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Improvement of Sludge Reduction Efficiency of Ozonation by Microbubble Aeration Technology and Catalysis

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Abstract - *The sludge reduction by ozonation was improved through the catalyst and microbubble aeration technology. H2O2 was used to reduce the amount of ozone and increase the biodegradability of the sludge. The results showed that the biodegradability of sludge was improved. The oil content decreased from 77.48% to 51.7% after ozonation due to the ozone content, catalyzer and the time of ozonation. The MLSS and MLVSS of sludge were affected by ozonation and depended on microbubble aeration technology, they decreased from 53.54% to 31.42% and increase 17.15% to 36.5% during ozonation. The COD and NH4+ increased significantly by 220.8% and 26.2% after ozonation, but the increasing tendency of COD became very high 392.45% when the volume of H_2O_2 was above 34 ml. The H_2O_2 catalyst affected the pH of sludge during the ozonation to the neutral zone. The physical properties of sludge got modified by the microbubble aeration. The several experiences using different tubes <linear mixer> and <jet pip> showed the better one is Linear Mixer; it did a very important role in changing the bubbles on the top of the reactor and MLSS & MLVSS of sludge during the ozone treatment.*

Key Words: Ozonation, Active sludge, Biodegradability, Catalyst, Microbubble,

1. INTRODUCTION

The activated sludge generated by the exhaust system of wastewater from a petrochemical industry is a threat to the environment and to human health because of its dangerous nature; the minimization of that sludge is a promising strategy what meets development tendency. that can reduce the cost of sludge disposal and solve the problem of secondary pollution.[1]

The effective cleaning of that oily sludge has become a global problem. Several processes chemical, physical and biological and methods of treatments were studied recently to consider the reduction of the activated sludge, including: thermo chemistry[2], the microbial consortium[3], aerobic digestion [4], the thermal hydrolysis[5], mechanical disintegration [6], the ultrasonic [7], ozonation [8][9] and Microbubble applied to ozonation [10] . In the above methods, ozonation is one of the most promising methods as ozonation can reduce 50–100% of the activated sludge production without toxic products [11]. The ozonation method has been studied and approved as a promising way to reduce the sludge, in this study we combinate it with the microbubble technology and catalyst. The catalyzer was hydrogen peroxide H_2O_2 because of it oxidize characteristic

In recent years, microbubble technologies have drawn great attention in water and wastewater treatment due to its unique properties. Especially microbubble technologies can improve the gas-liquid mass transfer coefficient and increase solubility of gas [12], The use of microbubble encourages the stripping and chemical oxidation of the oil, it increases the efficiency of the reaction with high oxidation surface and allows a low ozone content to be used in field applications involving removal of mg/L sludge[13].

1.1 Microbubble Ozonation

Microbubbles are defined as bubbles with diameters less than several tens of micrometers. Compared to common bubbles, which have diameters of several millimeters, microbubbles have a huge interfacial area and bubble density, low rising velocity in the liquid phase and high inner pressure [13]. Our previous work has shown that in addition to the enhancement of mass transfer, microbubbles, which have higher inner pressure, can accelerate the formation of hydroxyl radicals and hence improve the oxidation of dye molecules [14].

The application of microbubble technology in ozonation processes may thus provide a low cost and effective technique for sludge reduction [15]. We used the microbubble aeration technology to remove the oil in the sludge, increase its biodegradability, mineralize the waste and reduce its final mass.

1.2 Objective

The general objective of this paper was to improve the microbubble ozonation technology, the application of a catalyzer H_2O_2 to use a low amount of ozone, also the mechanisms of reduction of activated sludge by micro-ozone bubbles was also discussed. Prior to different physicochemical factors such as the efficiency of ozonation by microbubble Aeration technology, the properties of different pipes and the efficiency of H_2O_2 catalyst. Therefore, we analyzed the influences of ozonation on the physicochemical characteristics of the sludge such as Oil content, COD, NH_4^+, MLSS, and MLVSS. The properties of two different pipes linear mixer and jet pip and their efficiencies during ozonation,

2. MATERIALS AND METHODS2

2.1. Experimental set-up

The equipment used in this study is a conception of our own design in order to bring a selective and distinguished improvement for production of more micro Nano bubble to reduce oily sludge.

