

## Evaluation of leachate turbidity reduction in sanitary landfills following a coagulation/flocculation process enhanced by vegetable starch and thermal water

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### ABSTRACT

This study evaluates the reduction of leachate turbidity in landfills after applying a mixture of banana starch and thermal water. Principal component analysis was applied to study the combined effect with four variables: pH, concentration of starch and thermal water mixture, rapid mixing speed and slow mixing speed. The experimental design involved 16 experiments with repetition in the jar test to obtain the optimum dose while measuring turbidity as a response variable. The results showed that after the oxidation process of organic matter, under optimal conditions determined in the experiment, the mixture of starch and thermal water contributed to the reduction of leachate turbidity by 29.1%.

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## 1. INTRODUCTION

The management and proper disposal of solid waste is one of the greatest environmental problems in the world today. Solid waste generates liquids that harm both water sources and entire ecosystems and populations [1, 2]. One option for the final disposal of solid waste is to use sanitary landfills, where the leachate produced by its decomposition is treated, handled and disposed [3, 4]. Leachate is a liquid, with the capacity to contaminate soils and water sources, both surface water and groundwater. It contains heavy metals, nitrogen and phosphorus in high concentrations as well as various types of chemical compounds resulting from decomposition, with a high percentage of normally putrefiable organic matter, which, by fermentation, can produce strong odours that are harmful to surrounding ecosystems [2, 5, 6].

Waste that has been disposed of in open-air sanitary landfills inevitably comes into contact with rainwater [7]. This direct contact, and the capacity of the water to seep through the garbage, generates a series of biological, chemical and physical processes that lead to the generation of different intermediary compounds, both liquid and gaseous, during the stabilisation process. At the end of this process, stable liquid residues, known as 'leachate', are generated, which have a high concentration of mixed solids, both in suspension and in solution, as well as a high presence of heavy metals among other organic and inorganic compounds with a high level of toxicity and that generate risks for both human health and the environment [8-10].

Leachate has a high concentration of total organic carbon (TOC) and chemical oxygen demand (COD), which means that it contains a high concentration of organic and inorganic substances with great variability, depending on the area and time in which the solid waste was disposed of in the sanitary landfill [11]. Among other substances, it is possible to find heavy metals ionised or in colloidal state, nitrogen in ammoniacal state, high contents of total and dissolved solids, presence of chlorides and a variety of organic compounds and carboxylic acids as a product of anaerobic reactions and great variability in the potential of hydrogen ions, even when the leachate has passed through a buffer. The variability in the nature of the wastes depends on the pH, time of the wastes and the average temperature as well as the stabilisation process phase of the solid waste [12-15]. Finding the most suitable and economical way to treat leachate from a landfill generates many difficulties due to the fact that it contains high concentrations of various substances, both inorganic and organic [16]. Over the years, different studies and research have been carried out looking for the most appropriate and economical way to treat leachate [17]. Although solutions have been found in some cases, these results cannot be replicated in other landfills and their respective leachate [18, 19]. Therefore, each leachate from each sanitary landfill must be evaluated individually and tested for treatability in order to find the best way to treat and purify the particular leachate [14, 20, 21].

Coagulation–flocculation is a process whose main objective is to remove suspended material of very low weight and long sedimentation times, such as colloids, bacteria, clays and some inorganic salts. According to some authors, a low pH facilitates the removal of smaller particles that add turbidity and colour to the leachate [22, 23]. It also shows that the use of starches and thermal water in the treatment of wastewater or drinking water, as coadjutants in the coagulation process of ferric chloride and or aluminium sulphate, has positive effects on the treatment itself. The starch derived from banana peel combined with thermal water as a pre-treatment of leachate, may be a good option for its treatability; in addition, the sludge generated by the sedimentation of the flocs may be used in other types of digestion technologies. This mixture presents components and coagulating salts, as well as good flocculation capacity, both of which improve the physicochemical characteristics of the water body that receives this treatment. The importance of the use of vegetal residues in desalination is in harmony with reducing and mitigating the environmental impact and cost while obtaining the necessary primary inputs, given the richness of our soil and the optimal conditions in the acquisition of the resources for the pre-treatment. Several investigations have provided tools for treating leachate; these studies, which are framed within the large area of wastewater treatment processes, were carried out in an extended aeration system. Within the review, there is a treatment process called ‘Biodestil’, an integral technology for the treatment of leachate with high pollutant concentration from open-pit landfills [24, 25]. Wu [26] used submerged filters for this type of treatment. Both surface and subsurface water bodies are highly affected by contaminated leachate discharges [27] due to the difficulty of obtaining system designs that adequately manage and purify these wastes from a sanitary landfill [28, 29] Therefore, management and control of gases and leachate have gained importance in recent years.

