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COAGULATION - FLOCCULATION OF DOMESTIC WASTEWATER BY MOROCCAN CLAYS: REMOVAL OF ORGANIC MATTER AND SUSPENDED SOLIDS

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Abstract: Five clay Moroccan origins were employed during the coagulation-flocculation process which is evaluated in treatment of domestic wastewater on the basis of organic material as Chemical Oxygen Demand (COD) and Total Suspended Solids (TSS) removal efficiency. The influence of coagulant dosages and pH on the coagulation-flocculation efficiency process was studied and conditions were optimized corresponding to the best removal of COD and TSS. The optimum doses of each clay were 25 mg/l of perlite, 100 mg/l of clay of Safi and 200 mg/l of stevensite and two clays of Fes. The rate of COD removal was of 66.8%, 60.5%, 57.6%, 54.5% and 44.5% respectively of perlite, clay of Safi, clay of Fes1, stevensite and clay of Fes2. The influence of pH on coagulation effect was investigated under the optimum dosage. Results show that coagulants gave excellent COD and TSS removal efficiency when pH was 10. The clay of Safi has the lowest cost compared to other but perlite has been selected to avoid handling and storage of large volumes of clay at the treatment facility and particularly avoid high production of clayey sludge (to remove 1 kg of COD, we need 207 g of Sc while only 47 g of perlite are sufficient).

Keywords: Coagulation-flocculation, domestic wastewater, clay, perlite, dose, cost.

1. INTRODUCTION

The wastewater is contaminated with organic compounds from all Human activities: domestic, industrial and agricultural and it deserves advanced treatment before being discharged into the environment because of the ecological risk to animals and humans or before biological treatment (Al Ananzehl et al., 2012).

The literature reports a multitude of processes for treatment of wastewater such as physicochemical treatment (coagulation, precipitation, extraction, evaporation, adsorption on activated carbon...) and biological treatment. Each treatment method has its advantages and disadvantages. Coagulation-flocculation is one of the most popular unit operations in water and wastewater treatment trains, is a frequently applied process in the primary treatment of wastewater (and in some cases in secondary and tertiary treatment). It is based on the addition of chemical products which destabilize and flocculate dispersed solid particles and form separable clusters of flocks (Sekiou & Kellil, 2009). Coagulation-flocculation followed by decantation is used worldwide in the wastewater treatment process before the discharge. Coagulation is a very complicated process involving a series of physical–chemical interactions and the type of coagulant applied can play important roles in the removal of target pollutants. The coagulants used in most of the studies are traditional aluminum, ferric-based salts or synthesized organic coagulants (Baoyou et al., 2007). Using these chemical substances may have several environmental consequences as increase in metal concentration in water (which may have human health implications); and production of large volumes of (toxic) sludge (Renault et al., 2009). For these reasons, alternative coagulants and flocculants have been considered for environmental applications (Renault et al., 2009; Bolto & Gregory, 2007; Hamdani et al., 2004). A limitation of this technique is the disposal of sludge produced by coagulation.

Though chemical coagulation may be a method of choice for treating wastewaters before being fed to the biological treatment unit, it too has its drawbacks as the effectiveness is strongly pH dependent (Rashmi et al., 2006). The cost of importing chemical products for conventional treatment might be high and at times prohibitive. To minimize these drawbacks, natural flocculants can be used in treatment by coagulation-flocculation.

Clays are abundant, soft, secondary minerals formed by the weathering of parent rocks. Clays are defined as fine grained aluminosilicate minerals which exhibit plastic properties at a certain moisture content, but harden upon drying (Vanerek et al., 2006). Clays are widely applied in many fields of technology and science, for example, the removal of liquid impurities and purification of gases (Benhammou et al., 2005), the lead adsorption wastewater (Saad & Fares, 2009). We present in this work a study of comparative five clays: stevensite, clay pottery of Safi, perlite, and two clays of Fes.

Stevensite called locally Rhassoul is essentially marketed as a detergent or traditional shampoo. The layer of Rhassoul, resulting from a tertiary formation of Jbel Rhassoul, is located on the east side of the middle Atlas of Morocco (Benhammou et al., 2005). The perlite is a glassy volcanic rock with a rhyolitic composition and 2–5% of constitutive water. Commercially, the term perlite includes any volcanic glass that will expand when heated quickly, forming a lightweight frothy material. The expansion process takes place at temperatures from 760 to over 1200°C and increases the volume of perlite by 10–20 times of its original volume. The obtained material has low thermal conductivity, high adsorption of sound, low bulk density and is fire resistant. Over half of the produced perlite goes into the construction industry as aggregate in insulation boards, plaster and concrete (Alkan et al., 2005a; Ivankovic et al., 2010). Perlite is widely used in many purposes (Rihani et al., 2010; Talip et al., 2009; Ilker & Burak, 2007; Alkan et al., 2005b) but never used as coagulant.

