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Efficiency evaluation of *Luffa cylindrica* and *Mucuna sloanei* seeds in dye removal: A news approach

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ABSTRACT

The use of *Luffa cylindrica* (LC) and *Mucuna sloanei* (MS) seeds as coagulants in the removal of dye by coag-flocculation was evaluated using a new approach. The approach uses the coag-flocculation rate constant K_{11}, calculated using experimental data obtained while using LC and MS as coagulants in coag-flocculation process as a valid indicator to determine the optimum condition. The research also applied criterion for critical coag-flocculation rate constant K_c to satisfy World Health Organization's minimum allowable level of suspended substance for wastewater discharge. In the criterion $K_{11} \geq K_c$ indicates better performance. The results indicated that K_{11} of 0.00214 L/mg·min for LC and 0.00208 L/mg·min for MS surpassed 0.00016 L/mg·min K_c value. This was observed for LC and MS at pH 2, but 1400 mg/L and 1200 mg/L, respectively. From the above, in accordance with the WHO guideline, the dosage of both coagulants meets the requirement. The solution pH could not meet the standard. Nonetheless, MS satisfied the requirement at pH 6, for dosage and pH. These results correspond to what was obtained experimentally. Therefore, in coag-flocculation kinetic modeling, this novel approach can be considered to track the efficiency of these sensitive factors in water treatment plants to meet the effluent quality requirements.

Keywords: *Luffa cylindrica, Mucuna sloanei*, coag-flocculation rate constant, critical coag-flocculation rate constant, dye-based wastewater

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1. INTRODUCTION

The increased manufacturing of dye has resulted in the proliferation of industrial wastewater generation from dye-based industries, particularly to meet the textile needs of the ever-growing population. Throughout manufacturing processes, about 2 percent of the dye makes its way into the effluent and approximately 10 percent of the dye loss occurs throughout the colouring process in dye-based industries [1]. Dye colour is the principal pollutant found in wastewater, due to incompetent manufacturing in dye-based industries. The toxicity and presence of this colour is highly visible and harmful to aquatic life in very small amounts. The decomposition of organic contaminants from dye materials will produce toxic compounds which are considered to have mutagenic effects and are not biodegradable because of their high molecular mass and complex molecular structures [2-4].

Removal of these pollutants requires multiple treatment techniques, such as membrane separation, aerobic and anaerobic degradation with various microorganisms, chemical oxidation, coag-flocculation and reverse osmosis [5], chromatography, lime precipitation, and modified bleaching sequence [6]. Given the advancement of these techniques, coag-flocculation approach stands out as the most feasible primary treatment option for removing contaminants from dye dependent wastewater. This is due to its simple on-site implementation, high treatment performance, flexibility and low assembly and operating costs [7].

Coag-flocculation process is the application of coagulant to wastewater to destabilize and neutralize colour/colloidal dispersion and afterwards aggregate the individual particles resulting from it [7, 8]. Coag-flocculation mechanism that relies on the solution's physical and chemical properties, the contaminants present and the form of coagulant include hydrolysis, coagulation, peri-kinetic and orth-kinetic flocculation [9]. Coag-flocculation is one of the foundation processes at most water treatment plants and modern wastewater treatment plants where they are of great importance in the practice of solid-liquid separation. These are also commonly used for eliminating dispersion of colour/colloid in dye-based wastewater and untreated surface water [8, 10].

Because of their ability to reduce the aforementioned hazards and growing concern for environmental issues associated with the use of traditional coagulants, the search for and the use of biomass for wastewater treatment has gained importance. Natural biomass such as *Moringa olifera*, tannins, *Detarium microcarpum*, etc. have been investigated with effective results for the removal of contaminants [7, 11]. Wastewater treated with natural biomass using a coag-flocculation process, as opposed to synthetic coagulants, poses no danger to biological organisms. The sludge produced may be treated by biological means and used as soil conditioner [2].

Kinetic expression was introduced earlier for the distribution and aggregation of colloidal particles as time progresses based on Smoluchowski's classical work [12]. The rate of particle aggregation depends on the system's physical properties such as particle shape and size, composition, interactive mechanism, collision frequency and the probability of sticking once a collision occurs. These variables are influenced by conditions such as temperature, solution pH, or other chemical properties that influence particle correlations and an external field presence [10].

Modelling is a valuable engineering tool in the field of water treatment plant design and operation. It also helps develop a better understanding of the treatment processes and offers significant opportunities for addressing operational challenges as well as reducing operational costs in a specific process. Coag-flocculation kinetic modeling of water treatment plants can be used to automate processes and check control strategies at a reasonable cost to meet effluent quality requirements [10]. Coag-flocculation kinetics have been investigated and reported for several kinetic functional parameters such as order of coag-flocculation process, constant coag-flocculation rate, coag-flocculation period, coag-flocculation time, Brownian transport collision factor and collision efficiency applying different forms of analytically solved rate law equation [13-16].

Luffa cylindrica and *Mucuna sloanei* are growing throughout Nigeria with limited use. Both of these products are biodegradable, non-toxic and safe for both humans and animals. *Luffa cylindrica* was used as filter sponge and adsorptive biomass support [17], while mucuna seed extract was used in wastewater treatment as bio-coagulant and bio-adsorbent [18, 19]. The purpose of the present work was to develop a different approach using the constant coag-flocculation rate calculated as a valuable indicator based on experimental data to investigate the optimum process conditions and hence the performance of the coag-flocculation process using *Luffa cylindrica* seed (LCS) and *Mucuna sloanei* seeds (MSS) as coagulants. The research also provides an assessment criterion in terms of the critical caog-flocculation rate constant to achieve the minimum allowable level of suspended substance concentration to meet the recommended guideline for industrial wastewater discharge from world health organization (WHO).

2. MATERIALS AND METHODS

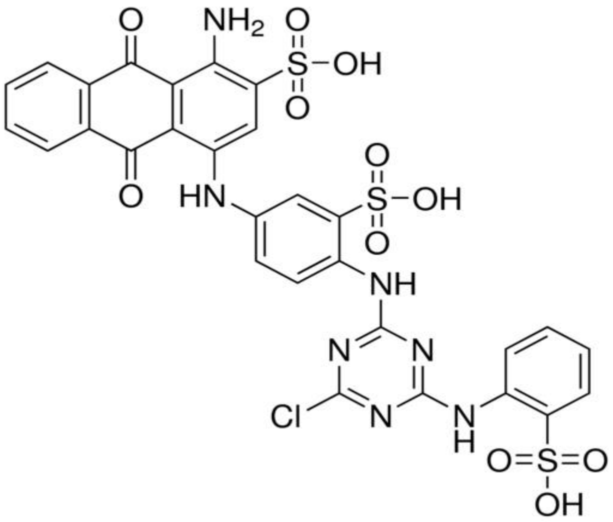

Figure 1. Cibacron blue (3GA) dye

The cibacron blue dye was collected in Enugu, Nigeria (Figure 1) at CONRAWS Scientific Equipment Nigeria Limited. The material for the dyeing was used without further processing. Dye-based wastewater was prepared to get a concentration of 1000 mg/L by dissolving cibacron blue in distilled water. Standard jar testing tools was used to induce coag-flocculation and sedimentation on dye-based wastewater. A total of 200 to 360 mg of LCS and MSS in 200ml aqueous dye-based wastewater at different pH levels (2 to 10) to provide a concentration of 1000 to 1800 mg/L were used and allowed to settle for 150 minutes. A 10 mL of the supernatant pipetted 2 cm below the solution was tested for absorbance at different time intervals and recorded using a UV-VIS spectrophotometer (UNICO 1100, wavelength range 330-900 nm with a 10 mm sample cell length). In this current work, the experimental data findings were used to analyze coag-flocculation kinetics modeling for the constant coag-flocculation rate and critical coag-flocculation rate constant in terms of second order of coag-flocculation process type.