In municipal landfills, leachate is produced by the percolation of rainwater through solid waste and the dilution of decomposed products and degradation of solid waste, all of which cause high concentrations of different pollutants in these waters [30, 31]. The concentrations of the different leachates vary widely according to the type of solid waste deposited as well as the density of the waste, the average annual rainfall and the covering or top layer material of the landfill [32].

## 2. RESEARCH METHOD

### 2.1. Coagulation/flocculation processes

In water treatment, there is a chemical phase whose objective is to clarify the solution by removing low-weight particles (colloids). This phase is divided into three parts: coagulation, flocculation and sedimentation (CFS). This chemical treatment is widely applied in drinking water treatment plants and, in recent years, has been applied to the advanced treatment of industrial and domestic wastewater [20, 33, 34].

In CFS, several important factors must be taken into account; one of these is the dosage of chemical compounds, or ‘coagulants’, which are usually inorganic salts or polymers. The objective of coagulants is to neutralise the charges of very small and low-weight particles, known as ‘colloids’. Once the charges have been neutralised, these colloids begin to form larger coagulated particles, known as ‘flocs’ (flocculation process). These flocs have enough weight to sediment out of solution and produce a sludge that can be used according to the treated water and the coagulant used [35].

Coagulants and flocculants can be divided into three important groups:

- Inorganic salts such as ferric chloride, polyaluminium chloride and aluminium sulphate, among others.
- Polymeric organic compounds such as polyethyleneimine and polymeric acrylamide.
- Biopolymeric coagulants of natural origin such as natural and modified starches.

## 2.2. Biopolymers

Starches are natural biopolymers with high molecular weight resulting from the union of several sugar monomers, which in this case is glucose. Different types of biopolymers of different molecular weights, structures, monomer compositions and types of charge can be found. In each biopolymer, the charge contained depends on the ionisation capacity of the functional groups contained in the molecule, the form of the copolymerisation and the way in which the functional groups are replaced. These polyelectrolytes (PE) have three important functions: 1) coagulation, cationic type polyelectrolytes (PEC) cause a reduction in colloid load; 2) the creation of bridges and/or links between colloids, especially anionic polyelectrolytes (PEA) and non-ionic polyelectrolytes (PENI); and 3), especially for high-molecular weight PEC. These types of high-molecular weight PE are used due to their effectiveness as coadjutants in the flocculation process. The PE species form branches that allow the connection between different types of colloids. In this way, the forces that make negatively charged colloids repel each other are broken.

## 2.3. Methodology

The leachate under study was obtained from the La Esmeralda landfill, administered by the SALAS group. This landfill is located to the north of the city of Manizales in Colombia, on the road that leads to the northern part of the department. This sanitary landfill receives between 400 and 500 tonnes of waste per day. In addition to the urban and rural populations of approximately 400,000 inhabitants, 31 municipalities are also served. The disposal service is provided to these municipalities because they do not have their own sanitary landfill. The sanitary landfill has canals and chambers built for the transport and treatment of leachate, and these separate rainwater from the leachate. The disposed waste is compressed in terraces that are divided into strips measuring 5 m high and 10 m wide; each terrace comprises several layers, each measuring 30 cm, with a preferred density of 1-1.5 tonnes per cubic metre. During the compaction process, in order to form the terraces, 'fishbone' channels are built with stone to function as filters on a waterproof layer and to help channel the leachate to the treatment train, which consists of an upflow anaerobic sludge blanket (UASB). It then passes through a reactor upward flow anaerobic filter (UFAF) and then on to the physicochemical treatment area where the coagulation, flocculation, sedimentation and filtration processes take place. There are stacks for the release of gases resulting from anaerobic digestion.

During the transportation of the leachate to the treatment tanks, hydrogen peroxide is added to reduce the biological oxygen demand (BOD) and fats that generate foams, facilitating the coagulation process. Between 10 and 15 mg/L of phosphorus is also added. In Ortega's work, a method of coagulation and flocculation is proposed for the treatment of leachate by applying banana starch. In this work, four study variables were considered: coagulant dose, hydrogen potential, speed or gradient of fast mixture and speed or gradient of slow mixture. A simple sample of 60 L of leachate in thermal water was collected and pre-treatment was performed by adding 0.9 mL (200 ppm) of hydrogen peroxide in a 4% dilution.