This study examines the performance of coagulation–flocculation using five Moroccan origin of natural clay and perlite as a coagulant–flocculants. It aims to assess the effects caused by the addition of these clays as coagulant at different doses and effect of pH on COD and TSS removal. In other words, to assess the coagulant (clay) optimum in terms of performance (reducing COD and TSS) and less cost.

2. MATERIAL AND METHODS

2.1. Origin and characterization of clay

Table 1 gives a list of the coagulants used in this study. First, the efficiency of each coagulant was assessed, according to the same operating mode for each of them. Then, the operating mode was adjusted depending on the result obtained.

The chemical analysis of clays, dried beforehand at 250 °C and crushed to size lower than 200 µm, was carried out by X-ray fluorescence spectrometry. In order to identify principal minerals present in clays, an X-ray diffractometric analysis was carried out **(**UATRS CNRST**)**.

2.2. Jar test

The samples were collected from the inflow El Jadida city (Fig. 1). Sampling was performed at 15 hours of the day during work days (Monday to Friday). The main characteristics of the wastewater are shown in table 3.

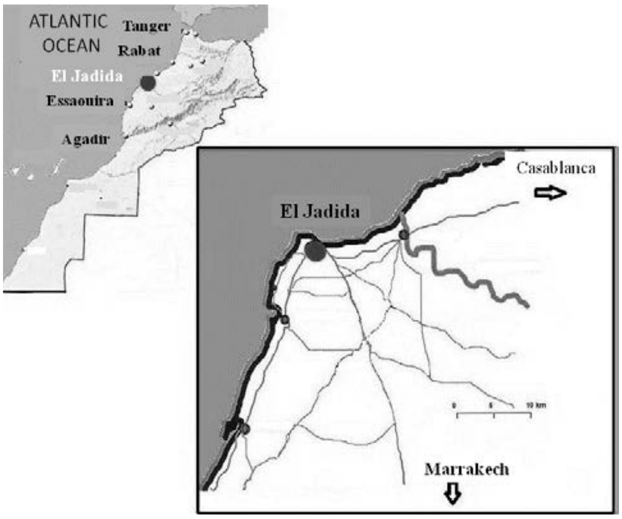

Figure 1. Location of the study area

The experimental study, comprised of coagulation/flocculation, was conducted under controlled laboratory conditions using a standard four-jar test FC4S apparatus manufactured by SBS Instruments S.A by using the Jar Test method. It was equipped with stainless-steel paddles (7.5 x 2.5 cm) and allowed to work simultaneously with four 1-l tall-form cylindrical beakers. Based on the existing bibliography (Hamdani et al., 2005) concerning the treatment of wastewater, after the addition of coagulant, the sample was immediately stirred at a constant speed of 150 rpm for 3 min, followed by a slow stirring at 30 rpm for 20 min.

Table 1. Origin and type of coagulants.

Coagulant	Names	Origin in Morocco and observation
Clay of Safi	Sc	Clay pottery used in Safi
Clay of Atlas (Morocco)	St	Stevensite from east side of the middle Atlas (Jbel Rhassoul)
Perlite	Pc	Expended clay from Tidiennit in south-west of Nador city
Clay of Fes	Fc1	Louajriyne in Fes city
Clay of Fes	Fc2	Region of Fes city

The experiments were performed at room temperature (i.e., 21°C ± 1.5°C). Following the settling time, the samples were collected at the top of the settling beaker (the top 1 cm of the water phase) using a siphoning pipe (Sekiou & Kellil, 2009; Carbonnier et al., 1997; Gibbs & Konwar, 1982).

2.3. Experimental procedure

2.3.1. Effect of adding coagulants on pH wastewater

A known dose of prepared Fc1, Fc2, Pc, Sc, and St was added to a 1000 ml jar containing 1000 ml of domestic wastewater. The coagulant dosages ranged from 12.5 to 250 mg/l as Fc1, Fc2, Pc, Sc, and St. After settling time, the pH value of each sample was measured.

2.3.2. Optimization of coagulant dose and pH in the coagulation process

The influence of coagulant dose on COD and TSS removal was studied by using Fc1, Fc2, Pc, Sc and St as coagulants at concentrations between 12.5 and 250 mg/l. Once the optimal dose coagulant had been determined, the pH was varied in order to observe any effect it caused in COD and TSS removal. The initial pH of the solution was measured and fixed at different values with NaOH and HCl (depending on the experiments), 4.0 ± 0.2, 6.0 ± 0.3, 7.0 ± 0.1, 8.0 ± 0.2 and 9.0 ± 0.3, to study its influence on COD and TSS removal under these conditions (Pallier et al., 2010; Szyguta et al., 2009; Allergre et al., 2004). Subsequently, the efficiency of the process was studied using a dose optimal for each coagulant at pH optimum.