Ozone was produced from pure oxygen by an ozone generator (JET-CFG Made in China). The sludge was ozonated inside a metallic cylindrical reactor of ozonation without ozone excess absorption which has a height of 150 cm and a diameter of 12cm with a total volume of 15L (Own design made in China) (Fig.1). Ozone was circulated through a small porous polymer tube connected to the dissolved gas pump (TYPE: YS7122 Eco Design China). Ozone from the dissolved gas pump goes through a mixer tube (pipe). We used two models of mixer tubes the Linear Mixer made by "OHR Technology" (Original Hydrodynamic Reaction) (Japan) and the second one was JET PIP Made by Wenzhou Lige Fluid System Co., Ltd. (China).

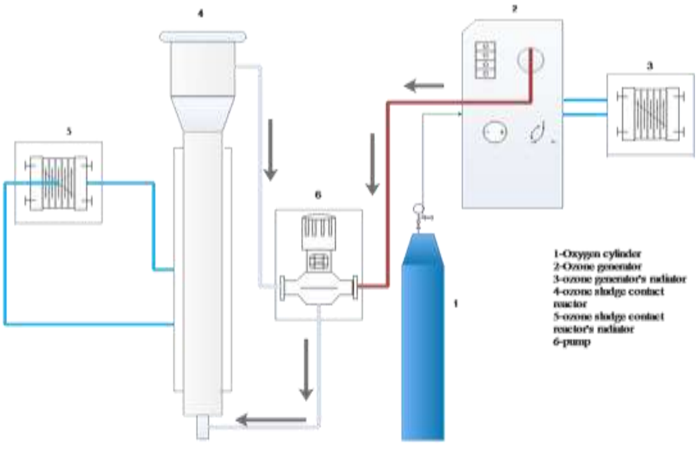

Fig -1: Experimental equipment for sludge ozonation

2.2. Sludge Source

The sludge used in these experiments were collected in the disposal system of Waste treatment of an oil refinery in Tianjin, a city located an hour from Beijing, China. A sample collected of activated sludge was precipitated about 14 hours to get a deposit at the bottom of the cans. The water above the deposit has been removed, the sludge turn was transferred to a vented container. The concentration was 6823 mg l^{-1}. A sample of 100 ml was used to determine the concentration analysis of the other compositions before the ozonation. This sample has been filtered through a filter paper to obtain a deposit of sludge that deposited in an oven at 105 ° for 72 hours to determine the initial characteristics of the Sludge see (Table 1)

Table 1: Characteristic of the active sludge

Parameter	values
pH	7.4 ± 0.44
MLSS (g /l)	1.68
MLVSS (g/l)	0.541
NH_4^+	18
Oil contains	263.6955

2.3. Operating Conditions

15 l of diluted sludge was fed into the ozonation reactor for a treatment. A sample of 200 ml was taken from the reactor every 20 to 30min during 120min before starting the analysis. We dealt with the sludge for each sample. 15L of the sludge introduced into the ozonation reactor was catalyzed by a volume of 33.4ml of H2O2. The hydrogen peroxide was added to, 5 min before the start of ozonation. 200 ml treated sludge were taken every 20 min for the first 60 minutes of treatment and then every 30 minutes to monitor the oxidation over time. For each collected sample was 200ml ozonized sludge and been analyzed as above. (Fig 2) presents the analysis made on each sample Several experiments were performed at different doses of ozone, (0.35 g O_3 g^{-1} TSS) (0.40 g O_3 g^{-1} TSS). The experiment was performed with the different pipes of the microbubble generator the results been discussed

