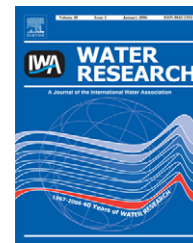


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New approach to optimize operational conditions for the biological treatment of a high-strength thiocyanate and ammonium waste: pH as key factor

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ABSTRACT

Biological treatment of coke and steel-processing wastewaters has to satisfy both industrial economic needs and environmental protection regulations. Nevertheless, as some of the pollutants contained in these waters or produced during the treatment are highly toxic, an effective and safe treatment has proved to be difficult to obtain.

This paper reports the study of a biological method for the treatment of wastewaters containing free cyanide, thiocyanate and ammonium (NH_4). Laboratory-scale activated-sludge reactors were fed with a synthetic solution reproducing a steel-processing industrial wastewater and inoculated with the same industrial bacterial seeding used on-site (Ecosynergie Inc.).

The results demonstrated that free cyanide and thiocyanate were efficiently degraded. Nevertheless, thiocyanate degradation and nitrification processes were actually inhibited by the free ammonia form (NH_3) in place of the ionized NH_4 form (NH_4^+) currently dosed and often improperly named “ammonia” [IUPAC, 1997. In: McNaught, A.D., Wilkinson, A. (compilers). Compendium of Chemical Terminology. Royal Society of Chemistry, Cambridge, UK]. Optimum degradation rates were obtained for very narrow ranges of ammonia nitrogen ($\text{NH}_3\text{-N}$) concentrations. This result can be explained by the role of pH, which mainly controls the NH_3/NH_4 equilibrium.

Pollutants and NH_3 concentrations influenced degradation rates of main pollutants. This influence was determined and expressed through elementary equations.

Although the Michaelis–Menten equation could have been used to describe thiocyanate degradation, a Haldane-inhibition model was used to satisfactorily describe cyanide degradation. On the other hand, a slightly modified Haldane model was applied to describe both NH_4 oxidation using $\text{NH}_3\text{-N}$ as substrate and thiocyanate degradation using $\text{NH}_3\text{-N}$ as inhibitor.

These findings emphasize the role of pH on degradation rates and allow one to optimize operational conditions in the biological treatment of coke and steel industrial wastewaters.

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1. Introduction

Industrial wastewaters from mining, coke or steel industries often contain high concentrations of toxic pollutants such as cyanide, thiocyanate and ammonia (NH_3). Several reviews summarized the treatment feasibility of these pollutants by biological methods (Akcil, 2003; Akcil and Mudder, 2003). Biological treatment appears less expensive than chemical and physical methods, and also much faster than natural oxidation, with a lower environmental impact.

Free cyanide and thiocyanate are biologically degraded to a combination of NH_3 , carbonate and sulfate. So, the resulting effluents may contain high concentrations of NH_3 , which is noxious for the environment and must be removed before effluent discharge. NH_3 is then commonly converted into nitrate through the conventional two-step nitrification process with nitrite (NO_2) as an intermediate product.

Various bacterial strains are able to perform cyanide, thiocyanate and NH_3 oxidation. However, several authors reported difficulties obtaining complete oxidation of NH_3 in this kind of wastewaters. It seemed that nitrifying bacteria were inhibited by high cyanide concentrations and too short acclimation periods. Nitrification stage and nitrifying bacteria recovery were definitely slower than cyanide degradation recovery (Dictor et al., 1997; White et al., 2000; Oliveira et al., 2001; Akcil and Mudder, 2003).

Significant information has been published with regard to the effects of process parameters on the efficiency of cyanide and thiocyanate biological treatment. Some of the parameters controlled were cyanide concentration, bacterial population, acclimation period, hydraulic retention time (HRT) and dissolved oxygen (DO) (Neufeld et al., 1986; Bodzek et al., 1996; Granato et al., 1996; Akcil et al., 2003).

Thiocyanate oxydative degradation leads to pH decrease due to sulfate formation. For an optimum thiocyanate biological treatment, the pH in the reactors is currently adjusted between 6.5 and 7.5 by NaOH or Na_2CO_3 addition (Dictor et al., 1997; Akcil, 2003). On the other hand, it seems that pH is an important factor for the nitrification process and that it must be strictly controlled in order to obtain a complete oxidation of NH_3 (Bernet et al., 2005). In the considered industrial wastewater treatments, pH suffers important variations due to sulfide and NH_3 oxidation. Hence, pH is strongly suspected to be a key parameter for treatment control through NH_3/NH_4 equilibrium control and NH_3 -N toxicity. However, many recent studies about nitrification kinetics did not pay any attention to this issue and considered pH as a constant parameter (Nowak et al., 1995; Dictor et al., 1997; Oliveira et al., 2001; Philips and Verstraete, 2001; Li et al., 2003).

This paper reports a laboratory study of the biological treatment of free cyanide, thiocyanate and ammonium (NH_4) in industrial wastewater. A synthetic wastewater reproducing the composition of a steel industry wastewater has been treated by means of classical activated-sludge pilot plants. Preliminary experiments were performed to determine substrate inhibition kinetics for free cyanide biological treatment. Further experiments allowed us to describe the effect of pH on the efficiency of thiocyanate and NH_4 biological treatment.

2. Materials and methods

2.1. Experimental setup

A first experiment on biological treatment of cyanide (experiment no. 1) was carried out in seven reactors, each of them consisting of a highly mixed aerated tank connected to a clarifier. Each reactor was equipped with two peristaltic pumps, one for feeding synthetic wastewater and one for recycling sludge from the clarifier. Reactor no. 1 had a 3-L aeration tank volume and reactor nos. 2–7 had a 2-L volume. Clarification systems were stirred and designed to ensure recycled sludge retention times smaller than 1/2 h.

The mixing of aeration tanks content was achieved by mechanical stirring. With a stirring power higher than 3WL^{-1} , reactor hydraulics was considered to be close to perfect mixing, and effluent concentrations were considered to represent the concentration of aeration tanks. DO in the reactor no. 1 was regulated at 3mgL^{-1} and the others reactors were continuously fed with air to maintain DO between 2 and 4mgL^{-1} . Temperature was maintained at $29.5 \pm 0.5^\circ\text{C}$ for all reactors (representative of full-scale conditions) and pH was monitored and manually adjusted in reactors by NaOH addition in the feed solution. Water residence times were adjusted using measured outlet flows.

The experiment about biological treatment of thiocyanate and NH_4 (experiment no. 2) was carried out in two reactors, similar to the two type of reactors previously described.

2.2. Synthetic solution

The synthetic solution was prepared three times weekly by dissolving reagent-grade quality dry sodium thiocyanate, dry sodium cyanide, dry sodium sulfate, dry ammonium sulfate and dry potassium hydrogen phosphate in tap water. The targeted chemical oxygen demand (COD) concentration was obtained by adding domestic fuel oil and polysorbate 20 (20mgL^{-1}) as a surfactant to homogenize the solution.

Pollutant concentrations were increased during the experiment up to concentrations similar to the ones found in

Table 1 – Characteristics of the influent fed to two experiments (concentrations in mgL^{-1})

Experiment	COD	Thiocyanate	Phenols	NH_4 as nitrogen	Sulfate	Cyanide	PO_4 as phosphorus
No. 1	1000	0	70	300	1600	5–120	6
No. 2	1000	5–500	70	50–400	1600	0	6

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