2.4. Analytical methods

Sample pH was measured under magnetic stirring with a Cyberscan 510 pH meter from Eutech Instruments equipped with a combined Ag/AgCl/KCl 4M glass electrode and with a platinum temperature probe. The parameters chosen to establish the optimal conditions of the process were: COD and TSS. The analytical methods were taken from APHA standards methods (American Public Health Association, 1995).

3. RESULT AND DISCUSSION

3.1. Characterization of clays

The chemical composition of natural clays, determined by X-ray fluorescence, is presented in table 2. It emerges that all clays are essentially made up of SiO_2: 53.4%, 53.2%, 52%, 51% and 33% respectively for Pc, St, Sc, FC1 and FC2.

The Clays Sc, Fc1 and Fc2 have a high rate of Fe_2O_3. This explains the red coloring for SC and brown coloring for FC1 and FC2, as well as the Sc present a high rate of K_2O (3.53%) compared to others clays. The MgO content is high for St (25.1%) compared with other clays. The diffractograms presented in figure 2 showed that the dominants phases are quartz, muscovite and calcite for Sc, quartz, montmorillonite and dolomite for St. For clays of Fes, the dominants phases are quartz, calcite, muscovite and kaolinite.

3.2. Effect of coagulants on pH of the wastewater

Removal of organic precursors by coagulation process is impacted by many factors such as coagulation conditions, characteristics of natural organic matter, nature and concentrations of coagulants (Uyak & Toroz, 2007).

The solution pH is an important factor in determining the physical and chemical properties of the sample, and it can be affected by the coagulants. As shown in figure 3, increase in the coagulant dose had no significant effect on pH of domestic wastewater.

The initial pH of the sample was 7.2. After addition of Fc1, Fc2, Pc, Sc or St, the pH increase slightly to 7.31 at 250 mg/l of St.

Invariably, additions of coagulants to wastewater show very little effect on the final pH.

