca(OH)_2$): Lime was used as a natural flocculant due to its ability to neutralize the acidity of water and facilitate the precipitation of impurities. The tests were conducted using different concentrations of lime to determine the optimal dose[30].

Starch: Starch, a natural polysaccharide derived from corn, wheat, or potato, was employed as another natural flocculant. Its flocculating properties stem from its chemical structure, allowing it to form bonds with suspended particles[31].

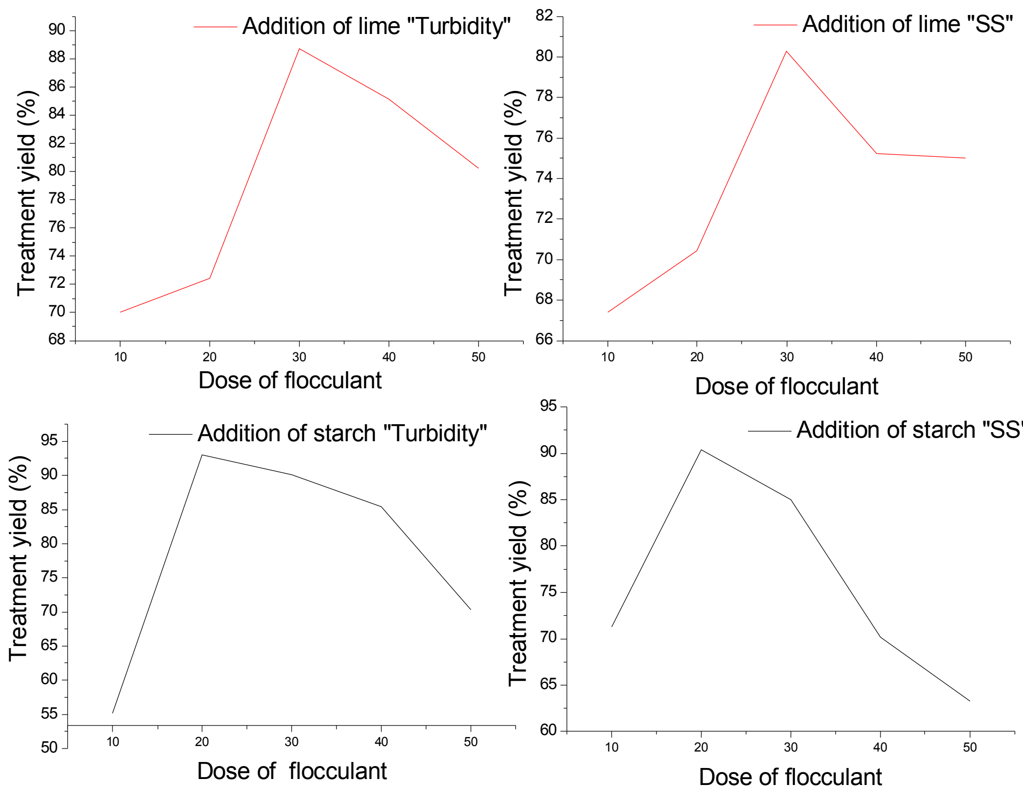

Fig. 5. Effect of flocculant type for coagulant (Walnut Shells)

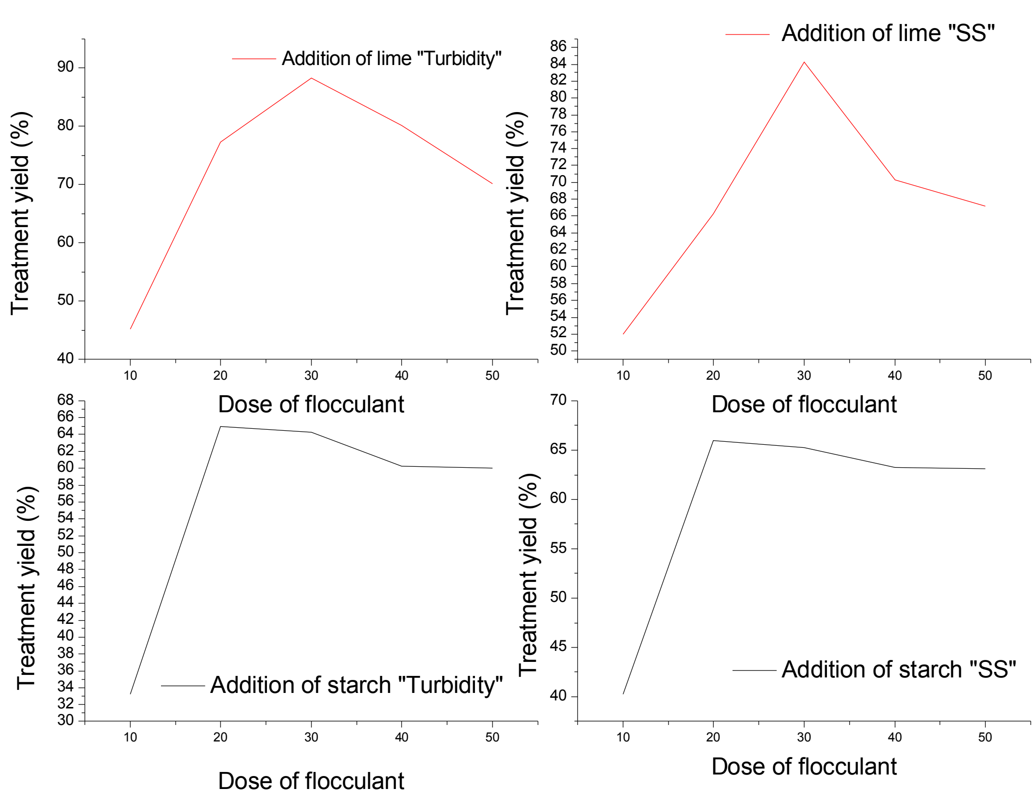

Fig. 6. Effect of flocculant type for coagulant (Opuntia Ficus)

