



In situ, one step removal of ammonia from onshore and offshore formation water of petroleum production fields



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HIGHLIGHTS

- Ammonia in petroleum formation water is a big environmental threat to living organisms.
- Electrochemical treatment could be used in situ for removal of ammonia.
- Ammonia was totally removed after electrochemical treatment.
- Other contaminants were obviously remediated.
- PV cells could be used in situ as the power source.

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ABSTRACT

Fish, crustaceans and other living organisms are threatened due to disposal of harmful contaminants in sea water. Ammonia is considered one of harmful contaminants due to industrial activities of oil companies, where excess ammonia in the formation water is discharged into sea water. Electrochemical treatment (EC) was used in one step for total removal of ammonia and remediation of other contaminants. Three working electrodes were examined EC cell, aluminum, iron and modified electrode (Ti/IrO₂). Graphite electrode was used as counter electrode in all processes of binary system. Both ionized and unionized ammonia of onshore (5.54 mg L⁻¹) and off shore (110 mg L⁻¹) were totally undetected after one step using all types of electrodes. The study was extended also to check the removal efficiency of other contaminants, where the analysis indicated the alleviation of them. Total suspended solid (TSS) of both onshore 64 mg L⁻¹ and offshore 228 mg L⁻¹ samples was reduced to 4 mg L⁻¹. Total dissolved solids (TDS), chemical oxygen demand (COD) and biological oxygen demand (BOD) of high values, 232,000, 8500 and 2442 mg L⁻¹ were also reduced to lower levels 18,400, 4000 and 1600 mg L⁻¹, respectively. The formed sludge after EC treatment was also investigated using XRD.

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1. Introduction

The quality of water is vital for human, animals and plants. Sea water is influenced by both natural processes and industrial petroleum activities (Dixon and Chiswell, 1996). Ammonia is one of harmful contaminants discharged into sea water as a result of oil industry either from onshore or offshore sites (Admiraal, 1977; Pearce and Whyte, 2005; Helmy and Kardena, 2015).

High level of ammonia causes toxicity of living organisms in sea

water and it is highly toxic to seawater at high pH in the presence of microalgae. Toxicity due to ammonia may be resulted from both ionized ammonia (NH₄⁺) and unionized ammonia (NH₃). Researchers have shown that the unionized ammonia is the most harmful species to sea water and threat fish (Durborow Robert et al., 1992). Ammonia limits for fish are narrow according to reports of the European Inland Fisheries Advisory Commission (Poxton and Allouse, 1982). Fish and other sea living organisms are influenced and a large variety of physiological disturbances was observed by excess ammonia (Jensen, 1995). Arising from the complex relationship between total ammonia concentration, pH and temperature; there emerges a level for total ammonia of around 0.3 mg L⁻¹ NH₃ (EPA, 2001). Also, ammonia is one of strong

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cell poison and can cause damage to sea living organisms. If its concentration gets high enough, fish will become inactive and eventually fall and die (Hargreaves and Tucker, 2004). Moreover, most fish produce ammonia as an end product of protein metabolism and it is excreted via the gills to the surrounding water. High levels of ammonia in the sea water either impair ammonia excretion or result in a net disposal of ammonia from the environment (Yuen and Chew Shit, 2010). In marine and estuarine fish, the toxicity of ionized ammonia NH_4 in sea water should be considered since the gills show some permeability to this ion and fish is considered at danger (EDDY, 2005). There are different factors which result in a negative effect of water quality but recently, the fast industrial development is considered as important factor which has a high contribution in water quality distortion. The negligence of wastewater treatment before discharging into sea water and also the entry of pollutants in sea water are considered the important reasons lead to sea water pollution. Oil industry is one of the main sources, which pollutes the environments (air, water and soil) and consequently, all living organisms are threatened by harmful contaminants such as ammonia (Cerqueira and Marques, 2012). Hazardous compounds of petroleum industry of nitrogen ingredient e.g. amino and nitro pyridine, methyl pyridine, ethyl pyridine are poor biodegradable, highly toxic and lead to environmental pollution. These compounds may produce ammonia under relevant conditions. (El-Ashtoukhy et al., 2013; Bashaa et al., 2008). In recent years, wastewater was discharged by industries which result in increasing TDS, COD, BOD and heavy metals in wastewaters. Different industries e.g. tanning (Ayoub et al., 2011), buttermilk and yogurt (Koyuncu et al., 2001), cheese (Monroy et al., 1995), distillery (Piya-areetham et al., 2006), textile (Ahn et al., 1999), food industries (Álvarez et al., 2011) and oil industry (Pichtel, 2016) are of different types of contaminants in wastewater.

