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Electrocoagulation for Ammonium Removal in Nam Son Landfill Leachate

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Abstract: In this paper, an electrocoagulation reactor was set up to investigate the ammonium removal in Nam Son landfill leachate. The research focused on studying several factors that affect to the ammonium removal namely current intensity, operating time, initial pH and electrode materials. A mono-polar electrocoagulation reactor was set up in a batch system with iron electrodes and 1.8 L leachate. The research indicated that current intensity and operating time are directly proportional with NH_4^+ treatment performance. When applied current increased from 1 to 4A, the NH_4^+ removal percentage went up from 14.03 to 24.99% after a 1 hour treatment. The effect of initial pH in range of 5 to 10 has showed that the best NH_4^+ treatment efficiency in neutral and mild alkaline conditions. It is noticeable that iron electrodes had higher NH_4^+ removal than aluminum one during nearly the first 40 min, however this trend has been reversed later with the advantage belonging to aluminum anode. The optimum operating conditions found are aluminum electrodes, applied current of 3A, electrolysis time of 60 min, raw pH of 8, resulting in NH_4^+ treatment performance of approximately 24%. As a result, the electrocoagulation method is not really effective in NH_4^+ removal and might be applied as a pre-treatment.

Keywords: Ammonium, electrocoagulation, landfill leachate, iron, aluminum, electrodes.

1. Introduction

Landfilling is one of the most popular methods of municipal solid waste disposal because of its relative simplicity and low cost. The degradation of the organic fraction of the municipal solid waste in landfill in combination with the percolation of rain water produces a liquid called leachate and it is highly toxic liquid with dissolving organic compounds, heavy metals and different soluble materials. The leachate composition is usually unstable and depends on many factors such as landfill age, type of waste, seasonal weather variations,

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precipitation level and landfill temperature. All these factors make leachate treatment difficult and complicate. Apart from popular methods such as adsorption, chemical precipitation, coagulation/flocculation, chemical oxidation and biological techniques, electrocoagulation is an alternative wastewater treatment that promises a large quantity of advantages namely short retention time, easy operation, simple equipment, performance and economic efficiency [1]. Therefore, the present research has studied application of electrocoagulation process on leachate treatment with iron and aluminum sacrificial electrodes. The series of experiments were carried out in a batch mode to evaluate the effect of different operating factors such as current intensity, electrolysis time, initial pH and electrode materials on ammonium removal efficiency.

In its simplest form, an electro-coagulation reactor is made up of an electrolytic cell with one anode and one cathode [2]. The principles of these methods are the in situ formation of the coagulants as the sacrificial anode corrodes and the simultaneous evolution of hydrogen bubbles at the cathode, resulting in the contaminants removal by electro-flotation. Moreover, electrocoagulation is known as an efficient technique since adsorption of hydroxide on mineral surfaces are a 100 times greater on "in situ' rather than on pre-precipitated hydroxides when metal hydroxides are used as coagulant [3]. On the other hand, because of no chemicals added to aqueous solution, it is environmentally friendly technique that does not cause secondary pollution. In addition, the smallest charged particles can be removed effectively compared to conventional chemical and biological techniques as the smallest charged particles have greater probability of being coagulated by the electric field that sets them in motion [3].

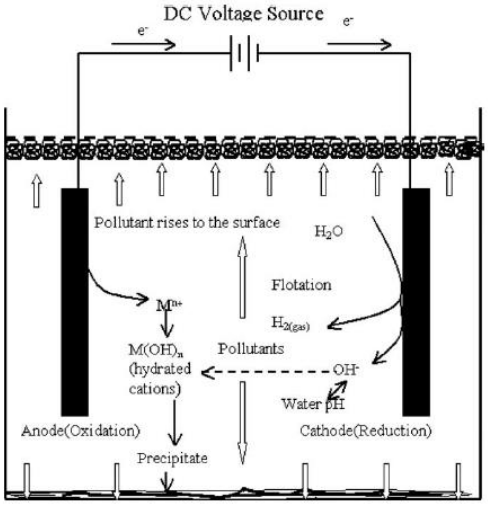

Figure 1. Several processes and reactions during Electrocoagulation [3].