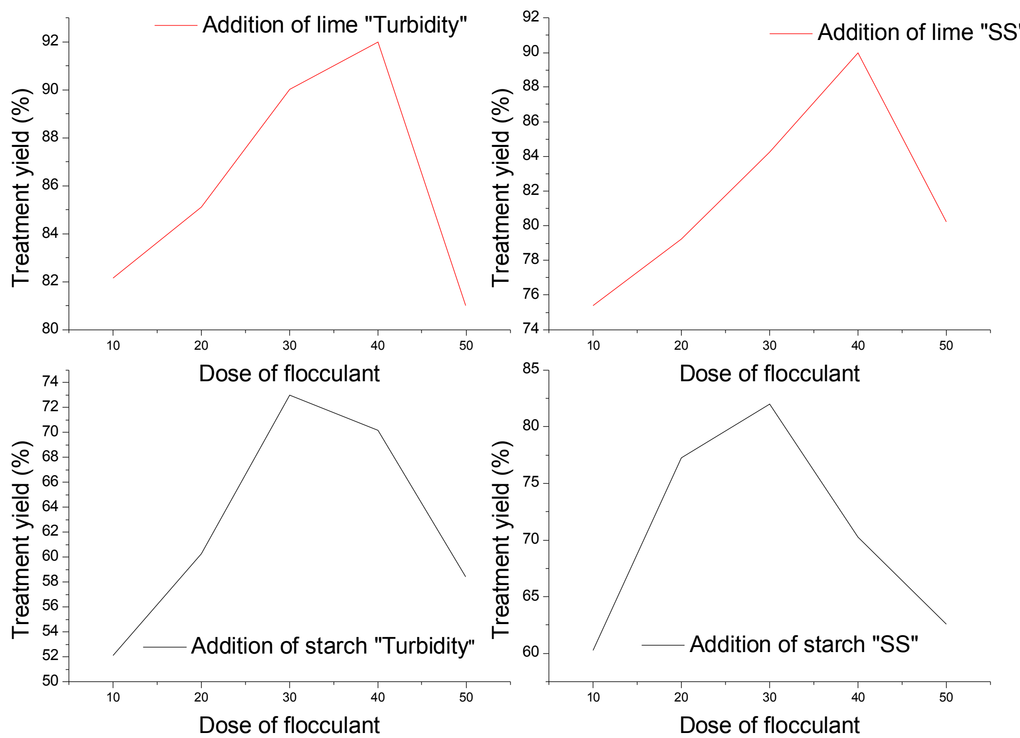

Fig. 7. Effect of flocculant type for coagulant (Moringa Oleifera)

The experimental setup involved varying doses of lime and starch in the leachate mixtures. The results were analyzed through histograms (Figs. 5–7) to visualize the reduction in color, turbidity, and suspended solids in response to different flocculant concentrations. In the conducted experiments, the optimal doses for various coagulants and flocculants were determined to enhance the efficiency of the treatment process. For Walnut Shells and Opuntia Ficus, an optimal lime dose of 30 g/L resulted in significant turbidity reduction, reaching 88.70 % for walnut shells and 88.2 % for Opuntia Ficus. Additionally, an optimal starch dose of 20 g/L led to substantial reductions in suspended solids, achieving 93 % and 65 % reduction for walnut shells and Opuntia Ficus, respectively. In the case of Moringa Oleifera, the optimal lime dose was found to be 40 g/L, resulting in a remarkable turbidity reduction of 92 % and suspended solids reduction of 90 %. Moreover, an optimal starch dose of 30 g/L led to notable reductions, achieving a turbidity reduction of 73 % and suspended solids reduction of 82 %. These findings underline the significance of natural flocculants in landfill leachate treatment. Lime and starch, derived from natural sources, proved effective in reducing turbidity and suspended solids. The study provides valuable insights into eco-friendly approaches for leachate treatment, emphasizing the potential of plant-based flocculants in environmental remediation efforts.

3.2. Modeling results by Box Behnken design

The Box Behnken design stands as a cornerstone in statistical experimentation, particularly in unraveling the intricate relationships among multiple variables. In both scientific research and industrial experiments, the exhaustive testing of all possible variable combinations is often infeasible due to limited resources. This is where the Box-Behnken design steps in, offering an efficient and systematic method to explore the response surface within a confined number of experiments[32]. In the context of our study, we harnessed the power of the Box-Behnken design to decipher the complex interplay of factors influencing the desired outcomes. By carefully selecting specific levels of each factor, this design enabled the creation of a balanced set of experimental runs. These runs provided deep insights into the main effects and interactions among the variables. Utilizing this approach, researchers can establish a response surface model, frequently in the form of a polynomial equation, through regression analysis. This model quantifies the relationship between input variables and the response variable, paving the way for accurate predictions and optimizations[33]. The Box-Behnken design offers a dual advantage: not only does it streamline the number of experimental runs, but it also excels in pinpointing the optimal conditions for desired outcomes. Researchers can systematically manipulate factors of interest, exploring their combined effects and deducing the optimal settings leading to the desired response. Furthermore, this design excels in detecting potential interactions between factors, providing invaluable insights into the underlying complexity of the studied system[34].

Table 6. Values of variables tested to obtain optimum value

Factors	Values		
	–1	0	+1
pH	2	5.5	9
Coagulant dosage, g/L	6	10	14
Stirring time (min)	10	22.5	35

The objective of this section is to delve into the modeling and optimization using the Box – Behnken design in the treatment of leachate. We focus on two key responses: turbidity removal efficiency (Y1) and suspended solids percentage (Y2). Three factors: pH, coagulant dose, and stirring time were examined across three distinct coagulants: Walnut shells (C1), Opuntia ficus (C2), and Moringa oleifera (C3). These investigations were conducted within the framework of the coagulation process, offering a comprehensive understanding of the interplay between factors and responses in the treatment of leachate. Values of each variable tested in this experiment were conducted in one replicate, as indicated in Table 6.

3.2.1. Study of the effect of variables using the experimental design methodology

The purpose of this section is to investigate the leachate treatment through coagulation with three coagulants (Walnut shells (C1), Opuntia Ficus (C2), Moringa Oleifera (C3)), using the coagulation process. A Box – Behnken experimental design was applied to optimize the coagulation parameters (pH, coagulant dosage, and treatment time). The turbidity removal efficiency (Y1) and suspended solids removal efficiency (Y2) were used as responses. The results of the experimental design were studied and interpreted using MINITAB 17 software to estimate the response of the dependent variable across all experiments. The outcomes from the Box-Behnken (BBD) design experiments are presented in Table 7, illustrating the statistical combinations of the independent variables: pH, coagulant dosage, and treatment time, along with the yields Y1 (%) and Y2 (%). Based on these results, empirical relationships between the responses and independent variables were identified, leading to the formulation of second-order polynomial multiple regression equations.

