Treatment of a Worn mineral oil Effluent by Electrocoagulation using aluminium Electrodes

Serge Raoul TCHAMANGO Dr
University of Ngaoundéré, tchamangos@yahoo.fr

God Promess LACTIO
Université de Ngaoundéré

Inès Estelle Nkenmogne KAMDEM
Université de Ngaoundéré

Martin Benoît NGASSOUM
Université de Ngaoundéré

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University of
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Abstract

Water pollution by oils is more and more a preoccupation because of its increasing occurrence and the hazard to human life and environment. Depollution of oily effluents can sometimes generate a secondary pollution (with chemical coagulation / flocculation processes) or be very costly (ultracentrifugation). In this work, electrocoagulation with aluminium electrodes was applied to the treatment of a worn mineral oil effluent. For this to be achieved, the influence of the pH and the current density on the turbidity and organic matter was first investigated. A liquid-liquid extraction was then carried out on the oil samples in order to isolate hydrocarbons and study their behaviour. Finally, these hydrocarbons were analysed by gas chromatography (GC-FID) to evaluate their removal efficiency. The results obtained showed that optimal conditions were as follows: initial pH=7 and current density = 10.98 mA.cm^{-2} . The results obtained with a supporting electrolyte concentration set at 0.5 mg.L^{-1} showed that optimal conditions were as follows: initial pH=7 and current density = 10.98 mA.cm^{-2} .) In these conditions, the turbidity was almost totally eliminated and organic matter removal efficiency was 93.30%. Analyses of the various chromatograms showed that 98.83, 99.56 and 93.02% respectively of aliphatic fraction, unsaturated hydrocarbons and polycyclic aromatic hydrocarbons (PAHs) were eliminated during electrocoagulation of the worn mineral effluent. However, the absence of regular peaks was observed on the chromatograms, and that was due to unidentified heavy PAHs. Thus, FID did not seem sensitive enough to identify and characterize PAHs compounds based only on retention.

Keywords

Effluents, Oil emulsions, Electrocoagulation, Aluminium electrodes, Hydrocarbons.

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1. Introduction

Oils constitute one of the most important hydrocarbon products in the modern world [1–3]. Generally, oil effluents come from refineries, agro-process; textile and leather industries, etc. [1,4]. Oil effluents can also be of urban origin, such as restaurants and waste inherent to human activities [5]. With the fast industrial development and the increasing urbanization during the last decades, the generation of worn oil effluents has largely increased [6]; thus in 2012, one considered the global amount of oily wastewater at approximately 9–14 billion cubic metres [7]. These oils can be released in the environment during the production, the transport, the refining process and their multiple uses, involving films formation and deposits on rivers and seas surfaces. This situation endangers the watery system and human health [4, 8, 9]. Because of their low biodegradation in the natural ecosystem, oily components can deteriorate biological functions for a long time.

Oils are present in water in three ways: immiscible mixtures, unstable oil/water emulsions and very stable secondary oil/water emulsions. In the first two cases, the treatment of the whole as an effluent is easy with simple mechanical or physicochemical processes. On the other hand in the case of secondary emulsions, these methods cannot be applied any more. For instance biological degradation, although being an eco-friendly process, is very slow when degrading hydrocarbon effluents and the treatment requires long reaction times, in the range of days [10]. In water containing oils and greases, the depolluting microorganisms can be intoxicated by the unavailability of air, due to oil surface films formed. The purpose of the chemical processes of treatment is to neutralize acid or alkaline effluents using chemicals, either by precipitation or flocculation to induce the elimination of the pollutants. However, the excess of reagents remaining in the treated effluent can in certain cases involve a secondary pollution, and seriously restrict its aptitude for industrial re-use [11]. Ultrafiltration is a very promising for the removal of oil from water, but it has been limited by economic obstacles due to severe membrane fouling [12,13].

With respect to new environmental standards on the aqueous wastes rejections, it becomes necessary to develop less expensive and efficient techniques, which are not hazardous for the human being and the

environment. During the last decades, electrochemical treatment processes have been more requested. In fact, these processes can mitigate the insufficiencies of the traditional physicochemical processes of water treatment. Electrocoagulation, an electrochemical process, is the electrolytic version of the chemical coagulation. This electrolytic process consists in the electro-generation of metallic ions when a current is imposed in a medium containing soluble aluminium or iron electrodes. At the anode, these electrodes release metallic ions (Fe^{2+} , Fe^{3+} or Al^{3+}) that combine then with hydroxyl ions generated at the cathode, leading to the formation of aluminium or iron hydroxides which favour flocks constitution [14–18]. Although the mechanism of electrocoagulation is similar to that of chemical coagulation, the cationic species responsible for the colloids charge neutralization and the coagulated flocks differ in both processes: during electrocoagulation, the coagulants (iron or aluminium ions) are gotten *in situ* by anode dissolution, which is not the case of the chemical coagulation where these ions are produced by iron or aluminium salts dissolution, accompanied by the liberation of counter ions [19–21].