The presence of high concentrations of ammonia in wastewaters will result in eutrophication spoilage, killing fish, and hindrance to the disinfection of water supplies, as well as an offensive smell and carcinogenesis (Larsen et al., 2007; Fernández-Nava et al., 2008). The environment was suffered from the emissions of industrial wastewater containing ammonia or nitrogen ingredients as a result of industrial development and human activities (Camargo and Alonso, 2006). Coking wastewater, generated from coke plants and coal gasification plants, contained high amount of ammonia and other hydrocarbons (Zhang et al., 2013a). Biological nitrification (Hou et al., 2014), electrochemical (Mook et al., 2012), adsorption (Markou et al., 2014), chemical precipitation (Li et al., 1999), microbial fuel cell (Zhang et al., 2013b), and ammonia stripping may be used for removal of ammonia from waste water (Zhang et al., 2012). The most common method for the treatment of ammonia from wastewater is biological nitrification, but it is not suitable for ammonia of high contents (Jiang et al., 2015). Still, electrochemical methods have been projected to have potential applications in wastewater treatment (Saracco, 2002; Jüttner et al., 2000). The electrochemical method has some important advantage as high treatment efficiency and relatively low investment costs (Wang et al., 2012).

The present study is aimed for characterization of both types of real formation water samples (onshore and offshore) produced during oil production from oil field located in Suez Gulf – Egypt. Formation water is a natural layer of water inside oil and gas reservoirs.

Also, this work is dedicated for finding out the most suitable process for the treatment and removal of highly hazardous ammonia and other contaminants in both onshore and offshore formation water. Finally, assessment of successful trials for removal of ammonia and remediation of contaminants was considered through measurements comprising, NH_3 , COD, BOD, TDS and TSS.

2. Materials and methods

2.1. Sample collection and preservation

A composite sample was collected by Central lab for industrial pollution studies, Tabbin Institute for Metallurgical Studies, Cairo. It was collected over a period of 24 h with sampling rate of 100 ml/15 min. The total volume is 9600 ml (9.6 L/24 h). The sample container was kept in icebox, so that the temperature of all samples was controlled at 4 °C during sampling and transportation from the oil field to the lab. The characteristics of raw samples of both onshore and offshore formation water are summarized in Table 1.

2.1.1. Sample location

Suez Gulf, Gabal El Zeit region in the Red Sea.

2.2. Equipment

Automatic sampler (ISCO 7600) was used for sample collection.

Electric DC power source (GW lab GPR-3030) was used as the source or direct electric current applied during electrochemical treatment of wastewater. Iron, aluminum and Ti/IrO₂ electrodes were used as working electrode into the electrolytic cell and connected to the positive terminal of DC Power, while graphite electrode was used as counter electrode and connected to the negative terminal of DC power. The following methods and equipment were used for analysis of raw water before and after electrochemical treatment to discern the quality of water and removal efficiency.

The concentration of ammonia was measured using the test ($\text{NH}_3\text{-N}$ SM 4500 NH_3C), where two apparatus were used Ion selective electrode ORION and Dual channel PH/Ion Meter Accumet Research, Fisher Scientific, USA Model AR 25.

BOD was measured using Dissolved Oxygen Meter with BOD Probe YSI Incorporated, USA Model 5100-230V S/N 15A 103,525 and BOD Incubator, Thermo scientific, USA Model 3730-146E S/N 300027894.

COD was measured using three devices Digital Dispenser 10 ml, Dispensette RS, Brand, Germany, Heater, Electro-thermal, UK S/N: 10953529 and Automatic Burette, DIGITRAT, JENCONS, UK.

Both TDS and TSS were determined using devices Iso-temperature Oven, Fisher Scientific, USA Model 738 F/S/N 126 and Analytical Balance: ADAM AFA Germany Max. 20 gm.

Electric conductivity was measured using Conductivity Meter, JENWAY, UK Model 4520 S/N 56,923.

Turbidity was measured using Turbidimeter, HF Scientific, Inc. USA Model Micro 1000, S/N 201503151.

As shown in Fig. 1, in situ a suggested electrochemical unit is divided into four main steps, first step is the collection tank of onshore or offshore wastewater from the sites of oil extraction. The second step is the clarification and removal of bulky materials to feed waste water to the main electrochemical unit (step 3). In this step, three types of electrodes, aluminum, iron and modified electrode (Ti/IrO₂) were examined and assessed for efficient removal and remediation of formation water. Graphite electrode was used as cathode in all processes. Each electrode was connected to DC power supply GW 30V- 3A. After EC treatment, suspended colloids were formed, so step 4 is used for filtration of water, where the removal of all suspended and precipitated sludge was carried out. Then, water was taken out for complete analysis and assessment of the removal efficiency.

A comparison between the cost of each type of electrodes was evaluated by passage of 1 A, where the external electric power was adjusted to pass the same electric current between the working and counter electrode through the wastewater bath. Using three different electrodes Al, Fe and Ti/IrO₂, the potential applied was

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