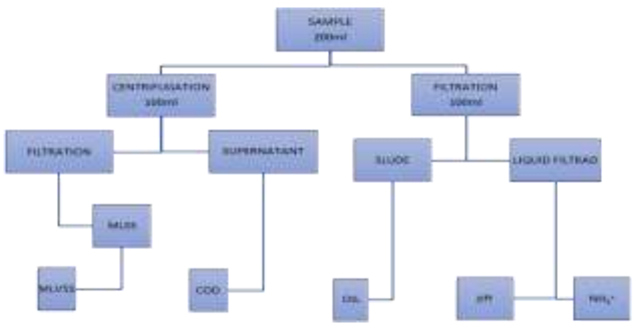

Figure 2: Physicochemical Characteristic Index

2.4. Analytical Methods

COD, NH_4^+, mixed liquor suspended solids and mixed liquor volatile suspended solids in this study were measured according to the Chinese NEPA standard methods [16]. The concentration of ozone in the gas phase was analyzed by the iodometric method with KI solution (Pires and Carvalho, 1998). The pH of the sample was monitored with a pH meter PHS-25 Chinese model MLSS, MLVSS were obtained by centrifuging, the ozonated sludge at 5,000 rpm about 15 min in the centrifuge (TD5). 100 ml of ozonized sludge was centrifuged and then sieved through a 0.35µm membrane of cellulose acetate and dry in an oven at a temperature of 105° for 72 hours and weighed to determine the MLSS, the same dried sludge been deposited in a muffle furnace at 600 ° for 120 min to calculate the MLVSS according to the Chinese NEPA standard methods [17][18].

The pH of the sample was monitored with a pH meter PHS-25 Chinese model

NH_4^+ was determined in the filtered liquid of each sample by Nessler's reagent colorimetric Method [1996 Chinese book] and calculated by the machine [Raster Spectrophotometer 722] a Chinese model. OIL was measuring from the filtered liquid through the membrane 0.35µm of cellulose acetate and dried in an oven at a temperature of 105 ° for 4 to 5 days and then the sample is crushed and washed with CCL_4 with the method:(ASTM D3921-96 (Reapproved 2003) GB / T16488-1996 The standard test for oil and Grease infrared spectrophotometry.GB3838-2002 Surface Water Quality Standards) and analyzed by the machine SYT700 Infare spectroscopy Oil Content Analyzer

3. RESULTS AND DISCUSSION

3.1. Performance of Microbubble Generator in Ozonation System

In the microbubble system, the smaller bubbles, which exist in great amounts, might rupture much faster and produce a higher speed jet, so that the associated mechanical stresses are likely to be[16]. The application of microbubble technology had brought a significant result in the sludge reduction, microbubble assisted the ozonation process successfully to simulate the flow of the sludge. The above Searcher reported that the microbubbles do contribute to sludge destruction and solubilization, as shown by the fact that 11% of sludge solubilization efficiency was reached in the temperature-controlled microbubble system. The microbubble generator adjusts conveniently the ozone amount from 0.45mgO3/ss to 0.35mgO/ss about a reduction of 22.22% ozone concentration what is a good result. Also reported the mass transfer coefficient of microbubble is 1.8 times higher than a macro bubble, while the pseudo-first order rate constant is 3.2-3.6 times higher. Also, TOC removal is about 1.3 times higher per g ozone consumed[19]. The used of microbubbles increase the COD 276%, NH_4^+ 26% in the same way [20] used microbubble ozonation and reported COD, NH_3 -N, and UV254 of the wastewater were removed by 42%, 21%, and 42%, respectively in the microbubble ozonation, while these removal rates by macro bubble ozonation were 17%, 12% and 7% in the same ozone dose

3.1.1. Efficiency of the pipe's Linear Mixer, the Jet pip and Direct Tube Use on Microbubble Generator

As innovation point, we used three types of pipes to the microbubble's technology aeration: the Linear mixer (Fig.3A.B) made by Original Hydrodynamic Reaction (Japan) ,the Jet pip (Fig.3C) made by Wenzhou Lige Fluid System Co., Ltd. (China) and the Direct Tube pipe (Fig.3D) according to the data from the experiment the tube direct didn't give a better result.(Fig.4D)

In order to choose between the linear mixer and the Jet Pip, we assume that both pipes have exactly the same microbubble characters but the design generates showed the different results (Fig.4ABC) made we chosen Linear mixer. Widely used method, the ozone generated by Linear mixer created a high pressure with a speed to inject ozone on the sludge; compare to the Jet pip as result (Fig.4A) showed the MLSS from linear decreased 2 times for than Jet Pip and MLVSS from Jet Pip increased 2 times more than linear mixer. Due at the teeth helix of linear mixer, supersaturated water is created at high pressure, the supersaturated gas is highly unstable and will easily escape out from the water when pressure decrease. As a result, a large number of microbubbles would be generated instantly. This kind of microbubble generation system usually contains a tank and a pump. The method[21] used is exactly this type.) no ozone gas bubbling tank, no was ozone decomposing, smaller ozonizer than the conventional ozone treatment method, in fact, the Jet pip did not have helix maybe that was the reason its result about the enhancement was not better as a linear mixer. The cost of Jet pip was a veritable economic good factor for this study[22][1].

