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## Hydrogen sulphide removal from the effluents of a phosphoric acid production unit by absorption into chlorinated seawater under alkaline conditions

*Élimination du sulfure d'hydrogène de l'effluent de l'unité de production d'acide phosphorique par absorption avec une solution chlorée et basifiée d'eau de mer*

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### ABSTRACT

Atmospheric pollution during the manufacturing of Wet process Phosphoric Acid (WPA) is a critical environmental issue. The aim of this study was to examine the absorption of hydrogen sulphide (H<sub>2</sub>S) emitted by this process into a chlorinated seawater solution under alkaline conditions. Parameters such as pH, sodium hypochlorite (NaOCl) concentration, gas flow rate, operating time and volumetric ratio of the liquid as well as gas phases (Q/Q) were investigated for their effects on H<sub>2</sub>S absorption efficiency. It was found that sodium hypochlorite is an effective and economical H<sub>2</sub>S oxidant. Moreover, according to the obtained results, a dose of 1 g Cl<sub>2</sub>/l and a pH of 11 are recommended. The preferred gas flow rate in this study was equal to 15 l/min. Under these conditions, more than 98% of the hydrogen sulfide was removed, even at relatively high H<sub>2</sub>S concentrations in industrial gas released from the plants of the Tunisian Chemical Group (GCT).

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### RÉSUMÉ

La pollution atmosphérique par le sulfure d'hydrogène issu de la fabrication de l'acide phosphorique par voie humide constitue une problématique environnementale critique. Le but de ce travail est d'étudier l'absorption de ce polluant par l'eau de mer chlorée et basifiée. L'effet des paramètres opératoires tels que le pH et la concentration en NaOCl de l'eau de mer, le débit de gaz, le temps de fonctionnement de l'absorbeur et le taux volumétrique ont été examinés pour évaluer leur effet sur l'efficacité d'absorption d'H<sub>2</sub>S. Les résultats ont démontré que, pour une dose de chlore de 1 g Cl<sub>2</sub>/l, un pH de 11 et un débit de gaz de 15 l/min, plus de 98% du sulfure hydrogène a été éliminé, même pour des

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teneurs relativement hautes en H<sub>2</sub>S. Ainsi, l'eau de mer chlorée et basifiée peut être considérée comme un agent efficace et économique pour dépolluer l'effluent gazeux industriel du Groupe chimique tunisien (GCT).

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## 1. Introduction

Particules, hydrogen sulfide, mercaptans and hydrogen fluoride constitute the major gaseous pollutants generated by the Wet manufacturing process of Phosphoric Acid (WPA). Those pollutants are frequently a source of olfactory nuisances likely to generate significant annoyance in the neighboring areas. Hydrogen sulfide (H<sub>2</sub>S) is the most common malodorous gas emitted due to both its high emission rate and its very low detection threshold [1, 2]. Its 'rotten egg' odor can be detected at a concentration of 0.4 ppb [3]. It can also cause corrosion in pipelines and thus reduce the lifetime of the plant [3,4]. Furthermore, hydrogen sulfide is a highly toxic gas which poses a serious health risk [5]. The maximum allowable exposure concentration for extended periods is 10 ppm [5]. It is reported that a long exposure to a concentration of 300 ppm of H<sub>2</sub>S in air can cause death. Concentrations exceeding 2000 ppm can be fatal if human beings are exposed to them for a few minutes [3]. To overcome this problem, the Tunisian legislation has issued regulations that deal with the emission of H<sub>2</sub>S into the atmosphere. A number of classical processes have been characterized and implemented to treat these odorous gases prior to their release in the environment. The main processes used to this end are amine absorption [6], alkaline absorption [7], dry oxidation adsorption [8], absorption using the Claus process or catalysts to recover the elemental sulfur [9, 10] and caustic absorption with chemical oxidation [5,11,12]. Even though biofilters can be an interesting alternative [12,1], absorption with chemical reaction is often the most economical method for controlling malodorous process emissions where large flow rates are involved [13].

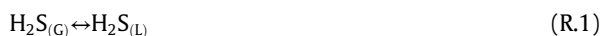
Compared to physical absorption, chemical absorption offers several advantages, mainly the irreversible degradation of the sulfur compounds which maintains a strong driving force for mass transfer [14]. The choice of the solvent and the design of the scrubber system are equally important for the achievement of high scrubbing efficiencies [4]. Sodium hydroxide solution (NaOH) is a very effective but non-regenerable absorbent for H<sub>2</sub>S and CO<sub>2</sub>. Therefore its use is usually limited to the removal of trace amounts of these impurities [4]. Waste gases containing significantly high concentrations of malodorous substances (up to 100 ppm) can be suitably washed by scrubbing with solutions of powerful oxidizing agents. Various oxidants such as chlorine gas (Cl<sub>2</sub>) [15], sodium hypochlorite (NaOCl) [7,11,16], hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) [17] and ozone (O<sub>3</sub>) [18,19,20] are able to remove H<sub>2</sub>S after its transfer to the liquid phase. Earlier work [13] showed that the preferred oxidant for a number of applications appears to be sodium hypochlorite. The most frequently

used is a combination of sodium hydroxide (NaOH) and sodium hypochlorite (NaOCl) because of low cost and good efficiency [5, 13,15].

In this work, the absorption of H<sub>2</sub>S using a chlorinated alkaline solution was studied. The design aspects of the chemical absorption equipment are well understood. Accordingly, we focused on the suitable chemical composition of the washing solution. Given that seawater is already available and is used by the processes belonging to the fertilizer complex of the Tunisian Chemical Group, it is used as a solvent for the chemical absorption of H<sub>2</sub>S. The concentration of sodium hypochlorite and the pH of the absorption media are the most important parameters that control the efficiency of the operation. A laboratory contactor was used to study the effect of these parameters on H<sub>2</sub>S absorption efficiency. In order to find the best operating conditions for the reactive absorption of H<sub>2</sub>S, different values were considered for the main operating parameters such as the gas flow rate, residence time, operating time and volumetric ratio.

## 2. Theoretical aspects

The absorption/oxidation of H<sub>2</sub>S with alkaline chlorinated seawater solution can be described by the following set of reactions. At the gas–liquid interface, the system is presented by reaction (R.1).



According to this reaction, absorption is limited by pollutant solubility in the liquid phase [17]. The solubility of hydrogen sulfide, resulting from the ionization in seawater, depends on temperature and pH in particular [21]. H<sub>2</sub>S is poorly soluble in water. Indeed, Henry's constant for H<sub>2</sub>S is equal to 899 Pa m<sup>3</sup>/mol (at 293.15 K) [22]. Thus, to increase its solubility and achieve good absorption efficiency, it is necessary to use a scrubbing solution of high pH [15, 4]. For this reason and at the effluent temperature (35–45 °C), the absorption of H<sub>2</sub>S is conducted under high alkaline conditions (pH ≥ 9).

After absorption (R.1), H<sub>2</sub>S is dissociated in the seawater solution according to reactions (R.2) and (R.3) and then oxidized by ClO<sup>-</sup> (R.4) into sulfur and/or sulfate compounds ((R.5) and (R.6)). Acido-basic ((R.2) and (R.3)) and oxidizing reactions ((R.5) and (R.6)) occurring in the liquid phase favor mass transfer by shifting the equilibrium of reaction (R.1) towards the formation of non-volatile species [17]. Therefore, reaction (R.2) increases directly the mass transfer by promoting the dissociation of H<sub>2</sub>S into HS<sup>-</sup> which leads to an increase in the pollutant apparent solubility and an acceleration in the inter-phase mass transfer.

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