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Degradation of recalcitrant compounds from stabilized landfill leachate using a combination of ozone-GAC adsorption treatment

Tonni Agustiono Kurniawan, Wai-Hung Lo *, Gilbert Y.S. Chan *

Department of Applied Biology and Chemical Technology and State Key Laboratory of Chinese Medicine and Molecular Pharmacology, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong Special Administrative Region (HKSAR), China

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Abstract

Laboratory experiments were undertaken to investigate the treatment performances of ozonation alone and/or its combination with granular activated carbon (GAC) adsorption for raw leachate from the NENT landfill (in Hong Kong). To improve its removal of recalcitrant contaminants from the leachate, the surface of GAC was oxidized with ozone prior to treatment. With respect to ozone dose and pH, the removal of COD and/or NH_3-N from ozonation alone and combined ozone-GAC adsorption were evaluated and compared to those of other physico-chemical treatments in some reported studies. The removal mechanism of recalcitrant compounds by ozone-GAC adsorption treatment was presented. Among the various treatments studied, the combination of ozone-GAC adsorption using ozone-modified GAC had the highest removal for COD (86%) and/or NH_3-N (92%) compared to ozonation alone (COD: 35%; NH_3-N: 50%) at the same initial COD and/or NH_3-N concentrations of 8000 and 2620 mg/L, respectively. Although the integrated treatment was more effective than ozonation alone for treating stabilized leachate, the results suggested that it could not generate treated effluent that complied with the COD limit of lower than 200 mg/L and the NH_3-N discharge standard of less than 5 mg/L. Therefore, further biological treatments to complement the degradation of the leachate are still required to meet the environmental legislation.

Keywords: Advanced oxidation process (AOP); Landfill leachate management; Municipal solid waste; Non-biodegradable compounds; Physico-chemical treatments

1. Introduction

Landfill is one of the most widely employed methods for the disposal of municipal solid waste (MSW) around the world. Up to 95% of the total MSW collected worldwide is disposed of in landfills [1]. More than 150,000 landfills have been built worldwide with over 55,000 and 35,000 sites located in the USA and Germany, respectively [2,3]. In the USA, about 57% of the 218 million tonnes of the MSW generated in 2000 was disposed of in landfills [4], while in China, over 80% of the 160 million tonnes of the municipal refuse generated in the same year was buried in 668 landfills [5].

After being landfilled, the refuse decomposes through a series of combined physico-chemical and biological processes, which may take a period of more than 50 years. During the decomposition process, a highly contaminated wastewater called "leachate" is generated by excess rainwater percolating through the waste layers in the landfill [6]. The common features of raw leachate from a local landfill are its high concentrations of ammoniacal nitrogen (NH_3-N) (2000–5000 mg/L) and moderately high strength of recalcitrant compounds (as reflected by its COD value) (5000–20,000 mg/L), as well as a low ratio of BOD_5/COD of less than 0.1 [7,8].

Of the toxic pollutants such as adsorptive organic halogen (AOX), heavy metals and xenobiotic compounds that are present in landfill leachate, NH_3-N, resulting from the decomposition process of organic nitrogen, has been identified not only as a major long-term pollutant [9], but also as the primary cause of acute toxicity [10]. Because NH_3-N is stable under anaerobic conditions, it typically accumulates in the leachate [9]. With a concentration of higher than 100 mg/L [11], untreated NH_3-N is highly toxic to aquatic organisms, as confirmed by toxicity tests using zebrafish (*Danio rerio*) [12], *Daphnia magna* [13] and luminescent bacteria [14].

Unless properly treated, leachate that seeps from a landfill can infiltrate and contaminate the underlying groundwater. Once the

* Corresponding authors. Tel.: +852 2766 5643; fax: +852 2364 9932.
E-mail addresses: bctlo@polyu.edu.hk (W.-H. Lo), bcyschan@polyu.edu.hk (G.Y.S. Chan).

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leachate escapes to the groundwater, it is difficult and expensive to have it controlled and cleaned up, thus posing potentially serious hazards not only to living organisms, but also to public health in the long term. In most cases, it is extremely difficult to restore the polluted groundwater to its former state. In recent years, the risk of groundwater pollution due to leachate seepage has become a major environmental concern worldwide.

A sound engineering design of a landfill can prevent or minimize the leakage of leachate from reaching the groundwater table. Measures include diverting the surface run-off from the site, proper soil cover, proper vegetation and a proper means for leachate interception and collection systems such as synthetic/natural liners, piping and pumping the leachate to a treatment facility [15]. However, most of these precautions can only be applied in newly designed landfills. Some old landfills still generate huge quantities of leachate, which must be treated prior to discharge.