- **Coagulant 1**

$$\text{Turbidity (\%)} = 83.00 + 2.12\,pH + 1.50\,Dose\ coagulant1 + 2.63\,Stirring\ time - 23.75\,pH \times pH - 3.00\,Dose\ coagulant1 \times Dose\ coagulant1 - 1.25\,pH \times Dose\ coagulant1 + 1\,pH \times Stirring\ time + 1.75 \quad (1)$$

$$\text{SS (\%)} = 96.00 + 2.75\,pH - 6.63\,Dose\ coagulant1 - 6.62\,Stirring\ time - 9.13\,pH \times pH - 9.38\,Dose\ coagulant1 \times Dose\ coagulant1 - 13.87\,Stirring\ time \times Stirring\ time - 2\,pH \times Dose\ coagulant1 \quad (2)$$

- **Coagulant 2**

$$\text{Turbidity (\%)} = 92.00 + 1.625\,pH + 2.375\,Dose\ coagulant2 + 4.250\,Stirring\ time - 26.13\,pH \times pH - 1.62\,Dose\ coagulant2 \times Dose\ coagulant2 - 11.87\,Stirring\ time \times Stirring\ time - 0.25\,pH \quad (3)$$

$$\text{SS (\%)} = 86.00 + 2.38\,pH + 3.88\,Dose\ coagulant2 + 5.25\,Stirring\ time - 36.25\,pH \times pH - 0.25\,Dose\ coagulant2 \times Dose\ coagulant2 - 11.50\,Stirring\ time \times Stirring\ time + 0.50\,pH \times Dose\ coagulant2 \quad (4)$$

- **Coagulant 3**

$$\text{Turbidity (\%)} = 49.00 - 4.13pH - 6.25\,Dose\ coagulant3 + 3.37\,Stirring\ time + 15.62\,pH \times pH + 8.38\,Dose\ coagulant3 \times Dose\ coagulant3 + 0.13\,Stirring\ time \times Stirring\ time + 0.50\,pH \times Dose\ coagulant3 \quad (5)$$

$$\text{SS (\%)} = 75.00 - 2.00\,pH - 5.75\,Dose\ coagulant3 - 2.25\,Stirring\ time + 10\,pH \times pH - 0.00\,Dose\ coagulant3 \times Dose\ coagulant3 + 0.00\,Stirring\ time \times Stirring\ time - 1pH \times Dose\ coagulant3 \quad (6)$$

The determination coefficients R^2 represent the percentage of variation in the response explained by its relationship with one or more predictor variables. This value is calculated as the ratio of the sum of squares of the model to the total sum of squares. The determination coefficient R^2 always ranges between 0 and 1 and should ideally be above 0.8 for a reasonable model. The model is considered well-fitted to the data if R^2 is high (relative relationship)[35, 36]. The higher value of R^2 is important. Experimental design, data analysis, and plotting of main effects and interactions were performed with MINITAB 17 statistical software[37]. The validation of the mathematical model requires comparing the model's Fisher F-values and lack-of-fit values with critical values obtained from the Fisher table for a given risk[38]. The results obtained indicated that the model is representative and confirms the strong correlation between degradation (response Y) and the selected factors for Coagulant 1 (R^2 of Y1 (96.07 %), R^2 of Y2 (91.61 %)), for Coagulant 2 (R^2 of Y1 (99.21 %), R^2 of Y2 (99.05 %)), and for Coagulant 3 (R^2 of Y1 (89.28 %), R^2 of Y2 (83.75 %)). The analysis of experimental data was conducted using Minitab 17 software, and the statistical validation of the model was performed by ANOVA with a 95 % confidence level.

Table 7. Experimental matrix of the three-factor Box – Behnken design (coded values)

Coagulant 1						Coagulant 2		Coagulant 3	
Experiences	pH	Dose of coagulant	Stirring time, min	C1 (Y1)	C1 (Y2)	C2 (Y1)	C2 (Y2)	C3 (Y1)	C3 (Y2)
1	–1	–	0	53	86	60	43	86	96
2	+1	–	0	60	90	64	47	80	93
3	–1	+	0	55	69	65	51	65	79
4	+1	+	0	57	65	68	57	61	72
5	–1	0	–	45	75	50	32	66	91
6	+1	0	–	50	87	53	39	63	87
7	–1	0	+	45	60	55	40	75	82
8	+1	0	+	51	70	58	42	55	80
9	0	–	–	62	74	70	62	50	77
10	0	+	–	65	82	75	71	52	74
11	0	–	+	70	77	82	80	70	77
12	0	+	+	80	58	87	84	58	72
13	0	0	0	83	96	92	86	49	75
14	0	0	0	83	96	92	86	49	75
15	0	0	0	83	96	92	86	49	75

Table 8. Analysis of variance for responses of three coagulants

	DF	C1				C2				C3			
		Y1		Y2		Y1		Y2		Y1		Y2	
		F-Value	*p*-Value	*F*-Value	*p*-Value	*F*-Value	*p*-Value	*F*-Value	*p*-Value	*F*-Value	*p*-Value	*F*-Value	*p*-Value
Source													
Model	9	13.58	0.005	6.07	0.031	69.52	0.000	58.12	0.000	4.43	0.053	2.86	0.130
Linear	3	1.77	0.249	4.49	0.035	14.19	0.007	12.07	0.01	4.25	0.077	4.04	0.083
pH	1	1.76	0.242	1.55	0.569	4.27	0.094	4.24	0.095	3.21	0.133	1.15	0.332
Dose coagulant	1	0.88	0.392	8.97	0.030	9.12	0.029	11.28	0.020	7.38	0.042	9.51	0.027
Stirring time	1	2.68	0.162	8.97	0.015	29.19	0.003	20.7	0.006	2.15	0.202	1.46	0.281
Square	3	38.60	0.001	10.01	0.038	194.34	0.000	161.85	0.000	8.67	0.006	4.48	0.070
Correlation coefficient of responses, %		$R^2 = 96.07$		$R^2 = 91.61$		$R^2 = 99.21$		$R^2 = 99.05$		$R^2 = 89.28$		$R^2 = 83.75$	

3.2.2. ANOVA for coagulants

To determine statistical significance, we analyze the *p*-values, which indicate the certainty level compared to a significance threshold for each drug ($\alpha = 0.05$). If $p \le \alpha$, the model explains the variation in the response; if $p > \alpha$, it does not. Lower *p*-values indicate higher correlation with the corresponding coefficient[39].