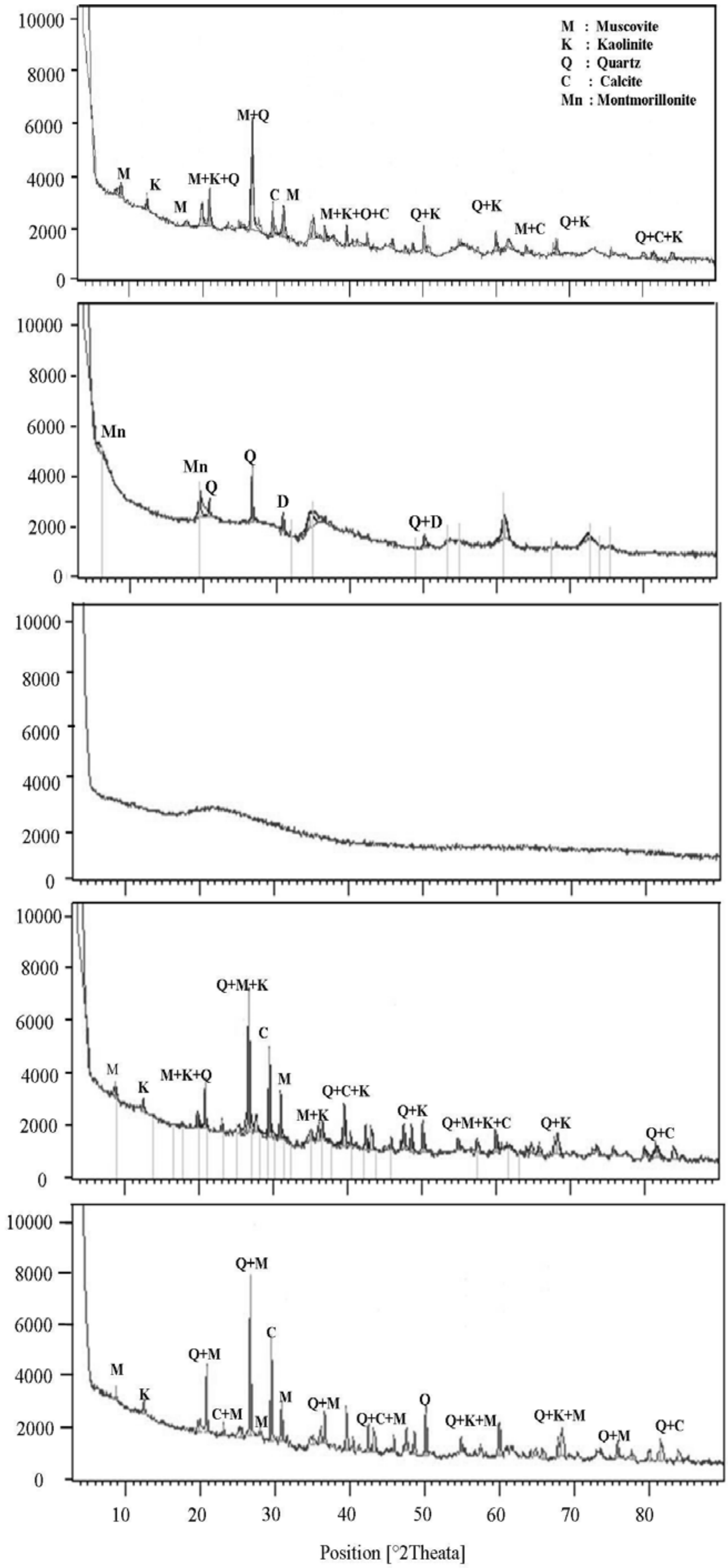

Figure 2. X-ray diffraction pattern of Moroccan clays.

Table 2. chemical composition of clays.

Oxides	SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	K_2O	LOI*	Total
Sc	52	22.3	4.34	3.34	3.03	3.53	9.29	97.83
St	53.2	2.99	0.806	25.1	0.945	0.419	11.8	95.26
Pc	53.4	10.2	0.252	0.489	0.248	1.88	30.7	97.17
Fc1	51	15.5	3.77	3.72	12.2	1.39	15.6	103.18
Fc2	33	10.3	6.18	2.49	6.94	0.75	15.5	75.16

*LOI: loss On Ignition

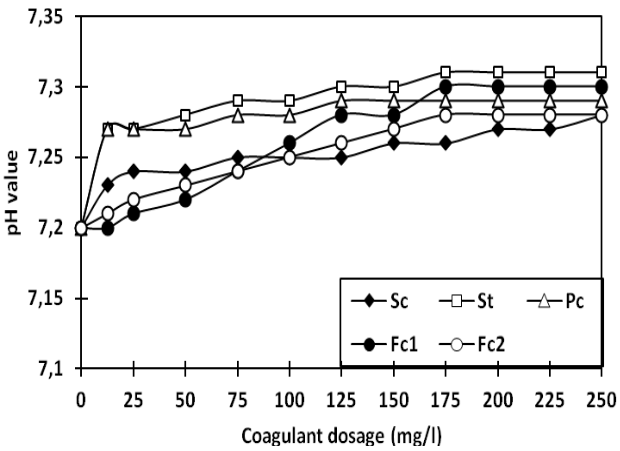

Figure 3. Effect of coagulant dose on pH of the wastewater.

The pH stability indicates that coagulation of domestic wastewater by these various coagulants could offer a possibility for treated wastewaters which may have a neutral pH value. However, compared with chemical coagulants such $Al_2(SO_4)_3$ or $FeCl_3(6H_2O)$, where pH can be affected by these coagulants (Song et al., 2004).

3.3. Optimization of the coagulant dose and the performance in the coagulation process

In the coagulation–flocculation process, coagulant dosage plays an important role in determining the coagulation efficiency (Huaili et al., 2011). The removal of organic matter from water with coagulation process varies widely, generally between 10 and 90% (Uyak & Toroz, 2007; Uyak & Toroz, 2006; Randtke, 1988).

3.3.1. Reduction of COD and Total Suspended Solids

Jar tests were carried out with coagulants (clays) at different predetermined Fc1, Fc2, Pc, Sc, and St doses of 0 to 250 mg/l at increments of 25 mg/l, keeping the other conditions constant (pH 7.2). The optimum dose was determined in terms of COD removal efficiency. When clays were used as coagulant and whatever its concentration, the decantation took place. Figure 4 shows the effect of coagulant addition on the percentage removal of COD at wastewater pH. The results indicate that COD removal efficiency increased substantially with the increase of the coagulant dosage, but for coagulant Pc when the dosage was of 25 mg/l, the highest COD removal efficiency was achieved and the curves approached plateau. Generally, the removal rate increased with the increase in dosage. After the addition of the Pc, Sc, St, Fc1 and Fc2 at a concentration of 12.5 mg/l, the COD concentration of wastewater decreased by 53.5, 46.5, 33.3, 30.3 and 6.1% respectively. On the basis of an average initial COD concentration of 800 mg/l, the results indicate that a maximum COD removal of 57.6, 54.5 and 44.5% at 200 mg/l using Fc1, St and Fc2 respectively; of 60.5% at 100 mg/l using Sc and of 66.8% at 25 mg/l be achieved by using Pc after settling. Nevertheless, in terms of COD removal, the optimum coagulant dose is similar for St, Fc1 and Fc2 about 200 mg/l, but Sc is more effective than St, Fc1 and Fc2 at 100 mg/l. Thus, 200 mg/l of St, Fc1 and Fc2, 100 mg/l of Sc and 25 mg/l Pc were the optimal doses in terms of COD removal. Coagulation using Sc appeared to be a little more effective than St, Fc1 and Fc2 in removing COD.
Increasing the dose of coagulant St of 100 mg/l to 200 mg/l did not further improve the COD removed.