To achieve a satisfactory removal of refractory pollutants from the leachate, several types of physico-chemical treatments such as ammonium stripping, chemical precipitation, coagulation–flocculation, ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) have been employed worldwide. In Hong Kong, leachate from local landfills has been commonly treated on-site using biologically aerobic reactors [16]. Despite reducing the volumetric loading of the reactors, this practice requires a large installation space and high operational cost of US\$ 12.5 m^{-3} of the treated effluent.

As one of the most powerful oxidants with an oxidation potential (E^0) of 2.07 V (Eq. (1)), ozone can act as a very strong oxidizing agent for the treatment of contaminated wastewater of high strength [17]:

$$O_3 + 2H^+ + 2e^- \rightarrow O_2 + H_2O, \quad E^0 = 2.07\,V \tag{1}$$

Due to its capability of transforming contaminants into innocuous substances within a short period, ozone has been identified as a potentially effective means to treat landfill leachate. Unlike chlorination, ozonation does not produce any secondary pollutant in the environment, since the ozonation of organic matter in the leachate leads to the formation of low molecular weight compounds such as acetic acid [18].

Because of its large surface area, microporous structure and surface reactivity, GAC adsorption is one of the most attractive methods for the removal of recalcitrant compounds from leachate. Basically, adsorption is a process by which a substance is transferred from the liquid phase to the surface of a solid and becomes bound by physical and/or chemical interactions [19]. Adsorption treatment using GAC may be technically applicable to meet the increasingly stringent discharge standards for refractory contaminants in developing countries.

In recent years, a combination of ozonation and GAC adsorption has emerged as one of the most promising options for the treatment of contaminated wastewater. Ozone is capable of oxidizing organic substances to their highest stable oxidation states and then, produces CO_2 and H_2O, while GAC can accelerate the kinetic rate of the ozone decomposition through the formation of $^{\bullet}OH$ radicals in the solution [20]. With a high oxidation potential of 2.80 V, $^{\bullet}OH$ radical is a highly reactive species (Eq. (2)):

$$^{\bullet}OH + H^+ + e^- \rightarrow H_2O, \quad E^0 = 2.80\,V \tag{2}$$

Through the $^{\bullet}OH$ radicals, the ozone rapidly reacts with most of the target recalcitrant compounds in the leachate for an ultimate degradation. An integrated treatment of ozonation and GAC adsorption may synergize the advantages of their treatment performance, while overcoming their respective limitations.

In this study, laboratory experiments were carried out to investigate the treatment performances of ozonation alone and/or its combination with GAC adsorption for raw leachate from the NENT landfill (in Hong Kong). To improve its removal of recalcitrant contaminants from the leachate, the surface of GAC was oxidized with ozone prior to treatment. With respect to the ozone dose and pH, the removal of COD and/or NH_3-N from ozonation alone and/or combined ozone-GAC adsorption were evaluated and compared to those of other physico-chemical treatments in some reported studies. The removal mechanism of recalcitrant compounds by combined ozone-GAC adsorption treatment is also presented.

2. Experiments

2.1. Materials and methods

An aged raw leachate was collected from the North East New Territory (NENT) landfill situated in Ta Kwung Lin, a remote part of the New Territories. It is one of the three on-going landfills in Hong Kong SAR, in addition to the South East New Territories (SENT) and the West New Territories (WENT) landfills. With an overall capacity of $37 \times 10^6\,m^3$ and a total area of 61 ha, the NENT landfill receives 3800 tonnes of MSW per day [21] from an urban population of nearly 7.5 million of inhabitants in 2005. Since its first commissioning operation in June 1995, the major component of waste dumped in the landfill is municipal refuse such as domestic, construction and industrial waste. The NENT landfill generates approximately 800 m^3 of leachate per day [22].

To study the effects of seasonal variations on the composition and the concentration of the raw leachate from the NENT landfill, the leachate samples were collected in December 2004 (winter) and July 2005 (rainy and summer). In Hong Kong, spring and summer are generally considered as rainy season, while fall and winter are dry seasons. Due to the impacts of rainfall on the change of physico-chemical characteristics of the leachate during the rainy season, the samples collected in December 2004 were used throughout this study.

The samples were collected in 20 L of polyethylene carboys that were filled to the capacity and capped tightly. They were then stored in a refrigerated storage chamber at 4 °C to minimize any further changes that might occur in its physico-chemical and biological properties prior to experiments. The leachate was immediately characterized according to the Standard Methods [23] for the following parameters: pH, COD, BOD_5, NH_3-N, alkalinity (as $CaCO_3$), total nitrogen, NO_3-N, total