The NTC 3903 (Colombian Technical Standard) indicates that a coagulant, or mixture of coagulants, can be added to water, but only if confirmed in the jar test to be appropriate. This coagulant, or mixture, should not negatively affect the quality of the water to be treated or of the receiving water body. The banana starch and thermal water mixture considered in this investigation complies with NTC guidelines. For the plantain starch sample, green bananas were taken, which according to the bibliographic review is ideal for the process to be implemented due to the properties of this starch. The starch is then dried and pulverised to obtain the coagulant starch from the leached starch in the jar test. For the preparation of banana starch, the method mentioned in Laines was used. The bananas were chopped and boiled for an average of 1 hour, and then liquefied and passed through filter paper. To reduce the moisture content, the resulting pulp was dried on the burner for 24 hours and then pulverised.

## 3. EXPERIMENTAL DESIGN

Four variables for turbidity reduction were identified in this investigation: coagulant mixture, pH and slow and fast mixing speeds. The coagulant mixture is the weight/weight percentage of the mixture of thermal water and banana starch. The response variable was turbidity in nephelometric units (NTU) and the variables that were not manipulated and were constant throughout the experiment were leachate (1 L per jar), coagulants (banana starch and thermal water), rapid (50 s) and slow (30 min) mixing time and sedimentation time (30 min). The sample used in the investigation was the same for each experiment, i.e. leached water obtained from the landfill by simple sampling. The design was factorial  $2^k = 16$  experiments. Each variable was identified with the letter A, B, C or D and maximum and minimum values were established as described in Table 1. Analysis of the results was based on a statistical design rather than a traditional analysis of jar tests. According to the factorial design of the experiment, 16 different coagulation and flocculation tests were performed. It was

planned to carry out eight runs, with each one carried out with two beakers, corresponding to maximum and minimum values. The tests were performed in duplicate to reduce the error, giving a total of 32 different tests.

Table 1. Identification of variable factors and maximum and minimum values

Identifier	Variable Factor	Min value (-1)	Max value (1)
A	pH	7.1	8.1
B	Dosage of banana starch and thermal water (coagulant)	0.45 g starch + 40 mL thermal water	0.15 g starch + 80 mL thermal water
C	Fast mixing speed	150 rpm	200 rpm
D	Slow mixing speed	25 rpm	50 rpm

#### 4. RESULTS AND DISCUSSION

Analysis of the results was carried out bearing in mind that in the initial jar tests, at the beginning of the run, each sample was conditioned with acid and base, either hydrogen peroxide or nitric acid and calcium hydroxide, to oxidize the organic matter, with the latter being more effective; however, both treatments increased the turbidity measured in NTU units. The latter being more effective; however, both treatments increased the turbidity measured in NTU units. It is from this turbidity that the tests with the natural coagulants were carried out.

##### 4.1. Statistical analysis

Table 2 describes the statistical treatment by stages. The first phase used the Yates notation. Interactions between the proposed treatments were performed. In Table 3 the analysis of variance ANOVA was performed. In table the effect for treatment AB is the greatest value found. The above implies the analysis of this value. Excel real statistics was used to complete the statistical analysis and help verify the assumptions of normality, homoscedasticity and independence. Analysing the p-values of the Shapiro–Wilk test, the values obtained are greater than the level of significance of the statistical test  $\alpha=0.05$ , which indicates that the data follow a normal distribution and accepting the null hypothesis. The quantil-quantile or QQ plot in Figure 1 corroborates a trend of the data to a straight line.

For the homogeneity test of variances, the Levene test was used, which states as  $H_0$ : The variances of both groups are not different and  $H_1$ : the variances of both groups are different. The obtained ANOVA results show that the p-value is greater than the value of the level of significance, which ratifies that the null hypothesis is correct. The Durbin–Watson test was carried out, showing that there is no self-correlation in the residuals and, therefore, there is independence from the analysed data.