In Table 8, linear effects for coagulants 1 and 3 (Y1: $p = 0.249$, $p = 0.077$, Y2: $p = 0.083$) exceed α, suggesting a non-significant correlation. Conversely, for coagulants 1 (Y2: $p = 0.035$) and 2 (Y1: $p = 0.007$, Y2: $p = 0.01$), linear terms are significant.

- **Main effects of coagulants**

In analyzing the main effects of the coagulants, it is evident that Coagulant 1's efficiency in turbidity removal (Y1) is significantly influenced by pH and treatment time, while coagulant dosage does not play a significant role. For suspended solids removal (Y2), only pH has a significant impact for Coagulant 1. Similarly, for Coagulant 2, pH and time are significant factors affecting both Y1 and Y2, with coagulant dosage not showing a significant effect. Coagulant 3 demonstrates a significant pH effect on both Y1 and Y2, while coagulant dosage and treatment time do not exhibit significant impacts on the removal processes.

- **Interactions between factors**

The study examined interactions among pH, coagulant dosage, and treatment time for each coagulant and response. In most cases, these interactions were not significant, indicating that the influence of each factor on the responses (Y1 and Y2) is largely independent of others. pH emerged as the most influential parameter for both responses across all coagulants. Adjusting the treatment pH can substantially impact coagulation process efficiency. Coagulant dosage was significant only for Y1 with Coagulant 1, suggesting its adjustment can influence turbidity removal in this specific scenario. Treatment time showed no significant impact on responses for most coagulants and parameters, indicating its potential for optimization without compromising process efficiency. Overall, the limited significance of interactions among factors (pH, coagulant dosage, time) emphasizes the predominant individual effects of these factors. This underscores the importance of their independent adjustment to enhance treatment efficiency.

The Fisher test is a statistical hypothesis test used to assess the equality of two variances by comparing the ratio of the two variances to a specific theoretical value found in the Fisher table. In the case of multiple variables, the observed test statistic is compared to the critical value (tabulated *F*). Based on our results shown in the ANOVA (Table 8), we can confirm the reliability and significance of the model represented by the F value, indicating the linearity and significance of the model. 40 From these results, several significant conclusions can be drawn regarding the effectiveness of the three coagulants (C1, C2, C3) in leachate treatment, considering responses Y1 (turbidity removal percentage) and Y2 (suspended solids removal percentage), as well as pH and treatment duration as the main study factors.

In evaluating the effectiveness of three coagulants (Coagulant 1, Coagulant 2, and Coagulant 3) in treating leachate, significant findings emerged.

Coagulant 1 demonstrated a considerable impact on turbidity reduction (Y1), with an *F* value of 13.58, surpassing the critical *F* value of 3.02, indicating a major role in decreasing leachate turbidity. Additionally, its effect on suspended solids removal (Y2) was significant, evident from the *F* value of 6.07 exceeding the critical threshold. Coagulant 2, on the other hand, exhibited exceptional effectiveness. Its *F* value for Y1, 69.52, greatly surpassed the critical *F* value, highlighting its highly significant role in reducing leachate turbidity. Similarly, for Y2, the *F* value of 58.12 exceeded the critical threshold, emphasizing the remarkable impact of Coagulant 2 on suspended solids removal. Coagulant 3, while showing a lower *F* value for both Y1 (4.43) and Y2 (2.86), still demonstrated significance. Its effect on turbidity reduction was notable, although not as pronounced as Coagulants 1 and 2. The impact on suspended solids removal, being less significant, remained noteworthy.

These results underscore the varying effectiveness of the coagulants, with Coagulant 2 being notably powerful in reducing both turbidity and suspended solids in leachate, followed by Coagulant 1 and Coagulant 3, each contributing significantly to the treatment process.