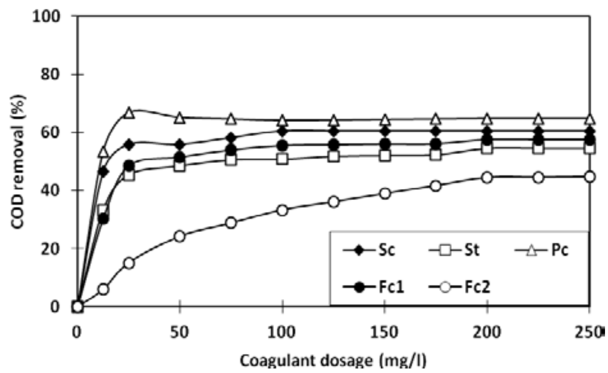

Figure 4. Effect of coagulant dose on percentage removal of COD.

With increase in Pc coagulant dose up to a certain level (250 mg/l), the percent removal of COD a decreasing trend was observed with further increase in dose level. However, an over optimum amount of coagulant would cause the aggregated particle to redisperse and would also disturb particle settling

(Rashmi et al., 2006; Chan & Chiang, 1995). Pc was found to be most effective for Sc followed by Fc1, St and Fc2, respectively, at a low dosage of 25 mg/l.

The dosages needed by the conventional coagulants were four to six times more than Fc1, Fc2, Pc, Sc, and St. Furthermore, Pc is proved to be a better coagulant compared with Sc, St, Fc1 and Fc2 even at lower dosage; 25 mg/l of Pc could remove 66.8% of organic matter.

Figure 5 shows the influence of coagulant dose on the removal of TSS at wastewater pH 7.2. After the addition of the St, Sc, Pc, Fc1 and Fc2 at a concentration of 12.5mg/l, the TSS concentration decreased by 83.9, 81.9, 67.7, 22.7 and 20.4% respectively. As the coagulant dose increased, the reduction of TSS increased. Considering an initial TSS concentration of 358mg/l, a maximum removal efficiency of 94.6% (remaining TSS of 19.2 mg/l), 91.6% (remaining TSS of 30 mg/l), 84.7% (remaining TSS of 54.9 mg/l) and 74.9% (remaining TSS of 89.9 mg/l) was reached at a dosage of 250 mg/l for Sc, St, Fc2 and Fc1 respectively and of 98.9% (remaining TSS of 4 mg/l) at 50 mg/l be achieved by using Pc.

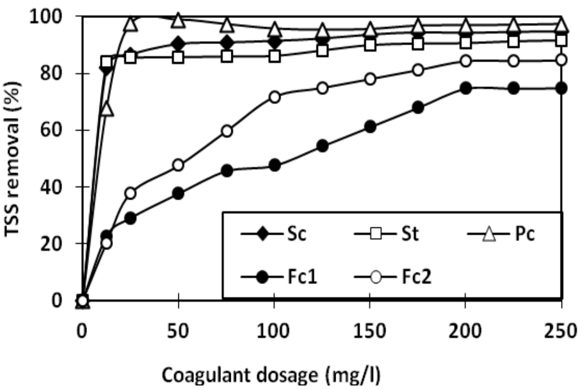

Figure 5. Effect of coagulant dose on percentage removal of TSS.

In this study, 100 mg/l coagulant dosage was chosen for Sc and 200 mg/l for St, Fc1 and Fc2. Moreover, even though 250 mg/l coagulant dosage resulted in COD removal lighter in domestic wastewaters coagulation, because of economical and engineering point of view and considerations, 100 mg/l coagulant dosages was selected for Sc, 200 mg/l for St, Fc1 and Fc2, and 25 mg/l for Pc were chosen for studies of pH of coagulation.

Once the optimal dosage for each coagulant had been determined, experiments were performed on elimination efficiency of TP, TKN, NH_4-N, FC and FS. The optimal doses chosen were 100 mg/l with Sc, 25 mg/l with Pc, and 200 mg/l with St, Fc1 and Fc2. The optimum dose of a coagulant is defined as the value above which there is no significant increase in removal efficiency with further addition of coagulant (Amuda & Alade, 2006). The addition of the different clays had hardly any effect on the elimination performance TKN, NH_4-N, TP, FC and FS (Table 3) while the removal efficiency of COD and TSS increased with coagulant dose.