Electrocoagulation has these interesting advantages with respect to chemical coagulation [22,23]:

- i) electrocoagulation makes it possible to treat the very small colloidal particles, contrary to chemical coagulation where these particles impose very slow stages of treatment and high quantities of coagulants;
- ii) sludge obtained are denser and less absorbent, this makes easier flotation and decantation; thus their volume is considerably decreased;
- iii) in chemical coagulation, the pH must be adjusted for the coagulation to occur whereas in electrocoagulation, water reduction at the cathode gradually generates hydroxyl ions which coalesce with the metal ions to produce metal hydroxide, that at the end of the process eliminate the pollutants;
- iv) the electrodes costs (aluminium and iron) are lower compared to iron and aluminum salts used during chemical coagulation, and which induce a secondary pollution. Nowadays, electrocoagulation is in full flight and various reactors were patented [24–27].

The mechanisms of a classical electrocoagulation are illustrated on the figure below, where M represents aluminium or iron.

When an electric field is imposed starting from a DC power supply, the dissolution of the metal M (electro-dissolution) occurs at the anode to release the metallic cations (M^{n+}) while concomitantly at the cathode, water reduction generates hydroxyl ions. These ions (M^{n+} and OH^-) combine then to form metallic hydroxides ($M(OH)_n$) which will fix the pollutants by inducing the flocks formation. These flocks will settle at the bottom, or float on the surface with the upward movement of the gas evolved at the electrodes (electro-floatation).

Various reactions occur when aluminium electrodes are used. At the anode, metal ions are produced following the reaction [28]:

In addition at the same electrode, oxygen formed can compete with aluminium dissolution [28–31]:

At the cathode, hydrogen formation that takes place helps in the floatation of the flocculated particles out of the water [29–32].

In the solution, the following reactions occur respectively in acidic and alkaline medium [31,34]:

At high pH values, hydroxyl ions generated at the cathode during hydrogen evolution may attack the cathode by the following reaction [32,35]: $Al + 3H_2O + OH^- \rightarrow Al(OH)_4^- + \frac{3}{2}H_2$

When there are chlorine ions, these following reactions take place:

Thus, Cl_2 and OCl^- can be produced from the anodic reactions of chloride ions according to Eqs. (7) and (8) [36]. Hypochlorous ion OCl^- is a strong oxidant, which could oxidize some of the organic molecules present in wastewater.

Aluminium and hydroxide ions released at the electrodes can react to form monomeric and polymeric species $Al(OH)^{2+}$, $Al(OH)_2^+$, $Al_2(OH)_2^{4+}$, $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}(OH)_{34}^{5+}$, $Al_{13}O_4(OH)_{24}^{7+}$, which are finally transformed into $Al(OH)_3$ [31,32].

This work investigates the treatment of worn mineral oils of a road conveyors company installed in Ngaoundere (Town of Adamawa region in Cameroon). These initially untreated effluents infiltrate the soils and are found in the surrounding streams located close to the husbandry places. Thus, animals can be contaminated, and this contamination can reach human beings through the food chain. Previous works showed that hydrocarbon effluents were successfully treated by electrocoagulation [10,37,38]. However, to the best of our knowledge no dedicated work to electrocoagulation of oily effluents in sub-Saharan Africa was found, from where the choice to apply this process to the treatment of oily effluents of this company.

2. Materials and methods

All the chemicals used were of analytical grade.

2.1. Electrochemistry

The electrolytic cell used in this study was a small parallelepipedic pyrex tank, with a total volume of 400 mL. The electrodes (two) were aluminium plates (dimensions: 10 cm × 4.5 cm × 0.2 cm). These electrodes were laid out in parallel to one another in the cell, and the inter-electrode distance was 2 cm.

The agitation of the effluent in the cell was assured by a Bioblock Scientific magnetic stirrer. The current was provided by a regulated JEULIN DC power supply (5A-30V) (see Fig. 1).

Fig. 2 presents a classical electrocoagulation cell with two electrodes.