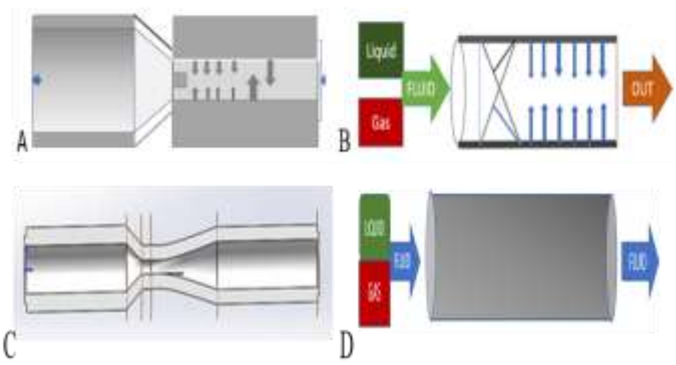

Figure 3: structure of pipes Linear mixer (A.B), Jet Pip(C), Direct Tube(D)

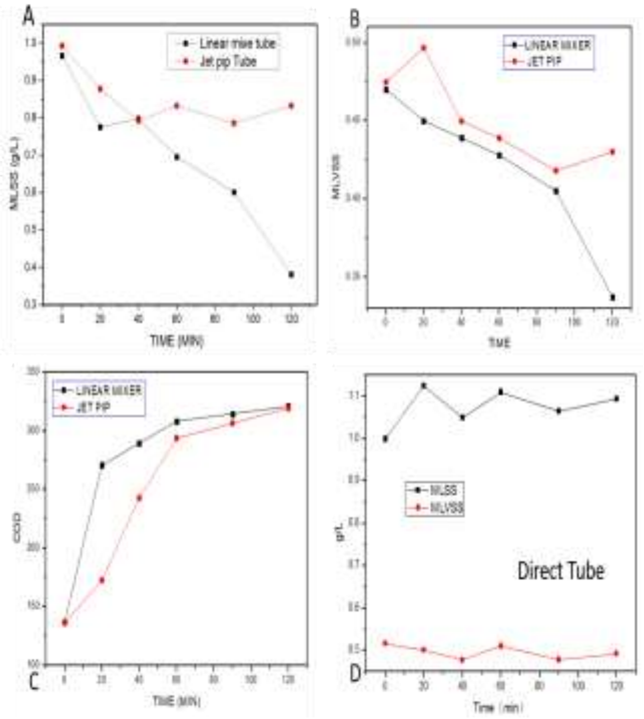

Figure 4: comparison between the linear mixer and Jet Pip MLSS & MLVSS, COD (ABC) Direct tube (D)

3.2. Effect of Microbubble Ozonation Catalyst on Mixed Liquor Suspended Solid (MLSS) and Mixed Liquor Volatile Suspended Solid (MLVSS)

The different ozone doses without the catalyst or with the catalyst H_2O_2 all showed a prominent decrease in the mixed liquor. Ozone reacts quickly and destroys the microorganisms and organic in the activated sludge that reduce the mixed liquor. When the dose of O_3 passed 0.35 g O_3 g^{-1} TSS the reduction of mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) in the treatment of 120 min corresponds to 31.42% and 17.15% (Fig.5). For the same dose 0.45 g O_3 g^{-1} TSS with the addition of the catalyst was reduced (53.45% and 36.5%) (Fig.5).