2. 1. Coag-flocculation kinetics modelling

The principles of particle size distribution and aggregation as time progresses began with Smoluchowski's classic work [7, 12, 14, 20, 21]. Through the work the differential equation below defined the generic rate of particle aggregation during the coag-flocculation process:

$$-\frac{dN_t}{dt} = K_{11}{N_t}^{\alpha} \tag{1}$$

where, $N_n(t)$ is the total particle concentration at time, t; K_{11} is the α^{th} order coag-flocculation rate constant, α is the order of coag-flocculation process. To simplify and overcome Eq. (1), the theoretical values of the order, α, of the coag-flocculation process stated to be within the range of ($1 \leq \alpha \leq 2$) [14, 21] were applied. Several literatures have indicated that the process of aggregation mainly takes the form of second order.

That is because the particle collision is proportional to the concentration product of the two colliding species [10, 19, 22]. In addition, taking into account the existing facts and experimental methods, widespread studies such as [11, 14, 18] used ($\alpha = 2$) and found that it was more appropriate primarily to reflect the aggregation rate of particle counts based on Brownian regulated and rapid coag-flocculation. Based on the above facts, in Eq. (1) replacing ($\alpha = 2$) and integrating it with the boundary conditions at the initial state ($t = 0$, $N_t = N_0$) and at the final state ($t = t$, $N_t = N_t$) will give Eq. (2).

$$\frac{1}{N_t} = K_{11}t + \frac{1}{N_0} \tag{2}$$

Using K_{11} as the reference we arrived at Eq. (3) from Eq. (2), Eq. (3) has been used mathematically in this analysis to measure the values of K_{11}.

$$K_{11} = \frac{\frac{1}{N_t}-\frac{1}{N_0}}{t} \tag{3}$$

where K_{11} is the constant of rate of second order coag-flocculation. The initial concentration of simulated dye dependent wastewater before (N_0) treatment and after (N_t) treatment. Such concentrations were used in Eq. (3) and are expressed in (mg/L) and unit of K_{11} becomes (L/mg.mins).

The absorbance was transformed to a concentration of suspended solid particles (N) in (mg/L) using Eq. (4)

$$A = \varepsilon_m N_n(t) L \tag{4}$$

where A = Absorbance, ε_m = molar extinction coefficient, $N_n(t)$ = number particle concentration as time evolves in (mg/L), L = path length of 1cm.

3. RESULTS AND DISCUSSION

Dosage and pH were defined as the most important parameter which influences the determination of optimum coag-flocculation process conditions. It has been determined that inadequate dosage or over-dosage during coag-flocculation process would result in poor results. This is because inadequate dosage results in incomplete destabilization and neutralization of charged colloidal/suspended particles, while over dosage results in the re-stabilization of these suspended particles. Likewise, due to the different reactions during coag-flocculation and the impact of solution pH on these reactions, the optimal pH for better performance in coag-flocculation needs to be determined. Such reactions depend on the composition of both the wastewater to be processed and the coagulant used in the process. It is therefore of paramount importance to assess the optimal state (dosage and pH) in order to reduce the costs incurred as a result of over dosage and thus achieve optimum treatment efficiency [23]. The optimum state is usually determined by systematic experiments, conventionally. Tables 1 - 2 summarize the experimental results obtained using LCS and MSS based on a settling time of 150 minutes.

The residual concentration of TSS particles in mg/L shown in Table 1 - 2 increased as the pH value increased from 2 to 10 by applying different coagulant dosages. From the tables it is clear that the values of residual total suspended substances in other pH values are higher than pH 2 in all dosages from 1000 to 1800 mg/L. The variability could be due to the influence of pH on the reactions resulting in process of coag-flocculation. Nonetheless, it was found that there were minor decreases at the extreme pH value of 10 applying 1200–1800 mg/L for LCS, while the residual suspended substance was observed substantially decreased at pH 6 for MSS.

This could be due particularly, at the extreme and middle values of pH to reactions correlated with changes in pH. In addition, the use of multiple coagulants (LCS and MSS) have shown significant differences in the residual substances suspended. This performance variation is expected because each coagulant has different ability to remove dye in terms of coag-flocculation activity. These abilities thus play a vital role in deciding the optimal condition for the process of coag-flocculation.

Table 1. Experimental optimum dosage and pH for LCS as a function of the residual total suspended substance for dye-based wastewater according to WHO TSS standard.

Dosage (mg/L) ▶ pH/number particle, N(mg/L) ▼	**1000**	**1200**	**1400**	**1600**	**1800**	**Optimum Dosage (mg/L)**	**WHO TSS standard < 40mg/L**	**Optimum pH**	**WHO pH standard = 6 -9.5**
pH 2	**Residual total suspended substance (TSS) after 150 minutes settling time**								
N_{150} (mg/L)	6.8	3.9	3.1	10.9	21.4	1400	Satisfied	pH 2	Not satisfied
pH 4									

N_{150} (mg/L)	391.7	261.8	211.7	120.8	216.5	1600			
pH 6									
N_{150} (mg/L)	432.3	433.9	423.6	344.5	365.7	1600			
pH 8									
N_{150} (mg/L)	431.0	537.7	408.7	373.9	496.3	1600			
pH 10									
N_{150} (mg/L)	591.1	375.9	351.1	94.3	174.3	1600			

Table 2. Experimental optimum dosage and pH for MSS as a function of the residual total suspended substance for dye-based wastewater according to WHO TSS standard.

Dosage (mg/L) ↘ pH/number particle, N(mg/L) ▼	**1000**	**1200**	**1400**	**1600**	**1800**	**Optimum Dosage (mg/L)**	**WHO TSS standard < 40mg/L**	**Optimum pH**	**WHO pH standard = 6 -9.5**
pH 2	**Residual total suspended substance (TSS) after 150 minutes settling time**								
N_{150} (mg/L)	12.5	3.2	36.6	40.7	58.1	1200	Satisfied	pH 2	Not satisfied
pH 4									
N_{150} (mg/L)	693.9	665.5	193.0	85.7	100.6	1600			
pH 6									
N_{150} (mg/L)	75.4	72.5	33.6	59.1	61.7	1400	Satisfied	pH 6	Satisfied
pH 8									
N_{150} (mg/L)	216.1	206.9	145.5	116.1	146.4	1600			
pH 10									
N_{150} (mg/L)	176.5	226.9	158.8	196.6	217.6	1400			

3. 1. Constant coag-flocculation rate as a parameter for optimal dosage of coagulants

The goal of this section is only to use coag-flocculation rate constant K_{11} as an investigative factor to identify optimum dosage at different pH for coagulants (LCS and MSS) regarding their success in meeting the WHO total suspended substance requirement for industrial wastewater discharge. Figures 2 - 6 presented the relation between the constant coag-flocculation rate and the coagulants (LCS and MSS) dosage. Figure 2 and pH 2 showed that the addition of 1400 mg/L and 1200 mg/L dosages resulted in K_{11} of 0.00214(L/mg·min) and 0.00208 (L/mg·min) for the two coagulants (LCS and MSS), respectively.

Other pH values of 4 to 10 for K_{11} recorded lower numerical values at different dosages. The best recorded K_{11} for pH 4 was 4.852e-5 (L/mg·mins) at 1600 mg/L for LCS, and 7.112e-5 (L/mg·mins) for MSS, respectively. Similar results were obtained for solution pH 6–8, applying 1600 mg/L of each of the two coagulants (LCS and MSS).

Nonetheless, pH 10 indicated K_{11}, a value of 3.53e-5 (L/mg·mins) obtained using MSS 1400 mg/L. In general, it can be seen from the results that with an increase in the initial coagulant dosage the values of K_{11} gradually increased to reach a maximum value that represents the optimum dosage.

It then gradually declines to a minimal value. This behavior is predicted as the optimum dosage induces instant load destabilization and neutralization, resulting in more solids being aggregated and settled in shorter time [24]. In this case the optimum dosage was selected based on the maximum values of K_{11}, which corresponds to the optimum dosage obtained experimentally. This is because the higher the K_{11} value, the greater the clarification efficiency [7].

From the findings, it is clear that numerical values of the coag-flocculation constant rate depend on several influencing parameters such as: the initial concentration of TSS particles, type of coagulant, solution pH, and coagulant dosage. However, the findings using the highest numerical values of the coag-flocculation rate constant K_{11}, gave a successful indicator for investigating the optimum dosage in the coag-flocculation process for each coagulant. That is in line with the literature available [10, 19].