There are two most widely-used electrode materials being iron and aluminum. For iron anode different mechanisms have been proposed based on the solution pH [4].

Mechanism I (acidic pH):

Anode:

$4Fe_{(s)} \rightarrow 4Fe^{2+}_{(aq)} + 8e^-$

$4Fe^{2+}_{(aq)} + 10\ H_2O_{(l)} + O_{2\ (g)} \leftrightarrow 4Fe(OH)_{3(s)} + 8H^+_{(aq)}$

Cathode:

$8H^+_{(aq)} + 8e^- \rightarrow 4H_{2\ (g)})$

Overall reaction:

$4Fe_{(s)} + 10\ H_2O_{(l)} + O_{2(g)} \rightarrow 2Fe(OH)_{3(s)} + 4H_{2(g)}$

Mechanism II (alkaline pH):

Anode: $Fe_{(s)} \rightarrow Fe^{2+}_{(aq)} + 2e^-$

$Fe^{2+}_{(aq)} + 2OH^-_{(aq)} \rightarrow Fe(OH)_{2(s)}$

Cathode: $2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} + 2OH_{(aq)}$

Overall reaction:

$Fe_{(s)} + 2H_2O_{(l)} \rightarrow Fe(OH)_{2(s)} + H_{2(g)}$

The Fe(OH)n (s) produced remains in the aqueous solution as a gelatinous suspension, which can remove the pollutants from wastewater either by adsorption, complexation

or by electrostatic attraction followed by coagulation.

With aluminum electrodes:

Anode:

$Al_{(s)} \rightarrow Al^{3+}_{(aq)} + 3e^-$

Cathode:

$3H_2O_{(l)} + 3e^- \rightarrow 3/2\ H_{2(g)} + 3OH^-_{(aq)}$

Overall reaction:

$Al^{3+}_{(aq)} + 3OH^-_{(aq)} \rightarrow Al(OH)_{3(s)}$

There are several monomeric and polymeric hydroxylated species formed and finally precipitated as $Al(OH)_{3(s)}$ which own a large specific surface area, allow rapid adsorption of organic pollutants and trap colloidal impurities inside. The flocks so formed either settle to the bottom or float on the surface by the evolution of H_2 bubbles [3, 5].

2. Materials and method

2.1. Objectives

The leachate samples were taken from Nam Son landfill being one of the most modern and famous landfill in Vietnam is located in Hanoi capital. Nam Son landfill was operated in 1999 with 83.5 hectares and expanded to 112 hectares in 2015. Currently, Nam Son landfill receives about 4300 to 4500 tons of waste per day and approximately 2000 m^3 leachate, yet only 1500 m^3 leachate is treated daily [6]. After collecting from Nam Son leachate reservoir, the samples were stored in obscurity at T=4 °C prior to the experiments.

Table 1. The NH_4^+ values of leachate in several landfills

No	Landfill	NH_4^+ (mg/l)
1	Nam Son landfill (7/2016)	1270 ± 38
2	Trang Cat landfill site (Hai Phong) [7]	104-620
3	Dong Ba landfill site (Quang Ninh) [7]	55-355
4	Go Cat landfill (Ho Chi Minh) [8]	3449 ± 233

As can be seen, the value of ammonium in Nam Son landfill leachate found very high, which outweighs leachate from other some landfills in the North Vietnam such as Trang Cat and Dong Ba landfill site. However, this value is smaller in Go Cat landfill leachate, Ho Chi Minh City, which may be caused by the different time in sample taking or landfill age or waste composition, weather condition and some other factors.