2.2. Preparation of oil emulsions

Emulsions preparation was achieved according to Mohamed method [39]. The samples of the worn mineral oil effluents were taken and diluted with tap water in order to obtain an artificially polluted water/oil emulsions. The preparation of the emulsion consisted in measuring a desired volume of worn mineral oil, which was then gradually added to water in a conical flask, and the whole vigorously agitated after each oil addition until the disappearance of the droplets. This step continued until obtaining a homogeneous mixture. The preparation lasted between 45 min to an hour approximately according to the oil initial concentration. After the preparation, the oil emulsions solutions at different concentrations were allowed to rest during 24 h in order to determine the critical

Fig. 1. Description of electrocoagulation mechanisms.

micellar concentration, i.e. the concentration which makes it possible to have a kinetically stable emulsion. The stable emulsions being obtained starting from 0.1% v/v, this concentration was thus selected for forthcoming experiments.

2.3. Characterization of the hydrocarbons extracted from emulsions

For this to be achieved, the liquid–liquid extraction was used, with hexane as solvent. According to Brum et al. method [40], 300 mL of the crude or treated emulsion was poured in a conical flask, and 25 mL of hexane added to this volume. The mixture was agitated during 30 min on a magnetic stirrer and then transferred in a separating funnel where it was vigorously shaken for 3 min while degassing from time to time. The phases were consequently formed after 10 min and the organic phase recovered. This operation was

repeated three times. The extract obtained was concentrated using a rotary evaporator before being fractionated.

2.4. Fractionation of the hydrocarbons concentrated extracts

For this to be achieved, Moreda et al. method was used [41] to separate aliphatic and aromatic hydrocarbons by chromatography. The column was progressively filled as seen in Fig. 3. Once the chromatographic column furnished, it was pre-eluated with 10 mL hexane. After the deposit of the sample on top of the column, the first fraction (aliphatic hydrocarbons) was eluted with hexane (10 mL). Then, with 10 mL of solvent mixture hexane/ethyl acetate in the ratio 83:17, the second fraction (constituted of a mixture of unsaturated aliphatic hydrocarbons and polycyclic aromatic hydrocarbons) was collected. Finally, the last fraction containing only heavy

Fig. 2. Electrocoagulation cell.

Fig. 3. Fractionation column of hydrocarbons extracts.

aromatic hydrocarbons with high molecular weight was collected (after extraction in 20 mL of the solvent mixture hexane/ethyl acetate in the proportion 65:35). The various fractions were concentrated and dried at 70 °C, and taken again with 1 mL hexane before being analysed (see Fig. 4).

2.5. Treatment of the worn mineral effluent

In order to treat our emulsions, 300 mL of the prepared emulsion was put in the electrolysis cell. NaCl (0.5 g/L) was then added to increase the conductivity and reduce the cell voltage. Finally the electrodes were connected to the DC power supply.

2.6. Analyses

The pH measurement was carried out using a EUTECH 510 Bioblock Scientific pH-meter. The pH was adjusted at the desired value using 0.1 M solutions of NaOH and H₂SO₄. All the experiments were achieved at ambient temperature.

The turbidity measurement was done using a HACH turbidimeter.

The determination of the organic matter (OM) consisted in evaluating the quantity of oxygen used for the reduction of potassium permanganate by the organic matter present in the worn mineral oil effluent (NF EN ISO 8467 of July 1995) [42].

Analyses of the various fractions of hydrocarbons were achieved by gas chromatography. The chromatograph (SHIMADZU GC-14B) was provided with a flame ionization detector (GC-FID) and a non-polar capillary column (SUPELCO HP-5MS). One microliter of each fraction was injected into the chromatograph using a micro syringe to analyse the compounds present.

To proceed to hydrocarbons analyses, naphthalene (hydrocarbon of reference) solutions at various concentrations (g. L⁻¹) were used, and the surface of the peaks was then measured by chromatography. The equation of the calibration curve obtained ($y = 10361x - 1099$ with $R^2 = 0.965$) clearly indicated that the peak surface (y) and the naphthalene concentration (x) are linearly dependent.

The different steps of the treatment are summarised in the following figure

Fig. 4. Synoptic diagram of the treatment of the worn mineral study.

Table 1
Initial characteristics of the worn mineral effluent.

Parameter	Value
Turbidity (NTU)	819.33 ± 1.00
Organic matter (mg.L^{-1})	22.40 ± 0.23
Sodium chloride (g.L^{-1})	0.5
pH	6.30 ± 0.05
Aliphatic hydrocarbon fraction mixture contents (g.L^{-1})	9.43 ± 0.06
Aliphatic/Aromatic hydrocarbon fraction mixture contents (g.L^{-1})	23.24 ± 0.15
Aromatic hydrocarbon fraction mixture contents (g.L^{-1})	14.66 ± 0.22

3. Results and discussions

Before the treatment, the initial characteristics of the effluent were measured. The initial characteristics of the effluent are reported in Table 1.