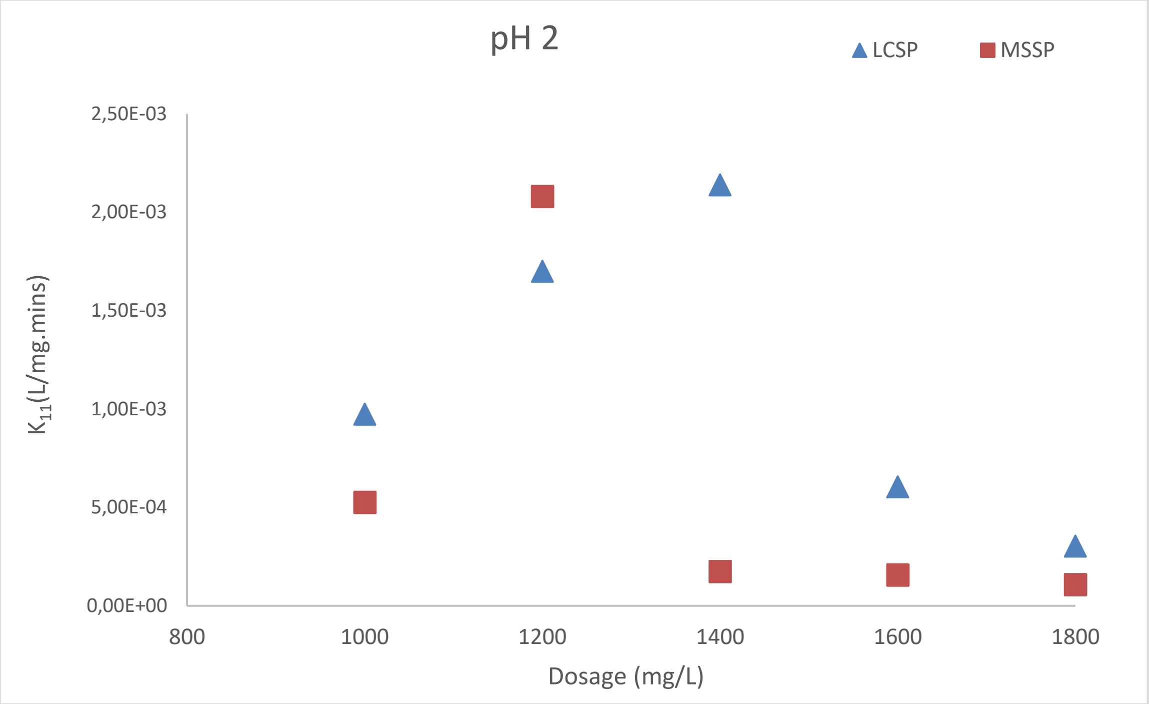

Figure 2. The effect of coagulant dosage at pH 2 on the constant rate of coag-flocculation, K_{11}, in dye-based wastewater

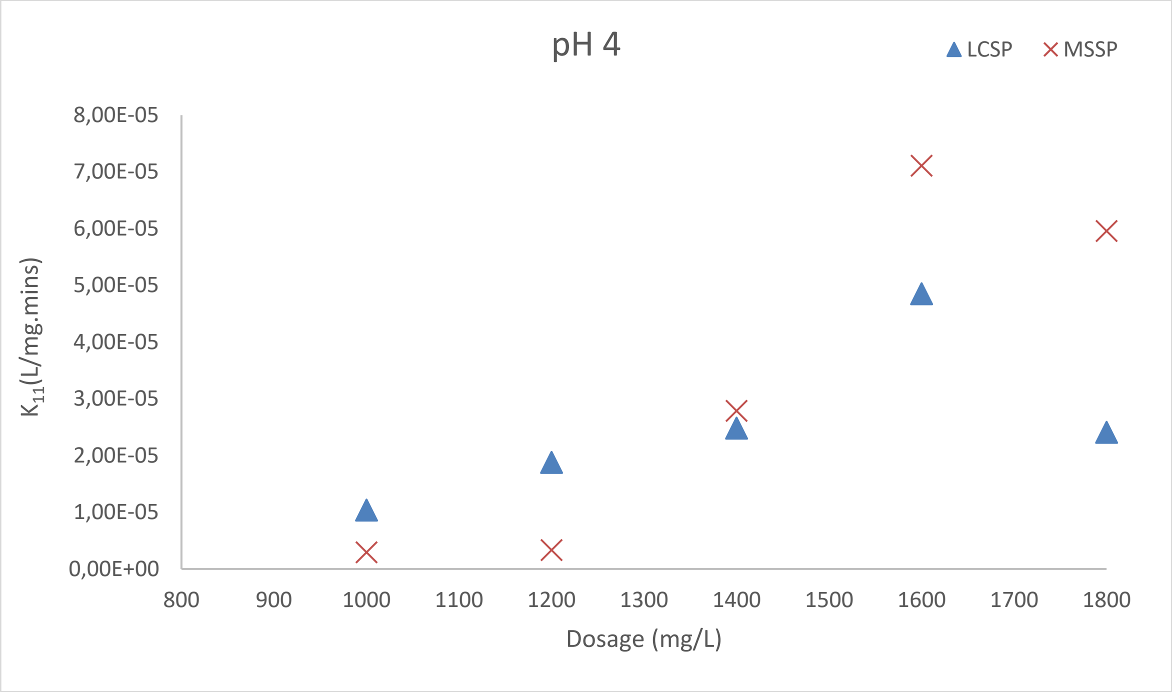

Figure 3. The effect of coagulant dosage at pH 4 on the constant rate of coag-flocculation, K_{11}, in dye-based wastewater

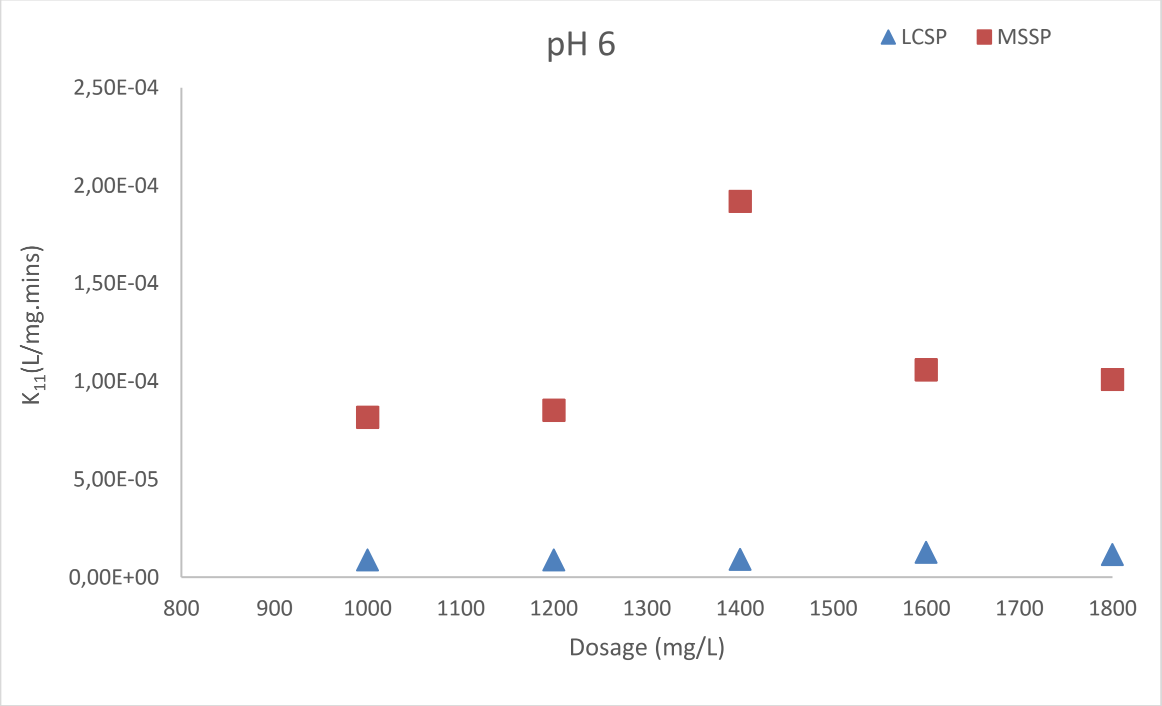

Figure 4. The effect of coagulant dosage at pH 6 on the constant rate of coag-flocculation, K_{11}, in dye-based wastewater