3.3.2. Sludge production

In general, the amount and the characteristics of the sludge produced during the coagulation-flocculation process depend on the coagulant used and operating conditions (Amuda & Alade, 2006). The effects of coagulants on percentage sludge volume from jar tests for coagulation of domestic wastewater are shown in table 3. The settled sludge volume is highly influenced by these different clays. This may be explained if different qualities of sludge formed as a result of high density of flocs forming during coagulation (Song et al., 2004).

The settling of the particles obtained with the optimum operating protocol allowed the determination of the settling time. Our objective was to show that the settling time was not too big and also that the supernatant could be used directly. At optimum dosage for all coagulants used, the settling time decreased considerably, giving a minimum for each coagulant.

Table 3. Efficiency of different clays in the removal of pollutants of domestic wastewater at optimal dose.

Parameter	Raw Wastewater	Optimal dose of coagulant (mg/l)				
		Sc: 100	St: 200	Pc: 25	Fc1: 200	Fc2: 200
Ph	7.2	7.25	7.3	7.27	7.3	7.28
COD removal (mg/l)	800	484	436	534	461	356
TSS (mg/l)	358	31	33	9	90	55
TKN (mg/l)	60.2	58.8	57.4	56	56	58.8
Ammoniacal-N (mg/l)	50.4	37.8	45	37.8	46.8	46.8
Total Phosphorus (mg/l)	0.212	0.176	0.184	0.203	0.19	0,200
Fecal coliforms CFU/100ml	$1.40\ 10^{8}$	$1.6\ 10^{7}$	$1.1\ 10^{8}$	$3\ 10^{6}$	$1.1\ 10^{8}$	$3\ 10^{7}$
Fecal streptococci CFU/100ml	$3,2\ 10^{4}$	$2.1\ 10^{4}$	$1.96\ 10^{4}$	$1.87\ 10^{4}$	$1.98\ 10^{4}$	$1.85\ 10^{4}$
Sludge volume (ml/1000ml)*	-	8	8.1	8	7	6.5
Settling time (min)**	-	30	25	20	25	35

*Maximum volume of sludge decanted, **Minimum settling time

Table 4. Correlation coefficient obtained in the linear regression analysis between pollutants variables and chemical composition of clays

Elements	Correlation coefficient, r						
	COD	TSS	TKN	NH_4-N	TP	FC	FS
SiO_2	0.7222	0.0934	0,309	0.2644	0.1866	0.113	0.7184
Al_2O_3	0.0734	0.0366	0.0583	0.1708	0.1681	0.0489	0.3976
Fe_2O_3	0.4689	0.3226	0.4256	0.1501	0.0077	0.0596	0.0011
MgO	0.0391	0.0096	0.0015	0.0985	0.1695	0.356	0.0171
CaO	0.1539	0.9415	0.0156	0.4418	0.0022	0.2952	0.0099
K_2O	0.3234	0.077	0.043	0.6178	0.2058	0.2527	0.4159

3.3.3. Relationship between pollutants variables and chemical composition of clays

Table 4 shows the correlation coefficient obtained in the linear regression analysis between pollutants variables and chemical composition of clay. The relationships between the chemical composition of clay (SiO_2, Al_2O_3, Fe_2O_3, MgO, CaO, and K_2O) and the physicochemical and microbiological variables were investigated by applying linear regression analysis using the data obtained during the coagulation/flocculation processes with this clay. The removal level of COD and TSS is significantly correlated to the presence of SiO_2 and CaO respectively of the clays used. More specifically, the correlation coefficient, r, is in the range of 0.7222 for SiO_2, 0.9415 for CaO and 0.6178 for K_2O.

3.4. Determination of the optimal coagulation pH

In the coagulation–flocculation process it is very important to control pH since the coagulation occurs within a specific range for each coagulant. With the previously established coagulants doses, the pH was varied within a range of 4–10. Literature findings reported that the pH of coagulation was dependent on the coagulant type and treated water sample (Uyak & Toroz, 2007; Volk et al., 2000). This parameter is a critical parameter in the efficiency of the coagulation-flocculation process. It influences the solution properties. Therefore, the 200 mg/l coagulant dosage of St, Fc1 and Fc2, 100 mg/l of Sc was chosen for all the subsequent experiments, and 25 mg/l coagulant dosage of Pc.