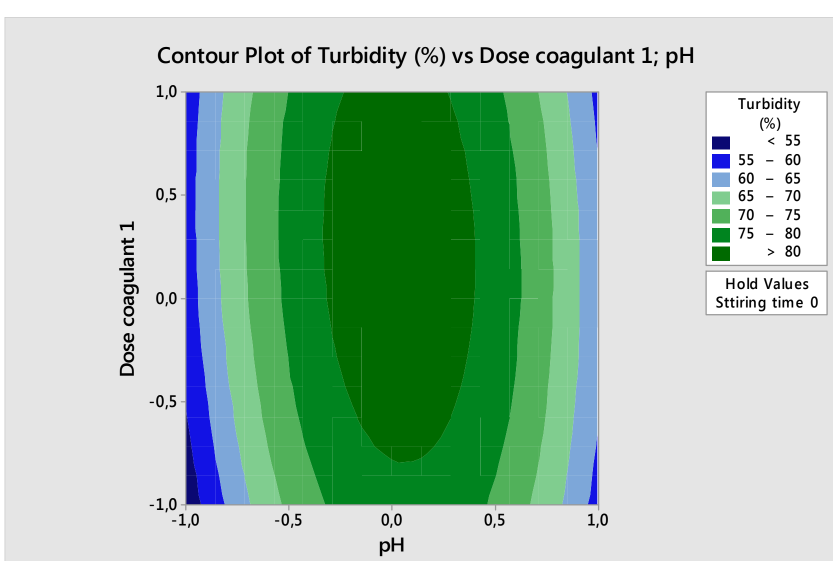

Fig. 8. Contour Plot for Coagulant (Walnut Shells), % turbidity

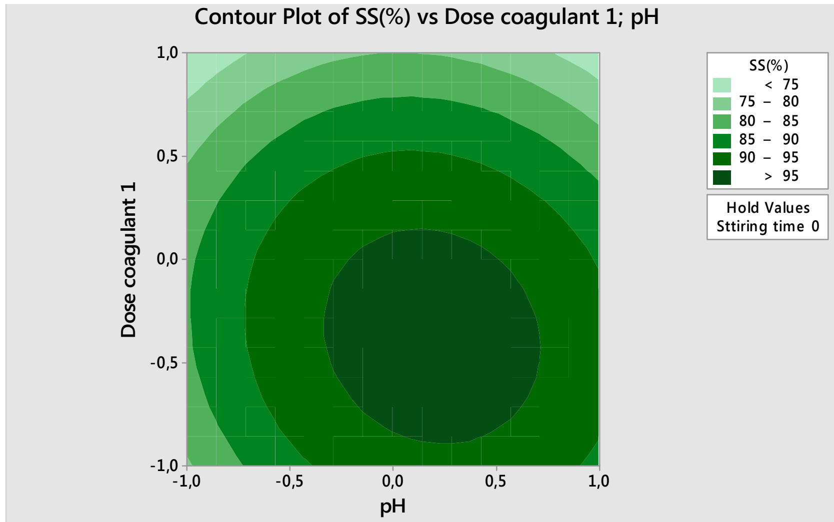

Fig. 9. Contour Plot for Coagulant (Walnut Shells), % SS

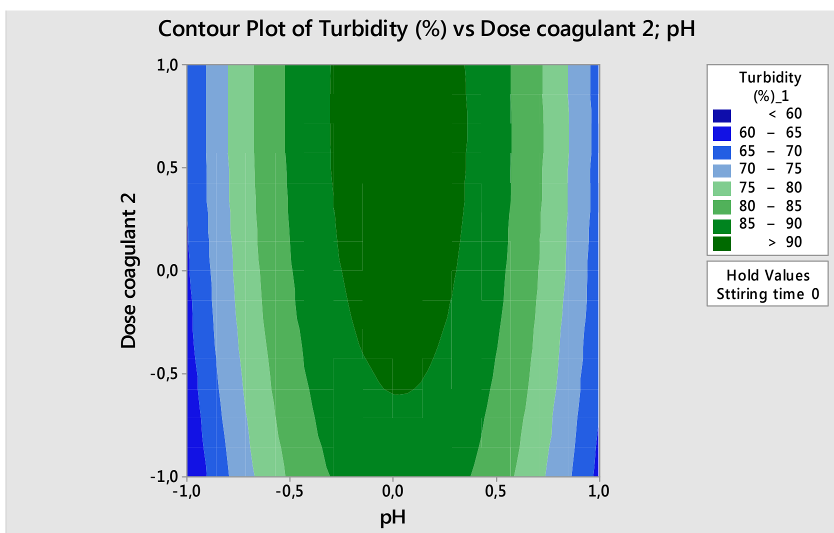

Fig. 10. Contour Plot for Coagulant (Opuntia Ficus), % turbidity

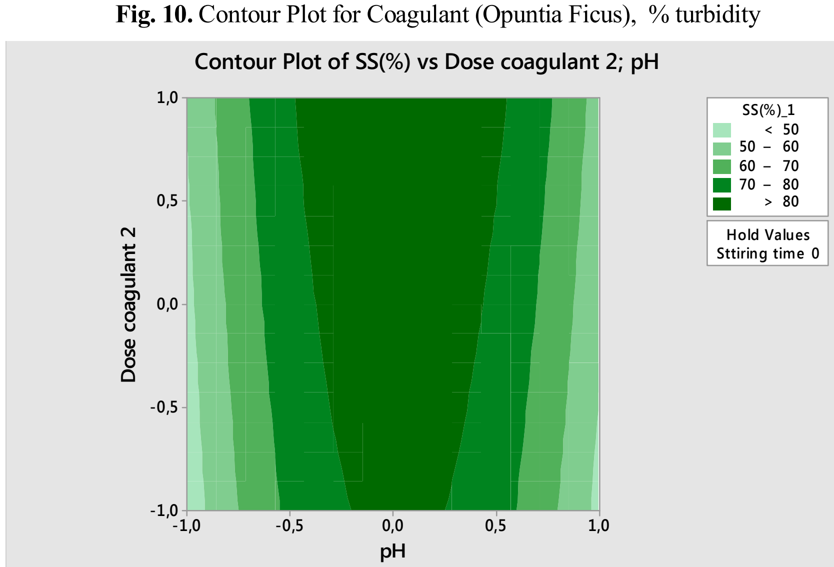

Fig. 11. Contour Plot for Coagulant (Opuntia Ficus), % SS

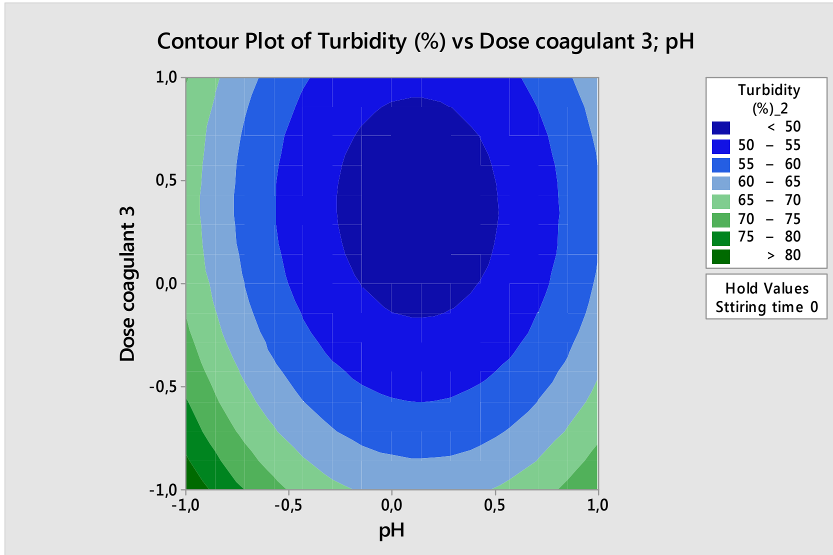

Fig. 12. Contour Plot for Coagulant (Moringa Oleifera), % turbidity

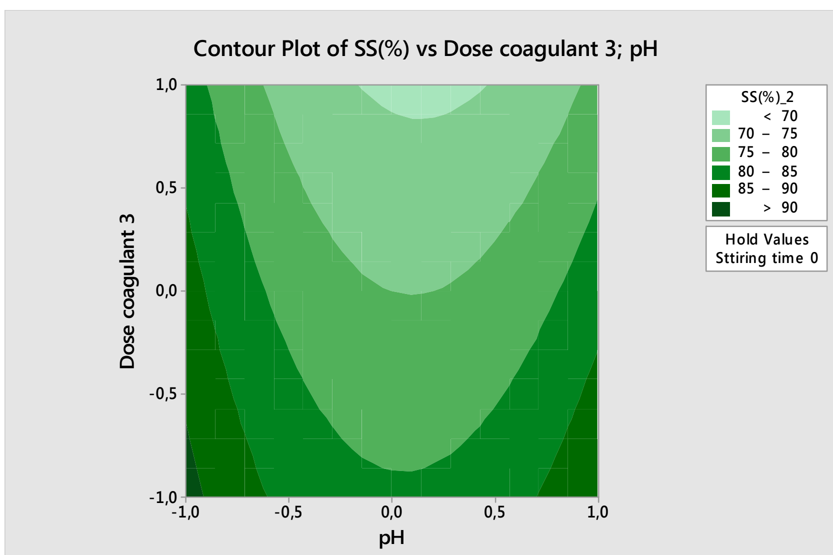

Fig. 13. Contour Plot for Coagulant (Moringa Oleifera), % SS

3.2.3. Contour plot

Contour plots, also referred to as contour maps or charts, stand as powerful tools extensively utilized across diverse scientific and engineering disciplines. These graphical representations transform intricate three-dimensional data into a comprehensible two-dimensional format, offering a lucid and intuitive means to dissect complex relationships among multiple variables. In our research, contour plots play a pivotal role by visually illuminating the interdependencies between various factors and the responses we investigated: turbidity removal efficiency and suspended solids percentage. Within our study, contour plots have been indispensable in elucidating the impact of three distinct coagulants: Walnut shells (C1), Opuntia Ficus (C2), and Moringa Oleifera (C3), on the responses under scrutiny. Figs. 8–13 showcase these contour plots, providing a comprehensive overview of how fluctuations in coagulant dosage and pH levels influence both turbidity removal efficiency and suspended solids percentage. It's noteworthy that the stirring time was consistently maintained at a medium level (0) throughout our experiments. This deliberate choice allowed us to meticulously isolate and analyze the specific effects of coagulant dosage and pH on the responses of interest. Each plot in Figs. 8–13 represents a unique combination of coagulant type and response variable, offering an intricate visualization of the intricate data emanating from our experiments. Through these contour plots, we discern discernible patterns, subtle trends, and areas ripe for optimization. These visualizations significantly augment our overall analysis, enriching our understanding of the experimental data.