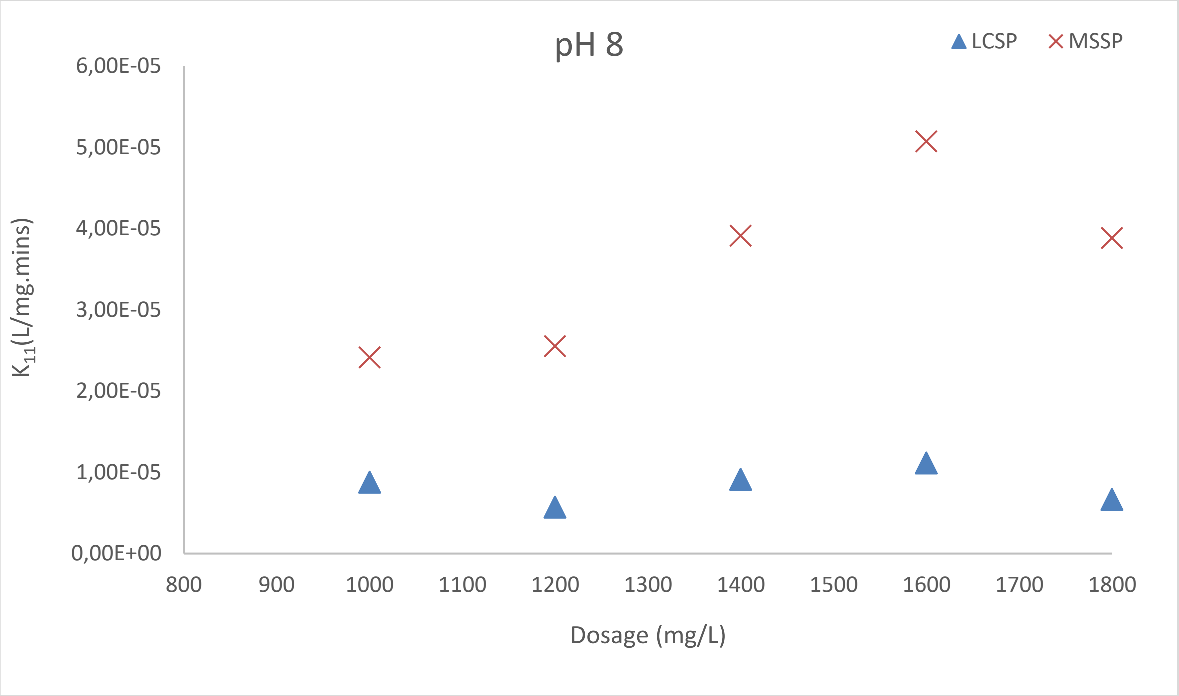

Figure 5. The effect of coagulant dosage at pH 8 on the constant rate of coag-flocculation, K_{11}, in dye-based wastewater

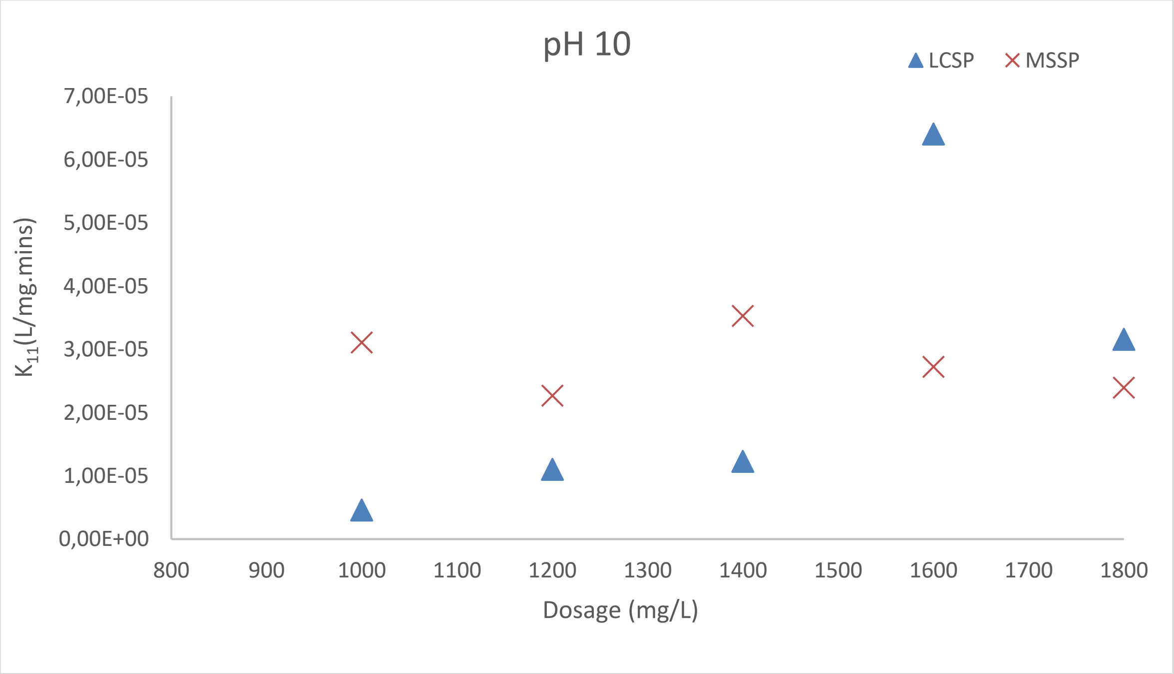

Figure 6. The effect of coagulant dosage at pH 10 on the constant rate of coag-flocculation, K_{11}, in dye-based wastewater

3. 2. Constant coag-flocculation rate as research parameter for optimum pH

Solution pH is one of the dominant factors which affect the process of coag-flocculation. The growing use of natural materials as coagulant has become important for treating wastewater. This is because of their green qualities and their friendliness to the environment. Such natural materials have been reported to be performing better at a lower pH value.

Determining this optimal pH can help to reduce coagulant wastage and achieve better output at reduced time. That will also reduce the cost of the service.

In this section the use of coag-flocculation rate constant K_{11} is studied as an investigative parameter to define optimum pH values for two coagulants (LCS and MSS) applying different coagulant dosages. This is directed towards evaluating their progress in achieving the WHO wastewater discharge standard. The relation between the constant coag-flocculation rate and the solution pH for two coagulants (LCS and MSS) was presented in Figures 7 - 11. Figure 7 showed the effect of solution pH on the constant coag-flocculation rate on the removal of colour from dye-based wastewater using 1000 mg/L of each coagulant (LCS and MSS). For both coagulants the highest constant of coag-flocculation rate K_{11}, values of 0.000527 (L/mg·min) for MSS and 0.000974 (L/mg·min) for LCS were recorded at pH 2.

A small increase in the constant of coag-flocculation rate K_{11}, was observed at pH 6, however. Similar results have been obtained at other 1200 to 1800 mg/L coagulant dosages. Nonetheless, with K_{11} of 0.000192 (L/mg·mins) at 1400 mg/L MSS, the performance at pH 6 was better than pH 2 which gave $K_{11,}$ 0.000175 (L/mg·min). From the observation it is clear that numerical values of the constant rate of coag-flocculation depend on several influencing parameters, type of coagulant, coagulant dosage and solution pH among others. Nevertheless, the results using the highest numerical values of the coag-flocculation rate constant, K_{11}, provided a viable predictor for investigating the optimum pH for each coagulant in the coag-flocculation process. This is in line with the literature available [10, 19].