For analytical deduction, the microbubble ozonation treatment could disrupt the structure of the sludge flocs and the transfer of solids to the soluble phase, which causes the decrease of the mixed liquor solids in suspension and MLSS the mixed liquor volatile suspended solid MLVSS. This deduction was verified solid mixed liquor suspended and mixed liquor decreased volatile suspended solid from 19412 ± 463 mg / L 16 800 ± 542 mg / L and 10,869 ± 254 mg / L to 9581 ± 340 mg / L. respectively[1]

When the ozone dose reached 0.35 g O_3 g^{-1} TSS, the mixed liquor suspended solids and the mixed liquor volatile suspended solids were all reduced approximately constant than the same of 0.35 g O_3 g^{-1} TSS with catalyst dose to disrupt the sludge. (Fig.5)

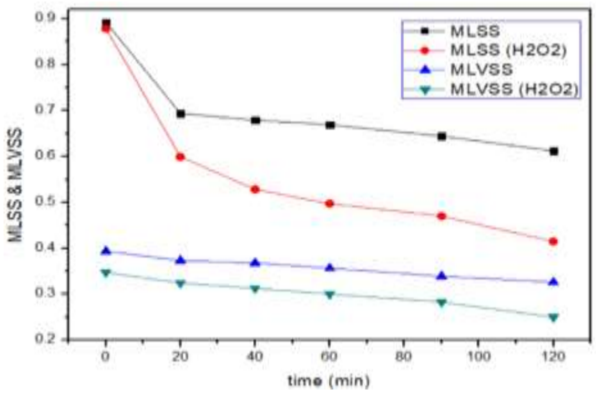

Figure 5: Effect of the microbubbles ozonation on MLSS & MLVSS

3.3. Efficiency of Microbubble Ozonation Catalyst on COD

Rapid injection of ozone at the beginning of reaction appears to be effective for the disintegration, ozonation can be determined by the increase in the value of COD during the experiment. This hypothesis has been supported by Sludge disintegration by ozone can be described as sequential decomposition reactions of the cell destruction, solubilization and subsequent oxidation of the released organics into carbon dioxide[19]. Ozone releases the soluble substances in the activated sludge which increases the COD, disrupts intracellular cell bodies and dissolution of sludge. By deducting the destruction of the organic cell in the activated sludge was the result of an increase in COD. Although comparison of the efficiency of sludge, in this study the COD increased 220.8% (Fig.6), two reasons were affected this result one was the property of H2O2 the second was the due of ozone-bubble amount, reported the efficiency of COD removal was 20% higher in the microbubble system. These results revealed that microbubble ozonation is a promising process in wastewater treatment H2O2 really affected the COD increasing which was 47.89% without catalyst with an ozone dose 0.35 g O_3 g^{-1} TSS. This hypothesis was based on previous studies for ozonation of activated sludge which reported. The value of SCOD in sludge mixture increased gradually along with ozone dose increasing, but its increasing rate decreased when the ozone dose was above 37.8 mg O3/g SS[4]