The contour plots, as depicted in Figs. 8–13, provide critical insights into the optimal conditions for turbidity removal efficiency and suspended solids percentage for the three coagulants: Walnut shells (C1), Opuntia Ficus (C2), and Moringa Oleifera (C3). The contour plots allow us to discern specific trends and patterns, guiding us toward the most effective combinations of pH levels and coagulant dosages to achieve superior results in the treatment of leachate.

- *For Coagulant 1:*

The contour plots reveal that the most effective turbidity removal (>80 %) occurs within the pH range of –0.5 to 0.5 and a coagulant dosage range of –0.75 to 1. Similarly, for suspended solids removal (>95 %), the optimal conditions lie between pH –0.7 to 0.7 and a coagulant dosage of –0.8 to 0.

- *For Coagulant 2:*

Coagulant 2 demonstrates exceptional performance in turbidity removal (>90 %) within a narrow pH range of –0.5 to 0.45 and a coagulant dosage range of –0.5 to 1. Additionally, for suspended solids removal (>80 %), the ideal conditions are found within the pH range of –0.5 to 0.5 and a coagulant dosage of –1 to 1.

- *For Coagulant 3:*

Coagulant 3 exhibits optimal turbidity removal (>80 %) between pH –0.3 and 0.5 and a coagulant dosage range of 0 to 0.8. Moreover, for suspended solids removal (>90 %), the contour plots indicate that the best results are achieved at pH levels between –1 and –0.9 and a coagulant dosage between –1 and –0.7.

The findings from these contour plots have significant practical implications for the treatment of leachate using different coagulants. By identifying the specific ranges of pH and coagulant dosages that yield the highest removal efficiencies for turbidity and suspended solids, treatment processes can be optimized for maximum effectiveness. Moreover, these results emphasize the importance of tailoring coagulant selection and dosages based on the specific characteristics of the leachate being treated. It is evident that each coagulant responds differently to variations in pH and dosage, highlighting the need for a nuanced and adaptable approach in wastewater treatment strategies.

3.2.4. Optimization of turbidity removal percentage and suspended solids removal percentage

To enhance the efficiency of the coagulation process in leachate treatment, optimization was conducted using Minitab 17's Optimizer tool. Key parameters, including pH, coagulant dosage, and treatment time, were adjusted to maximize the turbidity removal and suspended solids removal, ensuring optimal treatment performance.