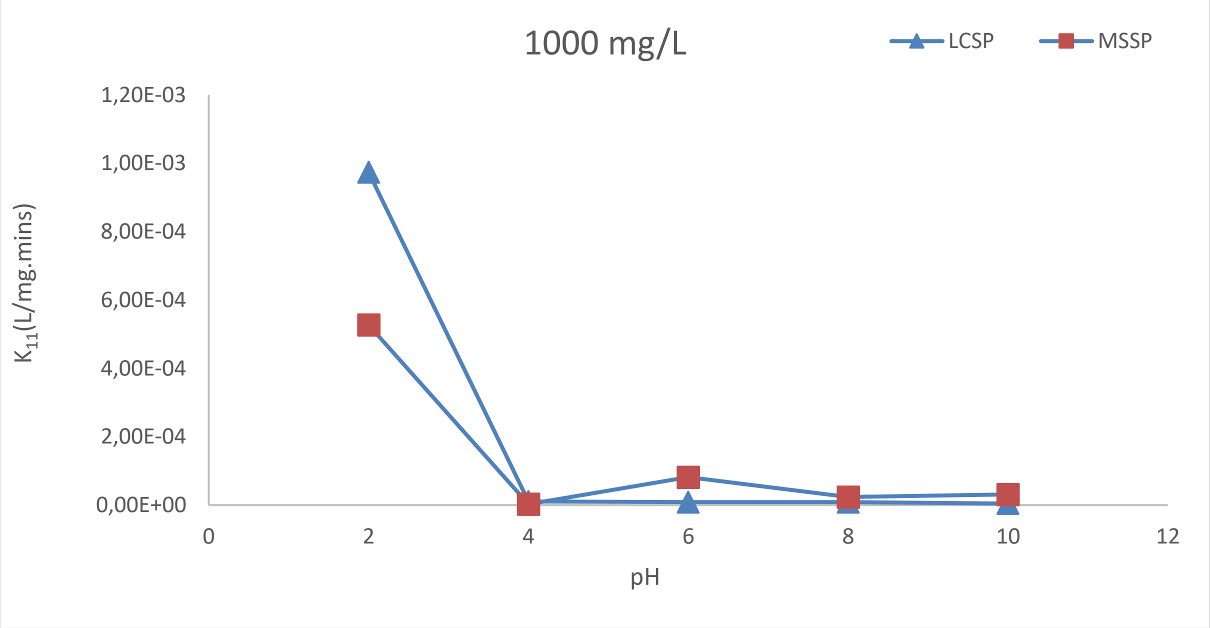

Figure 7. The effect of pH at 1000 mg / L coagulant dosage on the constant rate of coag-flocculation, $K_{11,}$ in dye-based wastewater

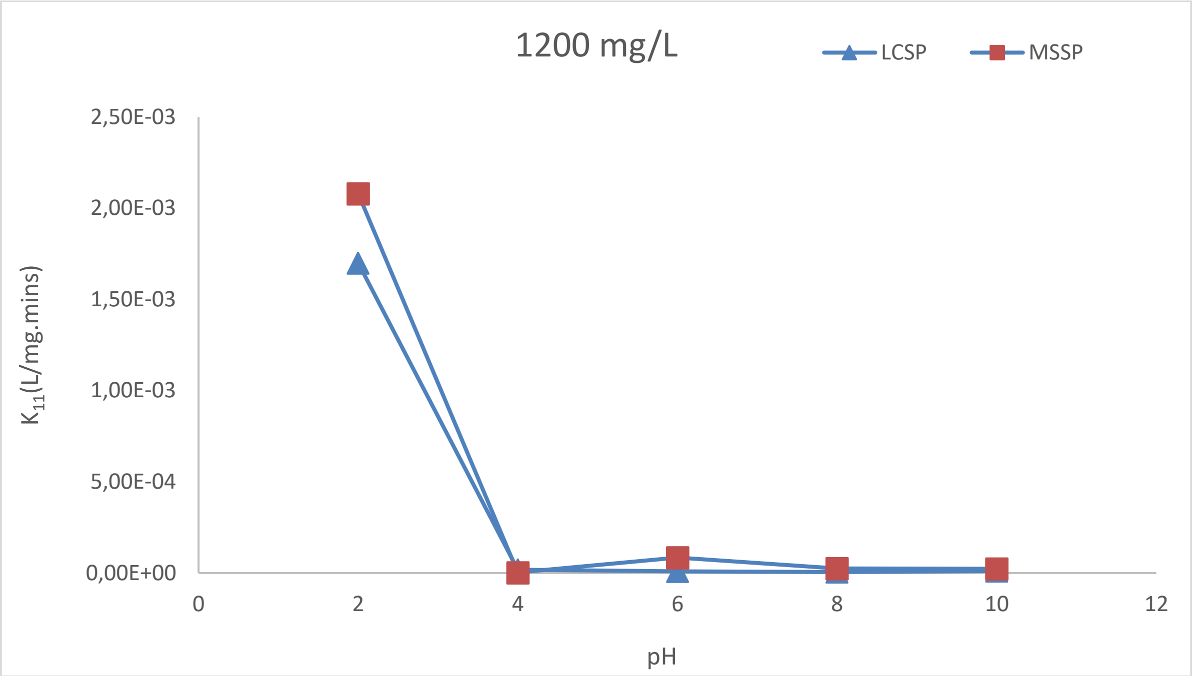

Figure 8. The effect of pH at 1200mg / L coagulant dosage on the constant rate of coag-flocculation, K_{11}, in dye-based wastewater

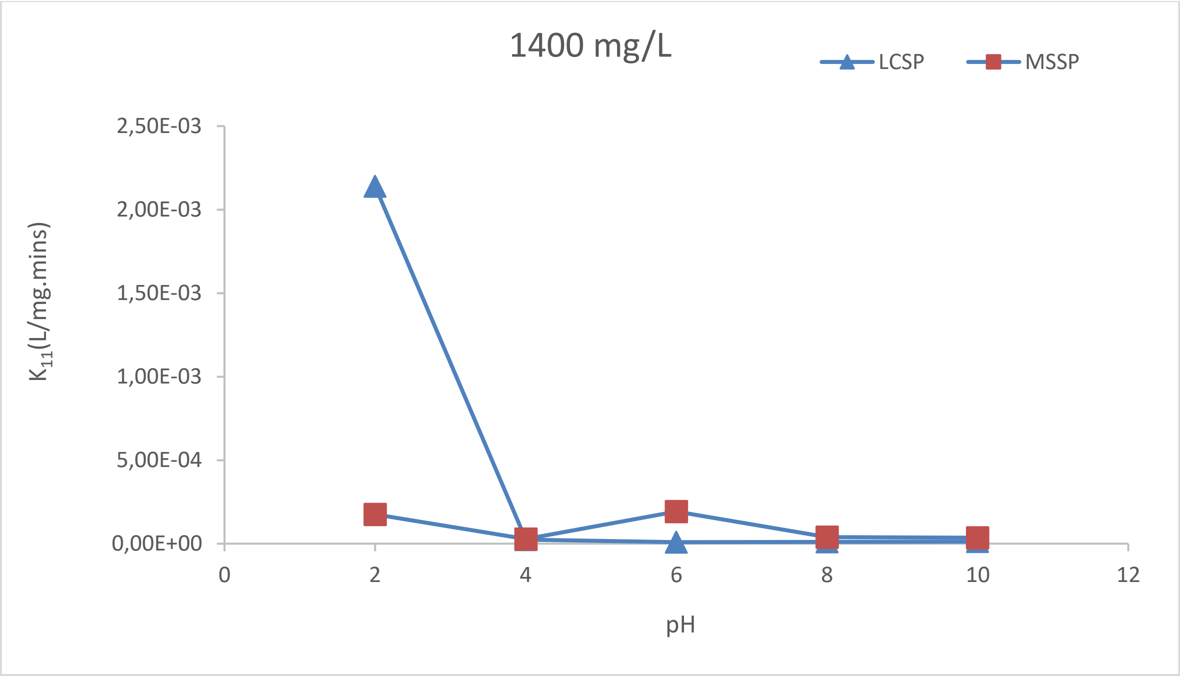

Figure 9. The effect of pH at 1400mg/L coagulant dosage on the constant rate of coag-flocculation, K_{11}, in dye-based wastewater