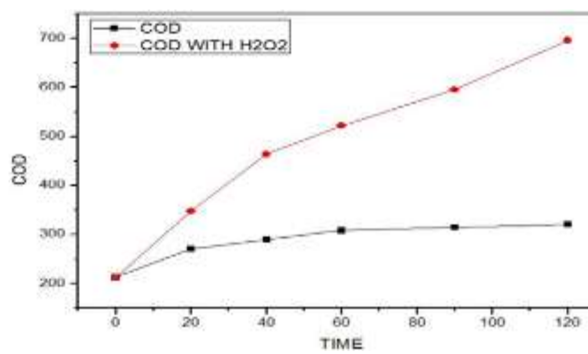

Figure 6: Effect of the microbubbles ozonation on COD

3.4. Efficiency of Microbubble Ozonation Catalyst on Oil Content

Oily sludge is considered as a hazardous solid waste, and its physical-chemical characterization is very complicated[6]. Immiscible pollution is due to the presence of oils and solvents used in industrial processes. Ozone is a powerful oxidizing gear to reduce the amount of oil in the activated sludge, in general, ozone stimulates the biological mechanisms that inactivate free radicals[15]. It exerts a cellular metabolism stimulus activity explains the associated revitalization effect Inhibits cellular immune reactions and Blocks tumor cell multiplication. Ozone has potent sterilizing activity by destroying bacteria, viruses, yeasts and parasites mud which causes a reduction in the amount of oil, according to the study reduced the amount of oil hardly dependent on others parameters like temperature, the microbubble, but it depends only on the introduced amount of ozone, catalyzer and time factor. For an amount of 0.30 g O_3 g^{-1} TSS, 0 min 120min we got 11.27% decrease, for a dose of 0.35 g O_3 g^{-1} TSS oil decreased about 17.48% (Fig.7) reported The results clearly indicate that oily sludge has a high percentage of hydrocarbons and it was 60.36 H_2O_2 catalyst may play an amplifier role of the introduced ozone dose that will give a good yield in a short time, in about 60min of H_2O_2 catalysis gives a satisfactory result decreased by about 51.7%. (Fig.7).

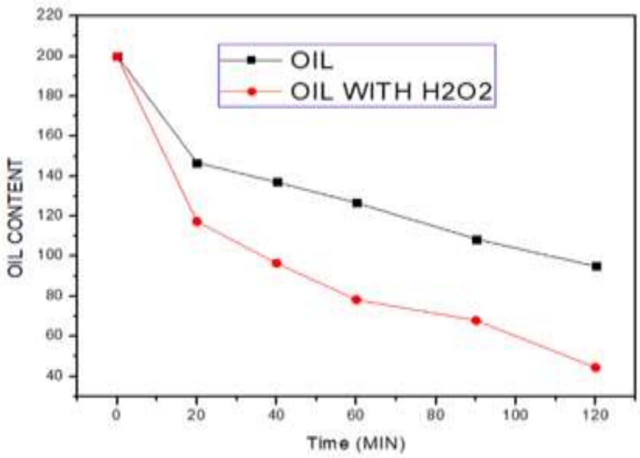

Figure 7: Effect of the microbubbles ozonation on Oil content

3.5. Efficiency of Microbubble Ozonation Catalyst on NH_4^+

Bacteria in the activated sludge were destroyed by ozone and the organic nuclei explode inside sludge, this explosion of the cell nucleus releases the DNA and protein which float in the slurry and increase the amount of nitrogen N. The nitrogen increases N increases and the NH_4^+ according to the reactions (ABC).

a- $NO + O_3 \rightarrow NO_2 + O_2$
b- $NO_2 + O_3 \rightarrow NO_3 + O_2$
c- $2\ NH_3 + 4\ O_3 \rightarrow NH_4NO_3 + 4\ O_2 + H_2O$

The catalyzer H2O2 increased the amount of NH_4^+ (Fig.8) and a bit disturbed and irregular, it was clear that a low amount of ozone was not able to destroy all nuclei of bacteria that float in the sludge and also the pipe Jet pip had no bacterial destructor propeller.

From a higher amount of O_3, the trend of evolution of the amount of NH_4^+ and was steadily increasing. The increase in nitrogen could be the following reason: The kernel is the main seat of the synthesis of DNA and RNA; destruction of microorganism's core causes the release of nitrogenous substances and intracellular proteins in the supernatant this assumption is affirmed. The Increase of nitrogen could Be the Following two Reasons: One Was That ozonation Could Destroy cytoderm of a microorganism resulting and in release intracellular of nitrogenous substances into the supernatant[11]. The other ozone was that which Could generate hydroxyl radicals, quite induced the ammonification of circumambient nitrogenous compounds, amino acids and Such As proteins. Previous research also says this: In the sludge ozonation may further oxidize a fraction of the released organic nitrogen[20]