3.1. Effect of the initial pH on the turbidity and the organic matter removals

The effectiveness of a treatment by electrocoagulation depends on the pH of the effluent

Fig. 5. Variation of the removal efficiencies of the pollution parameters against the initial pH. Electrocoagulation time = 15 min; oil concentration, $C = 0.1\%$ v/v; $I = 0.4$ A.

Fig. 6. Mineral oil effluent before and after electrocoagulation treatment.

Fig. 7. Variation of the organic matter as a function of electrocoagulation duration at different current densities. Oil concentration: $C = 0.1\%$ v/v; $\text{pH} = 7$; inter-electrode distance: $d = 10$ mm.

[30,32,43]. According to the nature of the effluents, the metal hydroxides which favour coagulation are formed at different pH, depending on the acid or alkaline behaviour of the medium. The pH also determines the nature of the coagulating species. To study the effect of this parameter on the treatment, the following pH

Fig. 8. Variation of the turbidity as a function of electrocoagulation duration at different current densities. Oil concentration: $C = 0.1\%$ v/v; pH = 7; inter-electrode distance: $d = 10$ mm.

values were imposed (3, 5, 7 and 9). The turbidity and organic matter (OM) removals were also studied as well as the pH variations when the electrolytic treatment occurred. These results are presented on the figure below, the electrocoagulation time being 15 min.

As seen on this figure, in all cases the final pH is higher than the initial pH. This is not surprising, owing to the fact that during electrocoagulation, hydroxyl ions continuously electrogenerated at the cathode contribute to increase this parameter. It is observed that while the initial pH increase until a value of 7, turbidity and organic matter removal efficiencies increase. For instance, respective turbidity and OM removals are 93.35 and 68.71% at pH 5. When the initial pH is 7, 99.88% turbidity and 91.07% OM are eliminated. Beyond this initial pH value (7), the decrease of the removal efficiency of both parameters is noticed. Thus, at pH 9 turbidity and OM removals are 93.07 and 67.72% respectively. This decrease is ascribed to the formation of aluminate ion $\text{Al}(\text{OH})_4^-$ following $\text{Al}(\text{OH})_3$ re-dissolution in the presence of an excess of hydroxyl ions. $\text{Al}(\text{OH})_4^-$ being a negatively charged and soluble species, could not fix the pollutants, but rather contributes to inhibit the electrolytic treatment according to the equation [31,32]:

Fig. 9. GC-FID chromatogram of the aliphatic fraction (a) before and (b) after electrocoagulation.

In all cases, the maximum of turbidity and organic matter removals are obtained at $\text{pH} = 7$, and this value is considered as optimum pH for the treatment.

After the treatment under optimal conditions, the impact of the turbidity removal is obvious, as observed on Fig. 5, where the effluent treated by electrocoagulation is clear compared to the initial one (see Fig. 6).

3.2. Effect of the current density

The current density determines the rate of electro-generated coagulant of the effluent to be treated, since the dissolved anode producing the metal ions will induce the coagulation of the oily particles dispersed in the solution. It was shown that the current density also controls the bubbles production rate and their size [44]. Moreover the bubbles size decreases with the increase of the current density, which is a benefit for electrocoagulation process [45]. Current density is the ratio between the imposed current and the electrode surface immersed in the

solution. To study the effect of this parameter on the electrolytic treatment, the current intensity was varied (0.2, 0.3, 0.4, 0.5 and 0.6 A) and the electrode area immersed in the solution maintained constant. This corresponded to the respective current densities 5.19, 7.79, 10.98, 12.98 and 15.58 mA cm^{-2} . Then the influence of the current density on the elimination of the organic matter (OM) and turbidity was studied, as depicted on Figs. 7 and 8, the electrocoagulation time being set at 30 min.

As shown on these figures, the turbidity and OM decrease with the increase of the electrocoagulation time and that of the current density imposed. In other words, the greater the current density is, the greater are turbidity and organic matter removals. This is explained by the increase of the rate at which gas bubbles are generated at the electrodes and the reduction in their size when the current density increases, thus involving a strong probability of these and the flocks to coalesce [39]. From $j = 10.98 \text{ mA cm}^{-2}$, the turbidity and OM variations

Fig. 10. GC-FID chromatogram of the mixture of aliphatic and aromatic fractions (a) before and (b) after electrocoagulation.

are not significant, whatever the current density. However, the turbidity elimination is faster than that of the OM. For instance, when $j = 10.98 \text{ mA cm}^{-2}$ (corresponding to $I = 0.4 \text{ A}$) after only 5 min of treatment, 92.74% of turbidity are eliminated against only 43.75% of the OM. After 15 min of electrolysis, the maximum removal efficiency of OM is 93.30% whereas turbidity is almost completely eliminated. The remaining organic matter at the end of the treatment could be due to dissolved organic matter contained in the effluent and which is not eliminated by electrocoagulation [31,32].