Table 2. Factorial design  $2^4$  and its effects of interest

No	Yates	A	B	C	D	I	II	Total	AB	AC	AD	BC	BD	CD	ABC	ABD	ACD	BCD	ABCD
1	I	-1	-1	-1	-1	184	173	357	1	1	1	1	1	1	-1	-1	-1	-1	1
2	A	1	-1	-1	-1	169	158	327	-1	-1	-1	1	1	1	1	1	1	-1	-1
3	B	-1	1	-1	-1	217	192	409	-1	1	1	-1	-1	1	1	1	-1	1	-1
4	Ab	1	1	-1	-1	192	180	372	1	-1	-1	-1	-1	1	-1	-1	1	1	1
5	C	-1	-1	1	-1	106	90	196	1	-1	1	-1	1	-1	1	-1	1	1	-1
6	Ac	1	-1	1	-1	170	169	339	-1	1	-1	-1	1	-1	-1	1	-1	1	1
7	Bc	-1	1	1	-1	169	173	342	-1	-1	1	1	-1	-1	-1	1	1	-1	1
8	abc	1	1	1	-1	161	165	326	1	1	-1	1	-1	-1	1	-1	-1	-1	-1
9	D	-1	-1	-1	1	196	203	399	1	1	-1	1	-1	-1	-1	1	1	1	-1
10	Ad	1	-1	-1	1	220	198	418	-1	-1	1	1	-1	-1	1	-1	-1	1	1
11	Bd	-1	1	-1	1	182	186	368	-1	1	-1	-1	1	-1	1	-1	1	-1	1
12	abd	1	1	-1	1	179	163	342	1	-1	1	-1	1	-1	-1	1	-1	-1	-1
13	Cd	-1	-1	1	1	214	170	384	1	-1	-1	-1	-1	1	1	1	-1	-1	1
14	acd	1	-1	1	1	139	124	263	-1	1	1	-1	-1	1	-1	-1	1	-1	-1
15	bcd	-1	1	1	1	181	177	358	-1	-1	-1	1	1	1	-1	-1	-1	1	-1
16	abcd	1	1	1	1	141	90	231	1	1	1	1	1	1	1	1	1	1	1

##### 4.2. Analysis of variance and Pareto test

Once the assumptions were verified, the analysis of the experimental design  $2^k$  data was carried out. The results of the ANOVA, taking as a critical value the one obtained for small samples with the t-Student distribution and the Pareto diagram are reported in Table 4. The significance of the analysis was set at a value of  $\alpha = 0.05$ . This guarantees the reliability of the test. With a critical value  $t$  of 2.120, Figure 2 can be analysed.

**Table 3. Contrast, effect, sum of squares and standardised effect**

Contrast	Effect	SC	SCt	Standardised Effect
			308,4414063	
189	a	11,8125	4,36047363	0,557820353
65	b	4,0625	0,51574707	0,191842978
-553	c	-34,5625	37,3302002	-1,632141032
95	d	5,9375	1,10168457	0,280385892
870	ab	54,375	92,3950195	2,567744481
-47	ac	-2,9375	0,26965332	-0,138717231
-315	ad	-19,6875	12,1124268	-0,929700588
85	bc	5,3125	0,88195801	0,250871587
-395	bd	-24,6875	19,0460205	-1,165815023
-29	cd	-1,8125	0,10266113	-0,085591483
-113	abc	-7,0625	1,55871582	-0,333511639
115	abd	7,1875	1,61437988	0,3394145
-435	acd	-27,1875	23,0987549	-1,28387224
13	bcd	0,8125	0,02062988	0,038368596
191	abcd	11,9375	4,45324707	0,563723214
			198,861572	57597,9375
			SCE	57399,07593

SC: sum of squares SCE: sum of error squares

**Table 4. ANOVA**

EFFECTS	SC	G. L	CM	F
A	4,36047363	1	4,36047363	0,001215483
B	0,51574707	1	0,51574707	0,000143765
C	37,3302002	1	37,3302002	0,010405798
D	1,10168457	1	1,10168457	0,000307095
AB	92,3950195	1	92,3950195	0,025755124
AC	0,26965332	1	0,26965332	7,51659E-05
AD	12,1124268	1	12,1124268	0,003376341
BC	0,88195801	1	0,88195801	0,000245846
BD	19,0460205	1	19,0460205	0,005309081
CD	0,10266113	1	0,10266113	2,86168E-05
ABC	1,55871582	1	1,55871582	0,000434492
ABD	1,61437988	1	1,61437988	0,000450009
ACD	23,0987549	1	23,0987549	0,006438781
BCD	0,02062988	1	0,02062988	5,75058E-06
ABCD	4,45324707	1	4,45324707	0,001241343
ERROR	57399,0759	16	3587,44225	
TOTAL	57597,9375	31		

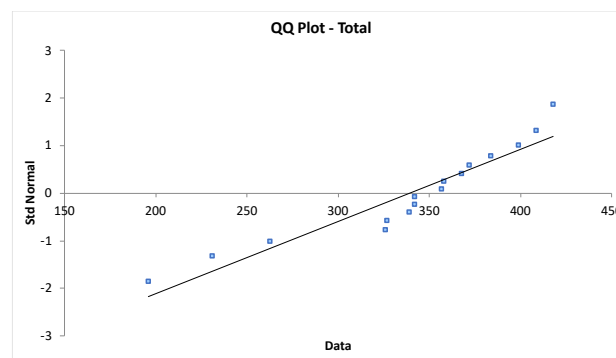


Figure 1. QQ-plot for normality test

**4.3. Results**

When performing the analysis of the experimental design  $2^4$  with the results obtained from ANOVA and Pareto, it was found that the standardised effect serves as a test statistic to test the null hypothesis  $H_0$  (population effect equal zero) against the alternative that the population effect is different from zero.  $H_0$  is thus rejected if the absolute value of the standardised effect is greater than the critical t-student value. The prefixed significance level was  $\alpha = 0.05$ , consulting table for  $t_{0,025;16} = 2.120$ ; any higher value is statistically significant. The only value that exceeds the critical value is the effect of the interaction between pH and coagulant dose, which is in accordance