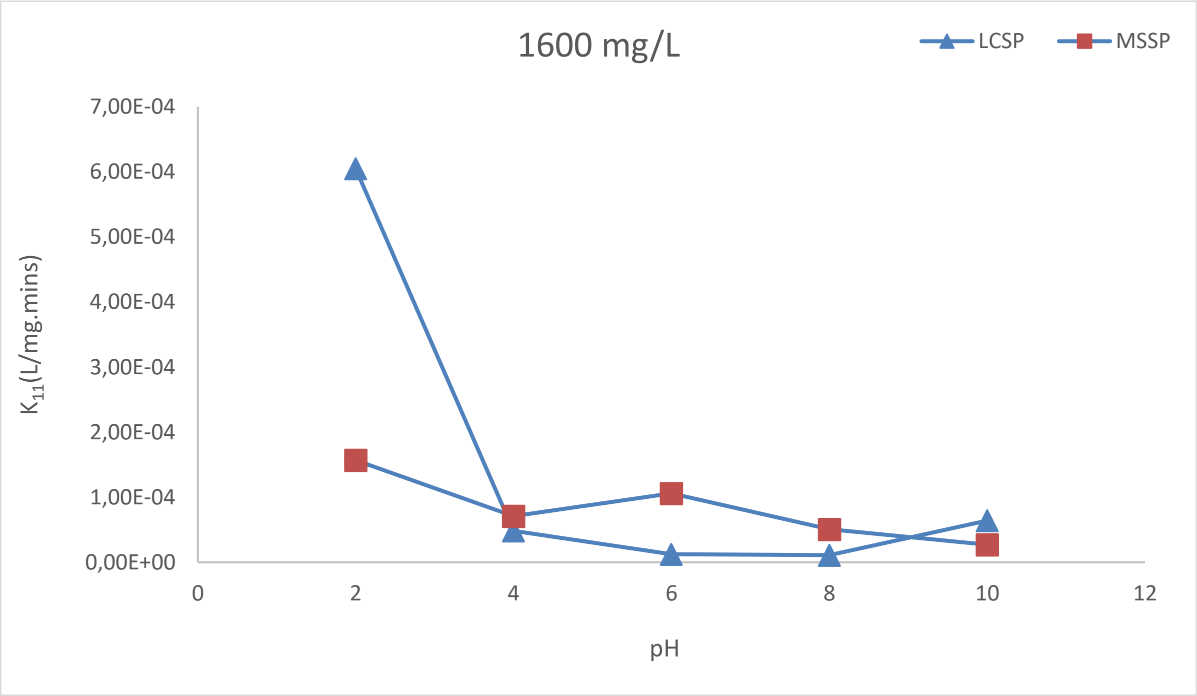

Figure 10. The effect of pH at 1600mg / L coagulant dosage on the constant rate of coag-flocculation, K_{11}, in dye-based wastewater

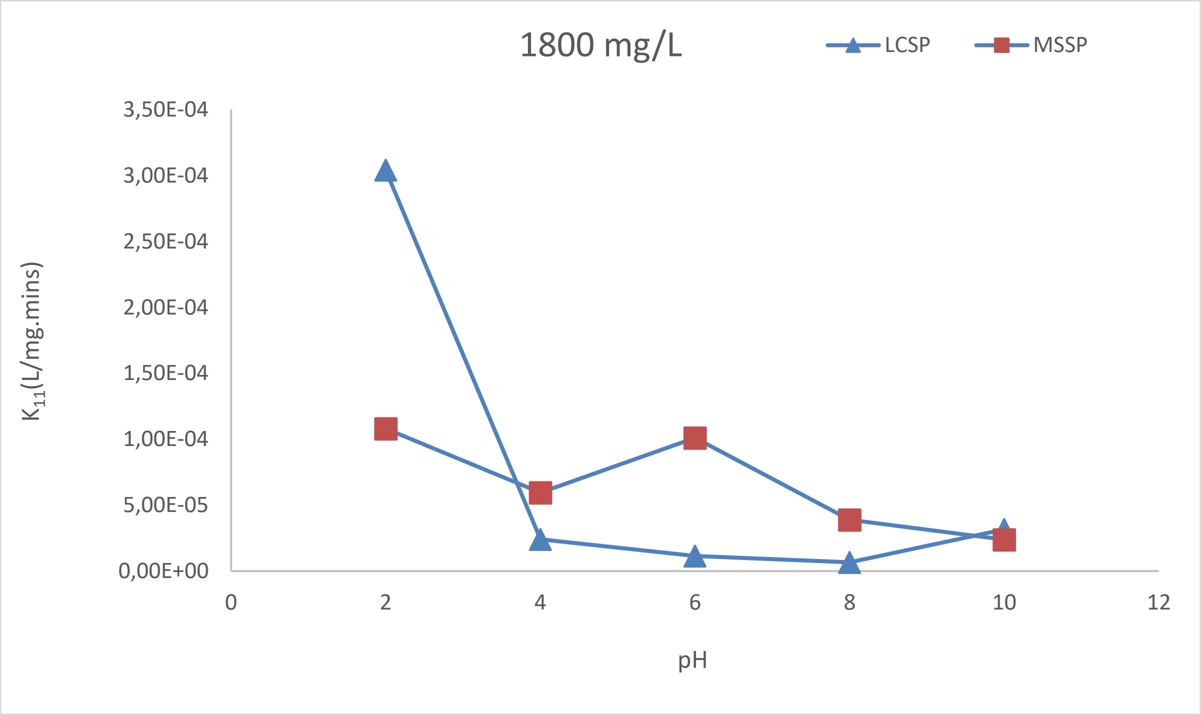

Figure 11. The effect of pH at 1800mg/L coagulant dosage on the constant rate of coag-flocculation, K_{11}, in dye-based wastewater

3. 3. Critical constant coag-flocculation rate as evaluation criterion for performance of coagulants at optimal condition

As recommended by the World Health Organization (WHO) guidelines, the general industrial wastewater discharge standard has an upper limit for total suspended substance (TSS) of 40mg/L [23]. The mathematical representation of the WHO recommendation was investigated by adding the following assumptions: Critical coag-flocculation rate constant, K_c; N_t: is equivalent to the maximum allowable level (40 mg/L) of WHO industrial wastewater discharge of the total suspended substances, N_0: is the original dye-based wastewater concentration (1000 mg/L) t: is the end of the predetermined settling period and is equal to 150 minutes. Replacing the values of (N_{150}, N_0, t) in Eq. 4 in order to obtain the value of a constant of critical coag-flocculation rate, K_c. The efficacy of (LCS and MSS) as coagulants in a coag-flocculation process was assessed in terms of meeting the optimal condition for industrial wastewater discharge of the total suspended substance (WHO) using the following criteria proposed:

$$K_{11} \geq K_c \tag{5}$$

The validity of the proposed criterion was tested with the results obtained experimentally in terms of the total residual suspended substance of the coag-flocculation process as described in Tables 1 - 2 in order to meet the minimum (WHO) suspended substance (almost 40mg/L) of industrial wastewater discharge. Based on the results shown in Figures 2 - 11, it can be observed clearly that the higher K_{11} values correspond to better performance of both coagulants. Such K_{11} values at their predetermined optimum state are evaluated on the basis of the proposed criteria, see Eq. (5) and shown in Figures 12 - 16. Figure 12 shows an optimum K_{11} value (pH 2 & 1400 mg/L) for LCS 0.00214 (L/mg·min) higher than a K_c value of 0.00016 (L/mg·min) and K_{11} value (pH 2 and 1200 mg/L) higher than a K_c value of 0.00016 (L/mg.min) for MSS 0.00208 (L/mg·mins). Consequently, it satisfies the criterion suggested Eq. (5). This implies that their output for industrial wastewater discharge will meet (WHO) total suspended substance standard as regards dosage. It also provided residual TSS of 3.1 mg/L and 3.2 mg/L for (LCS and MSS) respectively, below the WHO TSS standard of 40mg/L, as shown in Table 1. That is consistent with the literature available [10]. The prevalent pH value of 2 at which this efficiency was achieved, however, falls short of the WHO standard for industrial wastewater discharge (6 -9.5). Nonetheless, the WHO standard was achieved for MSS at pH 6, for both dosage and pH, even though it was not optimal dosage. Apparently, it was found that the efficiency at pH 2 was maximum, which happens to be the optimum during experimental. Choosing the optimal condition for MSS would depend on the cost of the coagulant whose dosage is higher at pH 6 and the cost of maintaining solution pH at 2 and applying lower coagulant dosages. Figures 13 - 16 showed the values of K_{11} for pH 4 & 1600 mg/L to be 4.852e-5 (L/mg·mins), pH 6 & 1600 mg/L to be 1.2685e-5 (L/mg·mins), pH 8 & 1600 mg/L to be 1.1163e-5 and pH 10 & 1600 mg / L to be 6.403e-5 (L/mg.mins) for (LCS) which are obviously below the WHO standard of 0.00016 (L/mg.mins). Also, for MSS, the K_{11} values for pH 4 & 1600 mg/L to be 7.112e-5 (L/mg·mins), pH 8 & 1600 mg/L equal 5.076e-5 (L/mg·mins) and pH 10 & 1400 mg/L equals 3.531e-5 (L/mg·mins) in the same Figures 13 - 16 were obviously short of the WHO standard of 0.00016 (L/mg·mins). Therefore, they could not fulfill the WHO criteria for discharging industrial wastewater. At coagulant dosage of 1600mg/L solution pH indicator effect fall short in performance and could be attributed to the suspended substance being re-stabilized due to overdosage. The pH value of 8 could not meet the WHO standard of suspended substance for industrial wastewater discharge, though within the WHO permissible pH level.

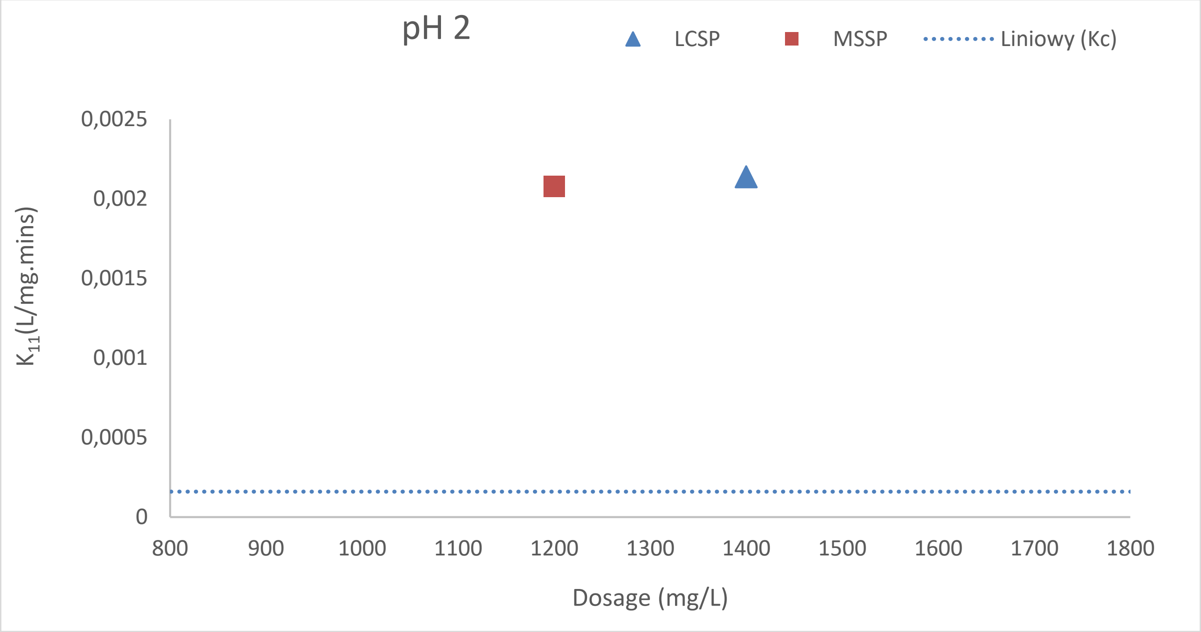

Figure 12. The critical coag-flocculation rate constant (Kc) as an evaluation factor compared to (K_{11}) at pH 2 for optimal dosage predetermined in dye-dependent wastewater

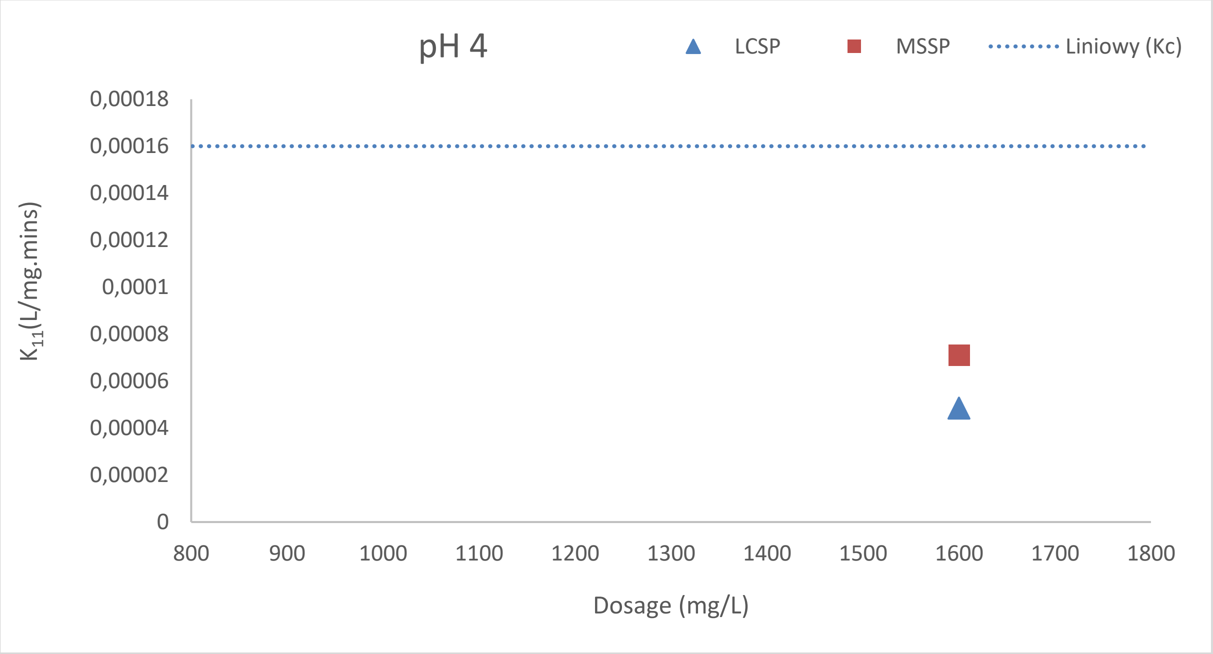

Figure 13. The critical coag-flocculation rate constant (Kc) as an evaluation factor compared to (K_{11}) at pH 4 for optimal dosage predetermined in dye-dependent wastewater