3.3. Results of hydrocarbons analyses

The calibration curve of a naphthalene solution was extended to the various hydrocarbons analyses. For this to be achieved, their elimination was evaluated by calculating the unresolved complex mixtures (UCM) surfaces of the chromatographs obtained. In fact, the

surfaces of the UCM depicted by the chromatograms are proportional to the hydrocarbons contents. Thus, the relation $\tau(\%) = \left[\frac{A_0 - A_f}{A_0} \right] \times 100$ (9) traduces the removal efficiency (removal percentage) of the concerned hydrocarbons during electrocoagulation. In this relation, τ is the removal efficiency of the UCMs, A_0 and A_f the humps areas of the UCMs before and after electrocoagulation treatment. The following Figs. 9–11 depict the chromatograms GC-FID) of the aliphatic/aromatic and aromatic fractions of the worn mineral effluent before and after treatment by electrocoagulation. The peaks observed at the beginning of the chromatograms represent the solvent.

As observed in these figures, all the curves (a) present chromatograms with large surfaces, whereas on the curves (b) the surfaces considerably decrease. Among the chromatograms of the influent worn mineral effluent, that of Fig. 10 (a) has a larger surface, while that of Fig. 9 (a) is the smallest. That shows that

Fig. 11. GC-FID chromatogram of the aromatic fraction (a) before and (b) after electrocoagulation.

initially the aromatic/aliphatic hydrocarbons fraction constituted the most abundant components and the aliphatic hydrocarbons fraction the less abundant ones in the worn mineral effluent. The significant surfaces decrease observed on the chromatograms (b) is ascribed to the elimination of the various hydrocarbons during electrocoagulation. The significant difference between the surfaces of the curves (a) and (b) undoubtedly indicates that after electrocoagulation, the hydrocarbons are highly degraded. Thus, the removal percentages obtained are 98.53, 99.56 and 93.02% for the aliphatic (n-alkanes), aliphatic/aromatic mixtures (unsaturated) and aromatic hydrocarbons fractions respectively. The fact that the aromatic fraction exhibits the smallest value of removal could be due to the presence of aromatic rings in its components that oppose a resistance to the treatment compared to the aliphatic ones. Because of the presence of very complex and heavy hydrocarbons, UCM obtained in this case become more intense after a retention time of about 18 min. Blumer et al. [46], showed that in the petroleum compounds, the UCM become intense from the carbons C_{12} to C_{22} and in certain cases, from the carbons C_{16} to C_{36} . The same authors mentioned that there are cases where the identification of hydrocarbon compounds is often very difficult, even almost impossible. This can happen on the chromatographic analysis of engine oils. The case of the studied worn mineral effluent is similar: no hydrocarbon compound was identified by the Kovats indexes [48]. Demirci et al. [49] also reported comparable results during their investigation for the treatment of petroleum refinery wastewater using chemical coagulants ($FeCl_3$, $Al_2(SO_4)_3$ and $FeSO_4$) and clays (coagulant aids). They achieved nearly 90% turbidity and COD removal with 100 mg L^{-1} clay together with 100 mg L^{-1} coagulant. However, according to us, the fact that the authors used chemical coagulation and clays for the treatment of these effluents increase significantly the treatment cost. Analyses of the pollution parameters and the various hydrocarbons fractions UCM in the studied mineral oil effluent amply shows the effectiveness of electrocoagulation to treat the recalcitrant compounds.

4. Conclusion

This study showed that the treatment of worn mineral oils effluent by electrocoagulation with aluminum electrodes was efficient. The initial pH of the effluent and the current density have a significant influence on the electrolytic treatment. In fact at optimum conditions, the turbidity was almost eliminated

and 93.30% the organic was removed. The results of the analyses of the hydrocarbons various fractions revealed that the aliphatic, the unsaturated aliphatic/aromatic compounds with weak molecular weight and the aromatic compounds with high molecular weight contents were also notably reduced, their respective removals being 98.53, 99.56 and 93.02%. The absence of the peaks on the chromatograms was ascribed to the fact that the worn mineral effluents studied was constituted by very complex hydrocarbons components, and thus difficult to identify by GC method. The implementation of electrocoagulation units in the petroleum or other industries is to encourage because the pollution parameter of the rejected effluent are significantly reduced to safe and accepted levels.

Declaration of Competing Interest

The authors declare that they have no competing interests.

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