with Laines for the run corresponding to maximum  $pH = 8.1$ , i.e. normal condition, minimum coagulant dose 0 being significant according to the statistical interpretation, 45 g of starch, 40 mL of thermal water, the rapid mixing speed with a maximum value of 200 rpm and the slow mixing speed with a minimum value of 25 rpm. The pH and the speed gradient are relevant variables of the jar test. Within the individual treatments a value that does not exceed the critical value, but is representative, is the rapid mixing speed.

According to the results, the Pareto diagram shows that the double interaction of the pH and the coagulant dose was the most significant; without exceeding the critical value, the rapid mixing speed appears to be a significant factor. An analysis of the main effects infers that turbidity decreases at the maximum pH as well as at the rapid mixing speed, but not at the minimum values of coagulant dose and slow mixing speed, confirming that the double interaction of pH and coagulant dose was the most significant effect from the experimental design.

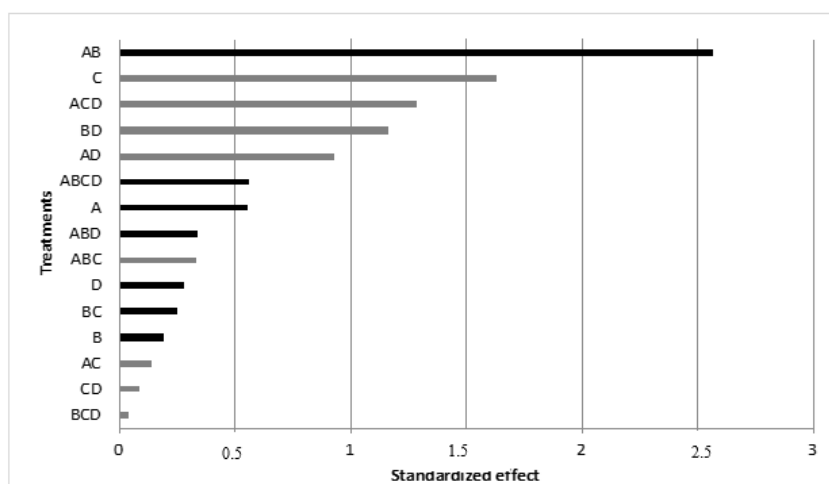


Figure 2. Standardised Pareto chart

## 5. CONCLUSIONS

The use of banana starch as an adjuvant in the flocculation process while mixed with thermal water is promising in the clarification phase because the thermal water provides a significant percentage of sulphates and phosphates to remove turbidity. The properties present in plantain and other vegetables are interesting for water and leachate treatment research, relying in turn on plant sustainability and the reduction of environmental problems while discarding products of consumption, thus generating added value. The use of experimental factorial design allowed for the establishment of factors of influence and relevance in the process in a consistent manner and with a smaller number of tests, thus minimising the costs and determining the optimal factors of the process. Although vegetable coagulants have the potential to be implemented in conventional decontamination treatments, they still do not completely satisfy and it is for this reason that it is recommended to start introducing them proportionally as assistants to chemical coagulants in two ways: 1) as a complement to conventional chemicals and 2) gradually involving the use of plant treatments in the replacement of conventional chemicals. The thermal waters around the city of Manizales contain high level of sulphates, which act as coagulants in the coagulation–flocculation process. At the same time, plantain starch fulfils the function of natural polyelectrolytes, replacing the synthetic chemicals used in conventional treatment.

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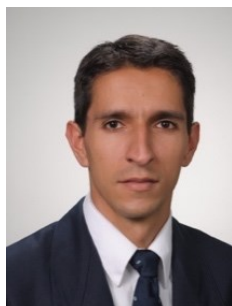
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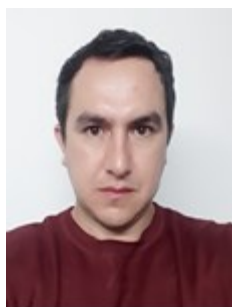
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