2.2. Electrocoagulation reactor

The experimental system is shown in Fig. 2. The electrocoagulation reactor was made of acrylic material with a dimension of 14 cm (with) _ 14 cm (length) _ 21 cm (depth) and the thickness of 1 cm. This experimental setup concludes 8 electrodes (11x10 cm) made of either iron or aluminum. All electrodes were connected directly with 8 electrode clips in which 4 are cathode and others for anode. A Direct Current (*Programmable PFC D.C.Supply 40V/30A, VSP 4030, BK Precision*) was used during the experiment. In this batch system, all the electrodes were submerged in approximately 1.8 liter leachate and the solution was agitated with a magnetic stirrer.

The effects of current intensity, operating time, initial pH and electrode materials on NH_4^+ removal were investigated throughout the experiment. The solution with flocks taken after each experiment was settle in 1 hour in the container before analysis. Only the limpid phase was used to chemical analysis.

Figure 2. The electrocoagulation reactor.

2.3. Chemical analysis

The ammonium concentration and pH values were determined by manual spectrometric method based on the TCVN 6179-1:1996, corresponding to ISO 7150-1:1984) and pH monitor (HANNA HI 991001), respectively. All the runs were carried out at room temperature.

3. Result and discussions

3.1. Effects of applied current and electrolysis time

Both applied current and electrolysis time play an important role in the electrocoagulation performance. Hence, this research focused on studying and assessing NH_4^+ removal efficiency by electrocoagulation method when the current intensity changes from 1 to 4A and the electrolysis time varies in range of 10 to 80 min.

One of the most important problems in leachate is nitrogenous compounds. The treatment of this type of wastewater is difficult due to very high ammonia content. The problems are especially related to fluctuations in nitrogenous compounds depending landfill age. As can be seen from Fig. 3, although electrocoagulation technique seems to work fairly-low, around 250-400 mg/l of in ammonia was removed to accomplish this removal efficiency. It is agreement with the research of Fatih et al, 2007 [9] which indicated a low NH_4^+ removal efficiency, just around 10% after 30 min treatment. The treatment efficiency seems to increase considerably with increasing current intensity and electrolysis time. In the first 30 electrolysis min, there were about 10 and 18.5% ammonia removal with applied currents of 1 and 4A, respectively; then these figures increase continuously and gradually to approximately 14 and 25% at the operating time of 60 min.

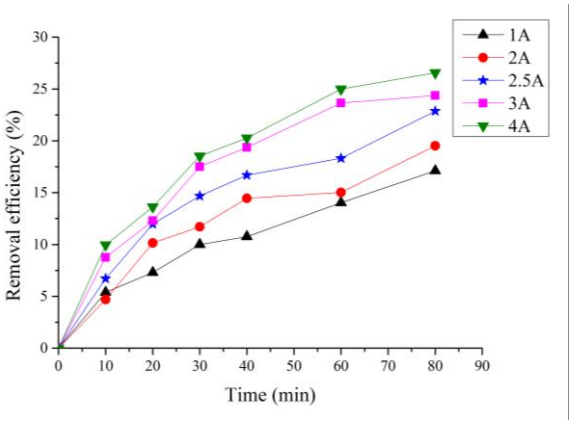

Figure 3. The effects of applied current and electrolysis time on NH_4^+ removal efficiency.

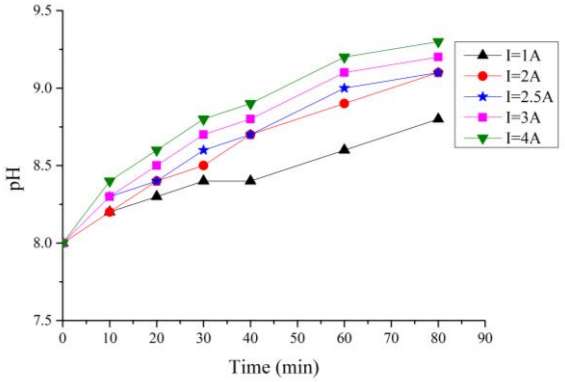

Figure 4. The variation of effluent pH during electrocoagulation process.