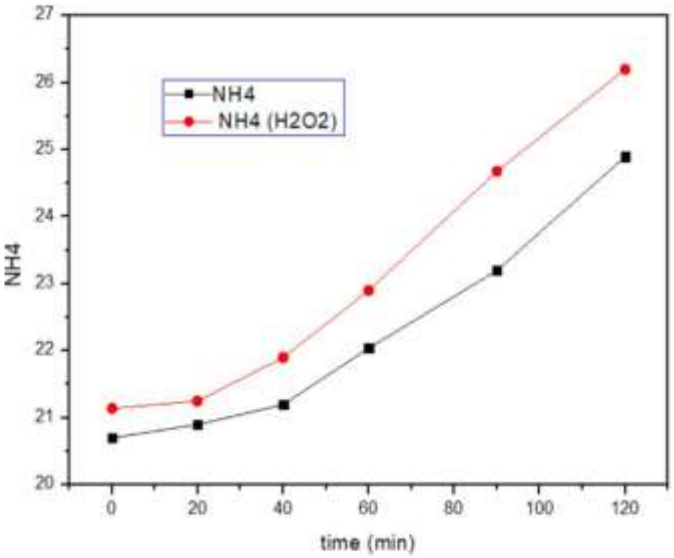

Figure 8: Effect of the microbubbles ozonation on NH_4^+

3.6. Effect of Microbubble Ozonation Catalyst on Ph

If the initial pH is greater than 7, the pH will tend to decline during ozonation due to the consumption of hydroxide ions (OH^-) in presence as in the absence of the catalyst H_2O_2.) the same been reported The inverse where the initial is inferior to 7, the pH will increase very slightly in p of H_2O_2 and in the absence of H_2O_2, the pH will still tend to decline[14]. This change in pH between the acidic medium and the basal medium is the migration of hydronium ions (H^+) where it is impossible to control the change of pH (Fig.9)

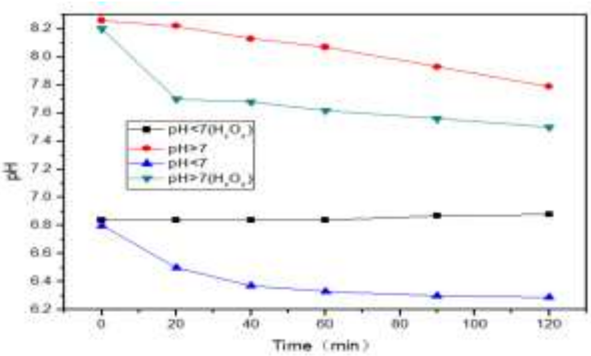

Figure 9: Effect of the microbubbles ozonation on pH

4. CONCLUSIONS

The reduction of sludge by ozone has already been the subject of much research in the world, particularly in the chemicals, environments laboratories of wastewater treatment. The use of microbubble aeration technology is an innovation for reducing sludge. this method creates a difference and yet provides a priori a number of benefits brought to ozonation as has been said in Result in and discussion.

Our problem in this study was thus to define: The reductional capacity of the activated sludge by the micro-ozone-bubbles and the efficiencies catalytic of hydrogen peroxide to enhance the solubilization of the organic and inorganic material from a sludge. The influence of certain process parameters such as the of the microbubble generator changed the physical properties of the sludge for a positive contribution on the solubilization capacity of the organic matter and mineral matter of the sludge.

The first results of the reduction by only ozone required a dose of 0.45 g O_3 g^{-1} TSS; MLSS decreased at 31.42%, MLVSS 17.15%, NH_4^+ increased to 20.29%, COD 47.89%, oil decreased at 51.7%. we noted a significant increase in the concentration of O_3 non-settling from the reactor. This result is negative because of the high dose of ozone (0.45 g O_3 g^{-1} TSS) not good for the environment it causes secondary pollution. The potential use of the microbubble (Linear Mixed pipe) and catalyzer hydrogen peroxide (H_2O_2) are very high for the main reason the ozone amount was reduced, the performance maximized. Indeed, the problem of secondary pollution was solved

The ozone from the dissolved gas pump into the mixer tube had a considerable effect on the bubbles, the sludge and ozone mixing process produces a maximum micro Nanobubbles.

The effectiveness of the use of the microbubble technology catalyzer hydrogen peroxide is ameliorating this result and minimize the issue of secondary pollution by reducing the specific amount of ozone from 0.45 g O_3 g^{-1} TSS to 0.35 g O_3 g^{-1} TSS and get a higher solubilization performance, the majority (85%) of the particulate organic material of the sludge can be solubilized showed: the COD increased about 220%, Oil content reduced to 77.48%, MLSS & MLVSS decreased 53.54% & 36%, NH_4^+ increased 25.35%, pH stabilized.

Finally, the use of a catalyzer and microbubble can reduce the Custom cost of the sludge reduction.

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