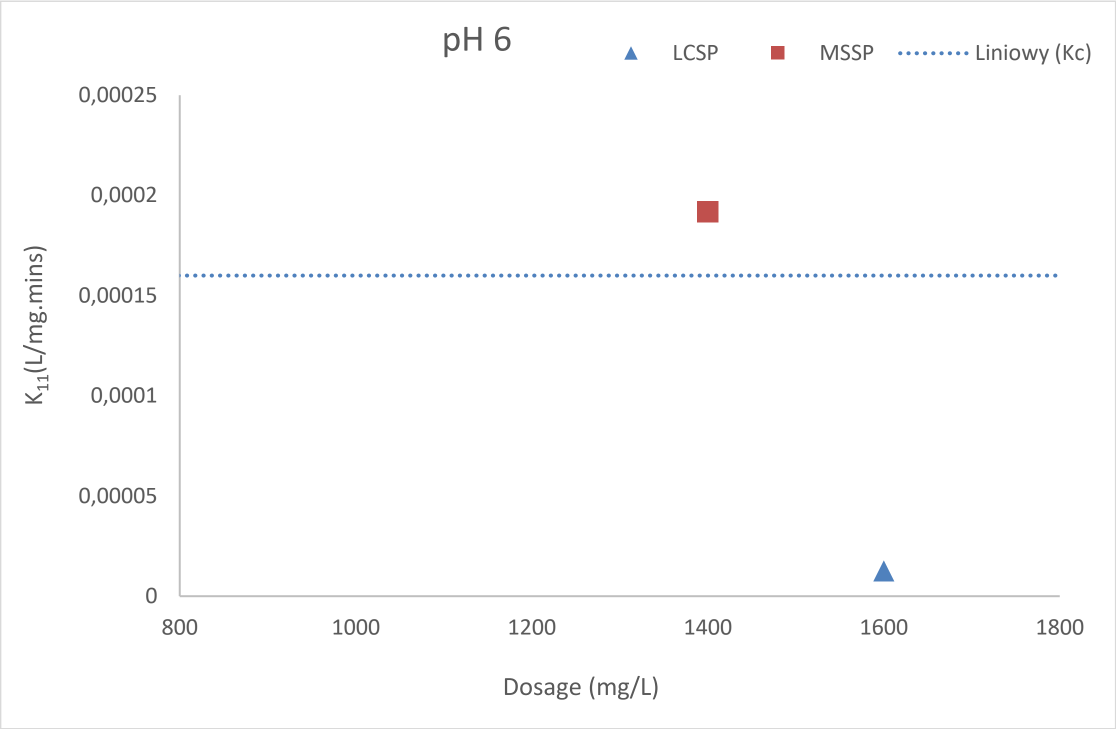

Figure 14. The critical coag-flocculation rate constant (Kc) as an evaluation factor compared to (K_{11}) at pH 6 for optimal dosage predetermined in dye-dependent wastewater

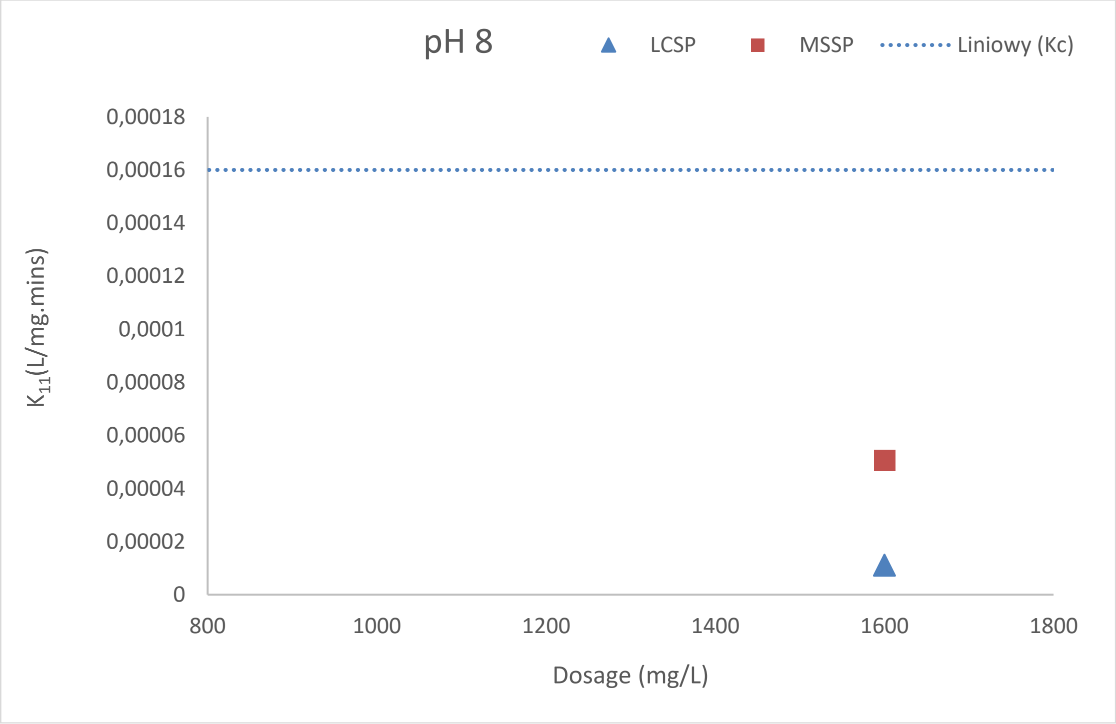

Figure 15. The critical coag-flocculation rate constant (Kc) as an evaluation factor compared to (K_{11}) at pH 8 for optimal dosage predetermined in dye-dependent wastewater

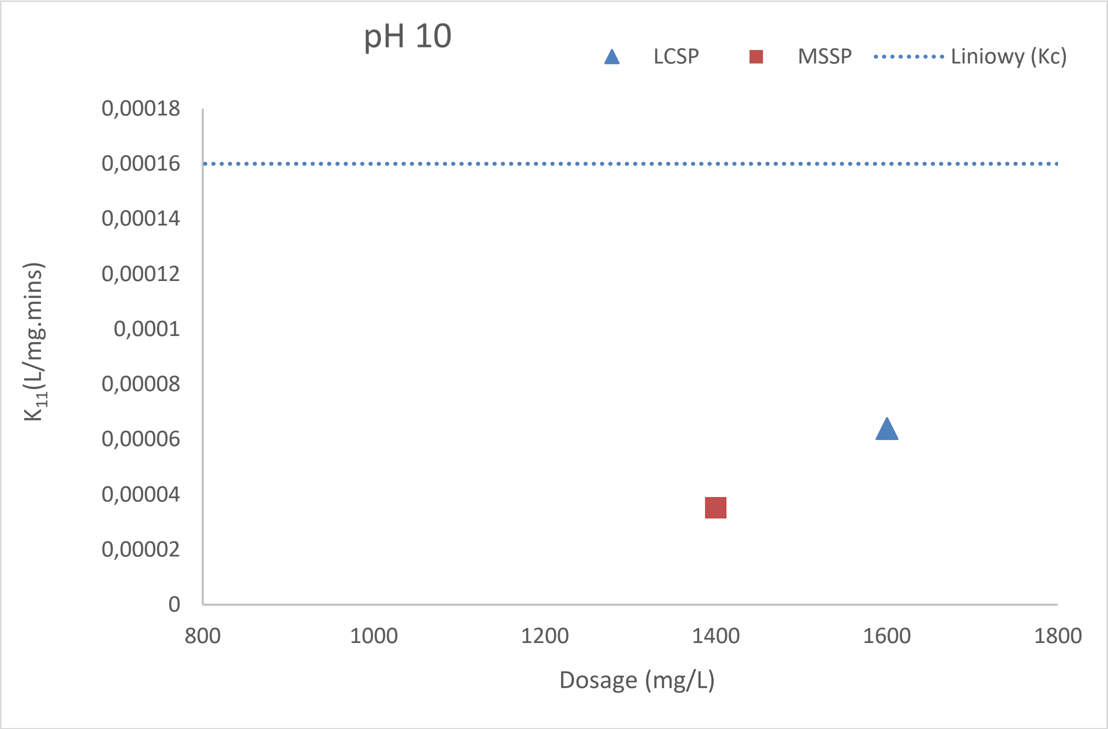

Figure 16. The critical coag-flocculation rate constant (Kc) as an evaluation factor compared to (K_{11}) at pH 10 for optimal dosage predetermined in dye-dependent wastewater

4. CONCLUSION

In the coag-flocculation method, the application of the use of coag-flocculation rate constant as an assessment parameter for determining the optimum condition was investigated using coagulant dosage and solution pH variables. On simulated dye-based wastewater two bio-coagulants (LCS and MSS) were applied. Using experimental data, the use of critical coag-flocculation rate constant as evaluating criterion to determine the efficiency of these coagulants was tested and validated. The results were in line with the experimental findings. Therefore, this novel approach can be considered in the coag-flocculation kinetic modeling to track the efficiency of these sensitive factors in water treatment plants to meet the requirements of effluent quality.

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