Moreover, it is obvious that the ammonia removal potentials with both applied currents of 3 and 4A, and electrolysis time of 60 and 80 min don't change considerably, just around 25%. Therefore, in order to save energy and reduce operating cost, the optimum intensity and electrolysis time for the following experiments were 3A and 60 min, respectively. This is explained by when applied current and time increase, the OH- group and H_2 gases created much more, leading to the increase in pH of medium (Fig. 4). Then, together with mixing process, much ammonium is converted to ammonia nitrogen is stripped with gases formed around the cathode and removed out.

3.2. Effect of initial pH

One of the most important factors influenced significantly on the electro-coagulation process is definitely pH [10]. In order to examine its effect, the initial pH of input leachate is adjusted to 5, 6, 7, 8, 9 and 10. The results of NH_4^+ analysis process is indicated in following Fig. 5 with constant operating time of 60 min and current intensity of 3 A.

It is evident that the electrocoagulation process works effectively in the range of pH from 6 to 8 in NH_4^+ removal, larger than 22%. The result has shown the similar trend with the research performed by Xiangdong Li et al, 2011 [11] which proved that the best NH_4^+ removal efficiency in range of pH from approximately 5.8 to 7.5. During the EC process, the change of pH during electrocoagulation process was observed when initial pH is either acidic or neutral. The interpretation for this phenomenon is that this increase is due to the release of over-saturated CO_2 because of hydrogen evolution at cathodes [12-14]. Hence, the NH_4^+ treatment performance increase remarkably as pH from 5 to 8, increasing from 14.33 to 24.89%, respectively. However, it is found that in the alkaline mediums, pH of the solution increases very gently or even decreases through the treatment, which due to precipitation of hydroxide ions with iron cations. This has explained why NH_4^+ removal efficiency reduced significantly in alkaline medium to only 11.23% at pH of 10 due to the reduction of ammonia in form of NH_3 gas. The results indicated that electrocoagulation can act as pH buffer. Fortunately, the raw Nam Son landfill leachate has the pH around 8. To take into account, it is not necessary to regulate raw water pH before treatment and the good pH value for the next experiments is 8.

3.3. Effect of electrode materials

Different anode electrode materials affect the performance of the electrocoagulation process. The most widely-used electrode materials are aluminum and iron due to their affordable price, readily availability, and effectiveness. In this study, the iron anodes and aluminum anodes were used in comparison on the same operating condition: 3A current intensity, raw leachate pH (8.0).

Overall, it is noticeable that the result is divided into 2 stages. The first period (from 0 to after 40 min) indicated that the ammonia removal efficiency by iron electrodes is higher from 1 to 3% than the one by aluminum electrode, and both have increased to around 18% after 40 min. In contrast, the latter period showed a higher NH_4^+ treatment yield by aluminum electrodes than iron electrodes, at about 30 and 25% after 80 min treatment time, respectively. However, the change in NH_4^+ removal efficiencies between iron and aluminum electrode is not really considerable; in particular, the gap between them is only from 1 to 5%. When optimum treatment time was 60 min, aluminum would be a better choice for ammonia treatment.

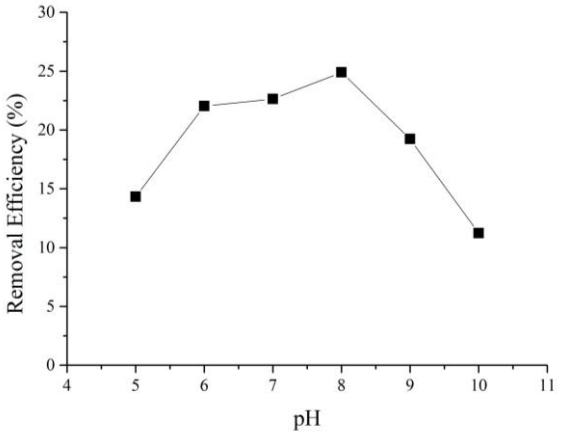

Figure 5. The effect of initial pH on NH_4^+ removal efficiency.