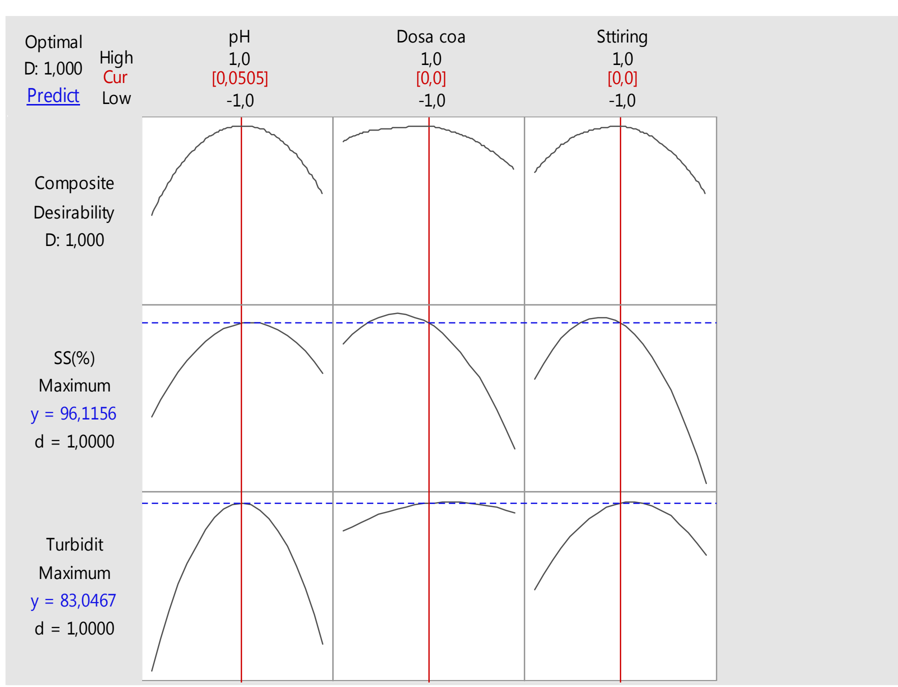

Fig. 14. Optimal Conditions for Turbidity (%) and Suspended Solids (%) Removal using Walnut Shells Coagulant

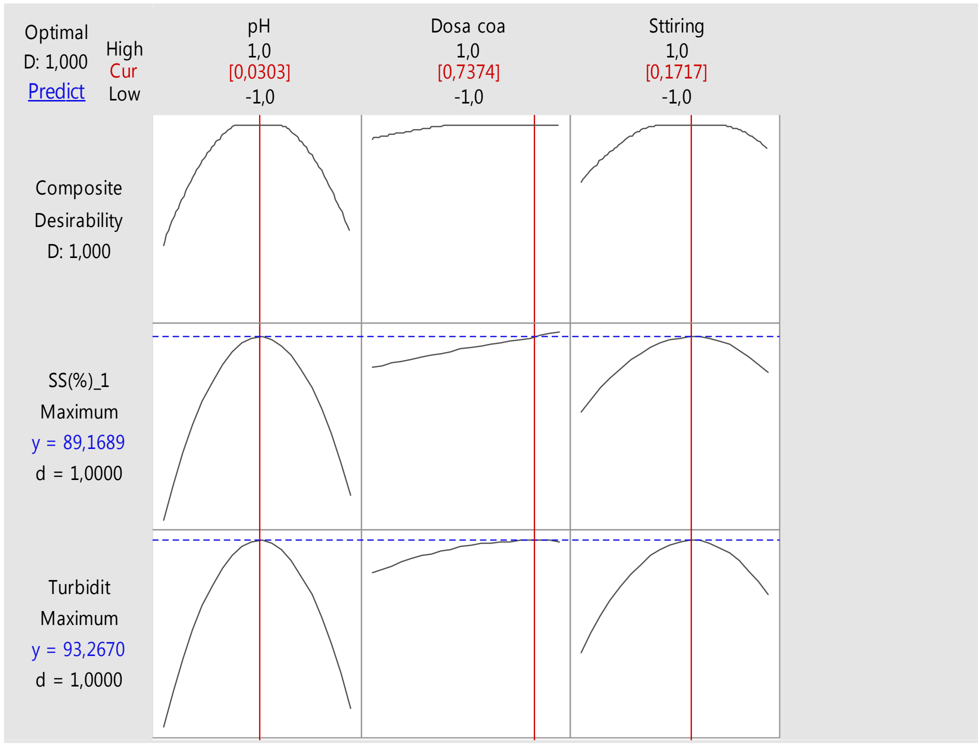

Fig. 15. Optimal Conditions for Turbidity (%) and Suspended Solids (%) Removal using Opuntia Ficus Coagulant

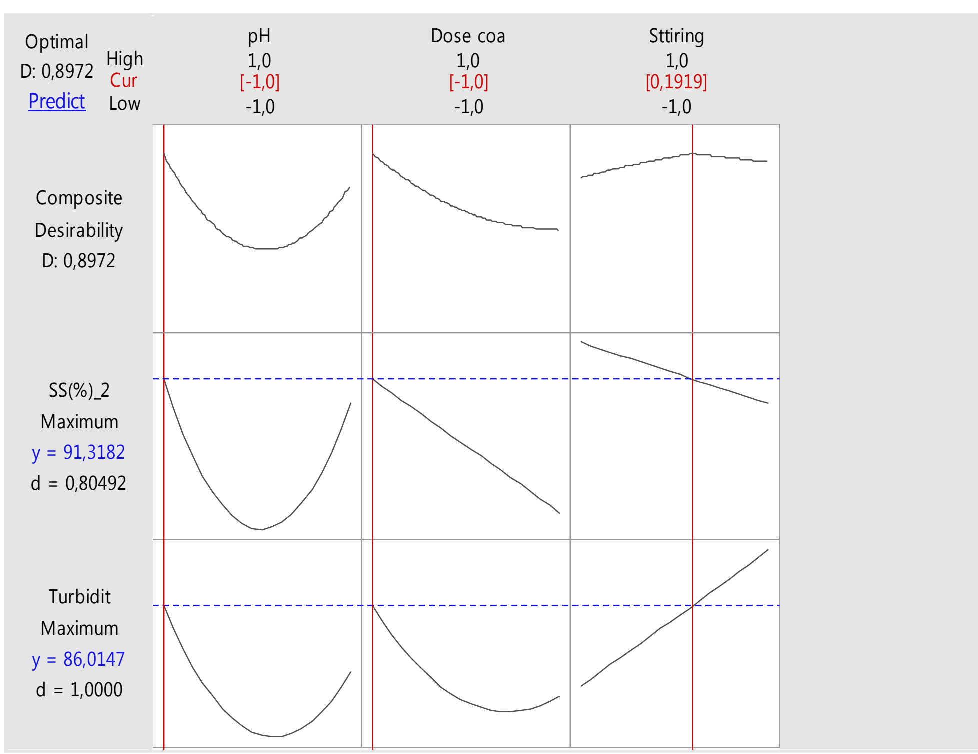

Fig. 16. Optimal Conditions for Turbidity (%) and Suspended Solids (%) Removal using Moringa Oleifera Coagulant

Figs. 14, 15, and 16 illustrate the optimal conditions for turbidity and suspended solids (SS) removal using the three different coagulants. The optimization results by Box Behnken design provide crucial insights into the ideal conditions for leachate treatment, focusing on turbidity and suspended solids removal. Here is an interpretation of the results obtained for each coagulant:

- *Coagulant 1:*

By adjusting pH to 5.68, using a coagulant dosage of 10, and treating for 22.5 min of time, a remarkable turbidity removal of 83.05 % and suspended solids removal of 96.12 % were achieved. These outcomes indicate significant effectiveness in reducing suspended particles in leachate, showcasing the potential of this coagulant in the treatment process.