3.4.1. Effect of pH on COD and TSS removal

The influence of pH on COD removal by different clays was studied. The results obtained are presented in figure 6. They have revealed that pH is the dominant parameter controlling the coagulation-flocculation. It was found that, for all the coagulants, the residual COD of the effluence decreased with the increasing pH. It can be seen that COD removal is most effective at a pH range between 7 and 10 for Sc, Pc, Fc1 and Fc2 and between 8 and 10 for St. All the curves for the five coagulants have a similar pattern. Sc, Pc, Fc1 and Fc2 have a broader optimum pH range compared to St. The maximum COD removal were 83.9, 74, 73.3, 69.1 and 67.5 % for Pc, Fc1, Sc, Fc2 and St respectively at a pH 10 corresponding to the dosage of 200 mg/1 for St, Fc1 and Fc2, 100 mg/l for Sc and 25 mg/l for Pc. The rate of COD removal decreased if the pH was lower than 7. In all cases at low pH values the performances are lower and they increase to maximum values for a determined pH or pH range.

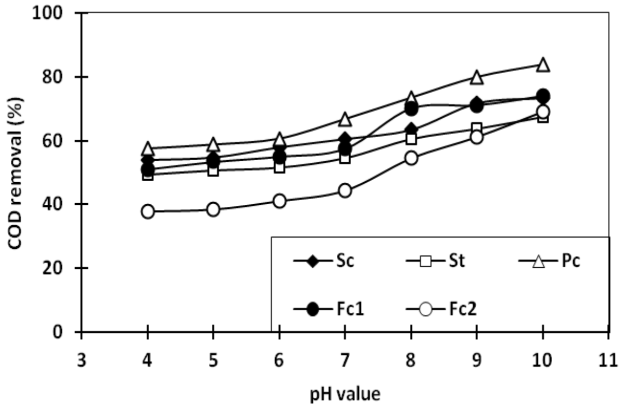

Figure 6. Effect of pH at optimum coagulant dosages on COD removal.

Numerous researchers have identified coagulation pH as the parameter having a great effect on achieving optimal organic precursors removal by coagulation process (Randtke, 1988; Bell-Ajy et al., 2000; White et al., 1997). The results of our study support the findings of literature, indicating that the pH of coagulation rather than the coagulant dosage was the determinant factor for COD removal. In an acid environment, aluminum of clays exists mainly in the form of Al_3^+, which is not conducive for the adsorption of colloid, adhesion, bridges and cross-linking, reducing the coagulation efficiency. On the contrary, in an alkaline environment, it can adsorb onto the surface of colloidal particles, promoting the colloid aggregation. However, when it exceeds a certain value, $Al(OH)_3$ will be generated which has a negative impact on coagulation efficiency (Huaili et al., 2011). Sc and Pc contain 22.3% and 10.2% Al_2O_3 respectively, hence its effectiveness in an alkaline environment in the removal of organic matter.

The effects of pH on percentage removal of TSS from jar tests for coagulation of domestic wastewater using Sc, St, Pc, Fc1 and Fc2 are shown in figure 7. It shows that pH is a predominant factor affecting the removal of TSS. It can be seen that TSS removal reaches its maximum at pH 7-10 for St, Pc and Fc2 (up to 98.9 % for Pc). However, for Sc, the removal of TSS is slightly affected by pH of the solution, therefore; in this case the coagulation–flocculation process takes place with relative independence of the pH. For St, Pc, Fc1 and Fc2 at low pH values the performances are lower and they increase to maximum values for a determined pH (7-10).

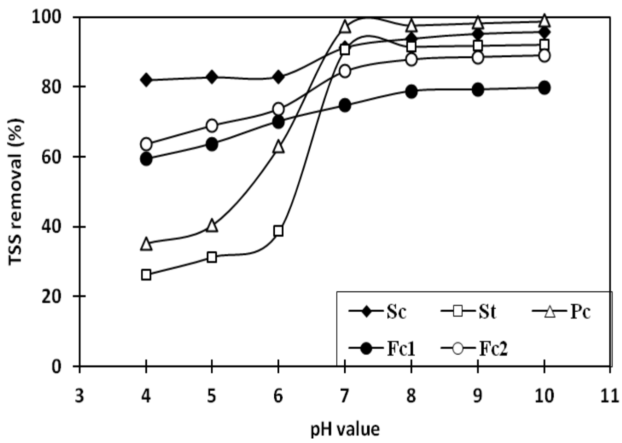

Figure 7. Effect of pH at optimum coagulant dosages on TSS removal.

For the 4 coagulants (St, Pc, Fc1 and Fc2) low pH values adversely affected the performance characteristics of coagulation-flocculation. In the case of Pc, within the range of pH studied, the removal efficiency of the TSS initially increased up to pH 8 where performances are at a maximum and from which stabilizes. When Fc1 and Fc2 are used, the elimination efficiency of TSS was very similar. Its use led to higher performance except at the lowest pH value studied (pH 4-6) where performances are slightly higher. For coagulants Pc, St, Fc1 and Fc2 can be used for the treatment of wastewater with a pH neutral and alkaline (8-10). Therefore, Sc can be used for any type of wastewater. For all coagulants, extending the optimal pH range allows efficient performance in the face of changing effluent characteristics or any pH variations that may occur during the coagulation–flocculation process and avoid the need to adjust pH.