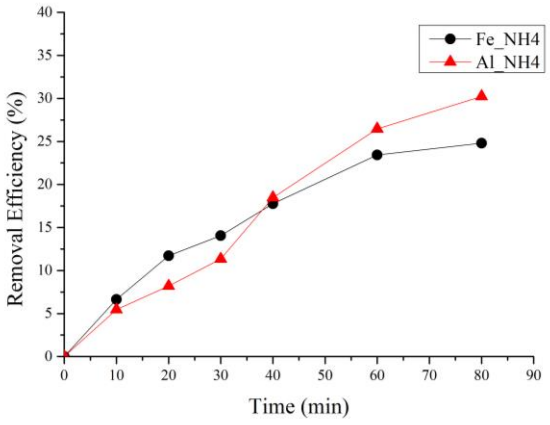

Figure 6. The effect of electrode materials on the NH_4^+ removal.

4. Conclusion

The study on NH_4^+ treatment performance from Nam Son landfill leachate by electrocoagulation method illustrated that with the input NH_4^+ concentration of approximately 1270 mg/l, the NH_4^+ removal efficiency is fairly low. With the increase in either current intensity or operating time, the NH_4^+ removal percentage increase; particularly this figure rose from 14.03 to 24.99% after 1 treatment hour with applied current of 1 and 4A respectively. The effect of initial pH on NH_4^+ removal shown that electrocoagulation performance ranged in pH from 6 to 8 and reached at pH of 8, this efficiency reduces significantly in both acidic and alkaline medium. Moreover, iron electrodes had much more benefit than aluminum one in NH_4^+ removal during nearly first 40 min, however this trend is reversed in the later. In conclusion, the optimum operating conditions found are aluminum electrodes, applied current of 3A, electrolysis time of 60 min, raw pH of 8, resulting in NH_4^+ treatment performance of approximately 24%. As a result, the electrocoagulation method is not really effective in NH_4^+ removal and might be applied as a pre-treatment.

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Loại bỏ amoni trong nước rỉ rác của bãi rác Nam Sơn bằng keo tụ điện hóa

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Tóm tắt: Trong bài báo này, một thiết bị keo tụ điện hóa đã được thiết kế để nghiên cứu loại bỏ amoni trong nước rỉ rác của bãi rác Nam Sơn. Nghiên cứu tập trung vào một số yếu tố ảnh hưởng đến việc loại bỏ amoni như cường độ dòng điện, thời gian điện phân, pH ban đầu và vật liệu làm điện cực. Thiết bị keo tụ điện hóa đơn cực làm việc ở chế độ theo mẻ với thể tích dung dịch nước rỉ rác là 1,8L, điện cực bằng sắt kim loại. Nghiên cứu chỉ ra rằng hiệu quả xử lý amoni tỷ lệ thuận với cường độ dòng điện và thời gian điện phân. Khi cường độ dòng điện tăng từ 1A tới 4A, hiệu suất loại bỏ amoni sau 1 giờ tăng từ 14,03% đến 24,99%. Kết quả cũng cho thấy hiệu quả xử lý NH_4^+ tốt nhất trong điều kiện pH trung tính và kiềm nhẹ. Đáng chú ý là trong khoảng 40 phút đầu tiên, các điện cực sắt đã loại bỏ NH_4^+ tốt hơn điện cực nhôm, tuy nhiên trong khoảng thời gian tiếp theo, điện cực nhôm lại xử lý NH_4^+ tốt hơn. Ở điều kiện tối ưu: điện cực nhôm, cường độ dòng điện 3A và pH8, sau 60 phút điện phân, khoảng 24% NH_4^+ đã bị xử lý. Như vậy, phương pháp keo tụ điện hóa không thực sự loại bỏ NH_4^+hiệu quả, tuy nhiên nó có thể được sử dụng như là một phương pháp tiền xử lý nước rỉ rác.

Từ khóa: Ammoni, keo tụ điện hóa, nước rỉ rác, sắt, nhôm, điện cực.