- *Coagulant 2:*

Maintaining pH 5.61, with a slightly reduced coagulant dosage of 7.05 and a treatment time of 20.35 min, resulted in impressive turbidity removal at 93.27 %, although suspended solids removal was slightly lower at 89.17 %. This suggests that this coagulant can be particularly effective in reducing leachate turbidity.

- *Coagulant 3:*

In this case, by significantly adjusting pH to 2 and using a coagulant dosage of 6 with a treatment time of 20.10 min, a turbidity removal of 86.01 % and suspended solids removal of 91.32 % were achieved. Although the pH value is extremely low, these results show that this coagulant can still be effective, although other considerations such as pH stability in the overall process must be taken into account.

4. Conclusions

The objective of this study is to determine the optimal conditions of the coagulation-flocculation process to eliminate the pollutants present in the leachates of the technical landfill center of Azzaba. Three natural coagulants will be used: walnut shells, Moringa oleifera seeds, and Opuntia ficus-indica leaves. The parameters assessed will be turbidity, suspended solids (SS), and color removal. In the leachate treatment study, three natural coagulants, Moringa oleifera, walnut shells, and Opuntia ficus, were evaluated, each under specific conditions. Walnut shells, with pH 4, coagulant dosage of 12 g/L, stirring speed of 300 rpm, and 25 minutes of treatment, displayed exceptional efficacy, reducing turbidity by 83.92 % and suspended solids by 92 %. Opuntia ficus, maintained at pH 6, with a coagulant dosage of 10 g/L, stirring speed of 300 rpm, and 20 minutes of treatment, achieved a notable decrease in turbidity (86 %) and suspended solids (90 %). Moringa oleifera, at a pH of 3, coagulant dosage of 6 g/L, stirring speed of 300 rpm, and 35 minutes of treatment, exhibited substantial reductions in turbidity (91 %) and suspended solids (85 %). These results underscore the effectiveness of these natural coagulants, particularly highlighting walnut shells as the most efficient in reducing turbidity and suspended solids in leachate.

The combination of flocculants (lime and starch) with the three coagulants shows that both give a significant improvement in treatment, with good results obtained for lime at an optimum dose of 30 g/L for walnut shells and Opuntia ficus, and at an optimum dose of 40 g/L for Moringa oleifera, with turbidity reductions of 88.70 %, 90 % and 92 % respectively. As for starch, the optimum dose is 20 g/L for walnut shells, Opuntia ficus, and 30 g/L for Moringa oleifera, with turbidity reductions of 84 %, 65 % and 73 %, respectively.

The Box Behnken design optimization highlighted the efficacy of the coagulation process in leachate treatment. Coagulant 1, optimized at pH 5.68, 10 g/L dosage, and 22.5 minutes, achieved outstanding results: 83.05 % turbidity removal and 96.12 % suspended solids removal. Coagulant 2, at pH 5.61, 7.05 g/L dosage, and 20.35 minutes, demonstrated impressive turbidity removal (93.27 %) with slightly lower suspended solids removal (89.17 %). Coagulant 3, despite its extreme pH of 2, exhibited significant performance, removing 86.01 % of turbidity and 91.32 % of suspended solids. These findings emphasize the effectiveness of Coagulants 1 and 2 in leachate treatment. However, maintaining pH stability remains critical for optimal results.

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ПОКРАЩЕННЯ ОЧИЩЕННЯ ФІЛЬТРАТУ СМІТТЄЗВАЛИЩА АЗЗАБА ЗА ДОПОМОГОЮ БІОКОАГУЛЯНТІВ З ОПТИМІЗАЦІЄЮ ЗА МЕТОДОМ БОКСА – БЕНКЕНА

Анотація. *Досліджено оптимізацію процесу коагуляції–флокуляції для очищення забруднювачів у фільтраті зі сміттєзвалища Аззаба із використанням природних коагулянтів: шкаралупи волоського горіха, насіння Moringa oleifera та листя Opuntia ficus-indica. Шкаралупа волоського горіха за pH 4, дозування коагулянту 12 г/л, швидкості перемішування 300 об/хв та тривалості обробки 25 хв продемонструвала значне зниження каламутності (83,92 %) і вмісту зважених твердих частинок (92 %). Opuntia ficus-indica за pH 6, дозування коагулянту 10 г/л, швидкості перемішування 300 об/хв та тривалості обробки 20 хв – значне зниження каламутності (86 %) і вмісту зважених твердих частинок (90 %). Moringa oleifera за pH 3, дозування коагулянту 6 г/л, швидкості перемішування 300 об/хв та тривалості обробки 35 хв – істотне зменшення каламутності (91 %) і вмісту зважених твердих частинок (85 %). Додавання вапна та крохмалю як флокулянтів додатково підвищило ефективність оброблення, особливо щодо зниження каламутності. Оптимізація за методом Бокса – Бенкена (BBD) підкреслила виняткову ефективність коагулянтів, особливо їхню надзвичайну продуктивність у видаленні каламутності та зважених твердих частинок. Однак підтримання стабільності pH залишається ключовим для оптимальних результатів. Ці результати підкреслюють ефективність природних коагулянтів, особливо шкаралупи волоського горіха, в очищенні фільтрату, демонструючи перспективний підхід до екологічної реабілітації.*

Ключові слова: *фільтрат сміттєзвалища, коагуляція, флокуляція, природні коагулянти, метод Бокса – Бенкена.*