3.4.2. Effect of pH on sludge volume

The interrelationship between pH and sludge volume was studied. We chose the concentrations of 25 mg/l of Pc, 100 mg/l of Sc and 200 mg/l of St, Fc1 and Fc2 for the determination the settling time at different pH. The settled sludge volume is highly influenced by pH values. With increasing pH (from pH 4 to 10), the percentage sludge volume increased considerably, giving a maximum for each coagulant at pH 10. Pc gives the sludge volume most important than Sc, St, Fc1 and Fc2 at pH 10.

In general, the amount and characteristics of the sludge produced during the coagulation–flocculation process were highly dependent on the coagulant used and on the operating conditions (Szyguta, et al., 2009; Amuda & Amoo, 2007). In the coagulation–flocculation process, the settling time is important since this will influence the overall cost and efficiency. The settling of the particles as function of flocculant obtained with the optimum operating protocol (dosages) allowed the determination of the settling time. The minimum settling time for each coagulant was obtained at pH 10.

3.5. Coagulant cost

The performance of Pc was found to be better than Sc, St, Fc1 and Fc2. However, the removal rates in COD and TSS increasing order for all the coagulants were Pc > Sc > St > Fc1 > Fc2. The proper determination of coagulant type and dosage will not only improve the resulting water characteristics (COD and TSS removal), but also decrease the cost of treatment.

Table 5, to optimal dose for each coagulant, shows cost of the different clays used. Of the five coagulants studied the cheapest is Sc (0.17 €/kg without counting transportation costs from natural mine to the treatment facility of wastewater). Furthermore, the dose of Sc, St, Pc, Fc1 and Fc2 to remove 1 kg of COD was respectively: 207, 459, 47, 434 and 562 g. To remove 1 kg of TSS was: 306, 615, 72, 746 and 660 g, respectively.

Moreover, even if the Sc has the lowest cost (0.035 € to remove one kg of COD by coagulation-flocculation) compared to St, Pc, Fc1 and Fc2, Pc has been selected (0.051 €) to avoid handling and storage of large volumes of clay at the treatment facility and particularly avoid high production of clayey sludge (to remove 1 kg of COD, we need 0.207 kg Sc while only 0.047 kg of Pc are sufficient).

Table 5. comparison of coagulants cost.

Coagulant	Clay of Safi	Stevensite	Perlite	Clay of Fes 1	Clay of FeS2	Lime	$Al_2(SO_4)_3$
Optimal dose of (mg/l)	100	200	25	200	200	100	200
TSS removed (mg/l)*	327	325	349	268	303	356	356
COD removed (mg/l)**	484	436	534	461	356	393	429
Amount of coagulant to remove 1 kg of COD (kg)	0.207	0.459	0.047	0.434	0.562	0.254	0.466
Cost of 1 kg of clay (€)	0.170	0.64	1.10	0.180	0.180	0.18	0.231
Cost of 1 kg COD removed (€)	0.035	0.293	0.051	0.078	0.101	0.046	0.108

* TSS of raw wastewater = 358 mg/l. ** COD of raw wastewater = 800 mg/l.

Table 5 indicate also differences in removal values for TSS and COD between the coagulation tests conducted with various clays (Sc, St, Pc, Fc1 and Fc2), lime and $Al_2(SO_4)_3$, especially that at maximum removal levels the values tend to overlap. These results lead to the conclusion that these coagulants (clays and chemical products) can work successfully in the coagulation of domestic wastewater. Though chemical coagulation by alum and lime may be a method of choice for treating wastewaters before their discharge, it too has its drawbacks as the effectiveness is strongly pH dependent (lime: alkaline pH, $Al_2(SO_4)_3$: pH acid) and finished water may have high residual aluminum concentrations, also in sludges and significant amounts of sludge are produced which complicates handling and disposal procedures. Further in many developing countries, the cost of importing alum and other required chemicals for conventional treatment might be high and at times prohibitive.

4. CONCLUSION

Physicochemical treatment of domestic wastewater using the coagulation-flocculation process was conducted to achieve maximum removals of COD and TSS. This study has demonstrated that coagulation with Moroccan clays origin is an effective method to clarify domestic wastewater by reducing the COD and TSS of the wastewaters with low costs. Pollutant removal efficiency at various pH and coagulant doses was evaluated. Pc produced better results than Sc, St, Fc1 and Fc2 in terms of COD and TSS removal. The results show a significant reduction of waste pollution of about 66.8% for COD and 97.4% for TSS at 25 mg/l of Pc. Sc can be efficient and less expensive in comparison with Pc, but of technical point of view, Pc was selected.

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