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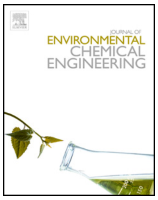

Cost estimation of COD and color removal from landfill leachate using combined coffee-waste based activated carbon with advanced oxidation processes

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ABSTRACT

Advanced oxidation processes (AOPs) effectively treat landfill leachate, but high energy demands and capital costs mean that further research is needed to improve AOP viability. In the present study, two AOPs (ultrasound + O_3 and solar/O_3; with and without H_2O_2) were compared in terms of landfill leachate depuration, specific energy consumption, and associated reagent and operational costs. Solar/O_3/H_2O_2 attained 59.7% color elimination and 34.4% chemical oxygen demand (COD) abatement, while ultrasound + O_3/H_2O_2 achieved 41.5% color elimination and 33.8% COD abatement. A novel adsorption pretreatment based on activated carbon (1 g/L) obtained from coffee waste was also investigated, resulting in 68.5% COD abatment and 56.8% color elimination. When adsorption pretreatment was followed by the solar/O_3 AOP, maximum color (58.3%) and COD (70.8%) removals were obtained. The least energy-demanding treatment was solar/O_3/H_2O_2, which required 0.011 Wh/mg of COD removed. Higher operating costs were associated with adsorption pretreatment, followed by ultrasound and H_2O_2 use. Finally, using solar energy resulted in substantial reduced energy costs. In conclusion, the results of this study support the industrial-scale application of ozonation combined with H_2O_2 and solar energy as a technically and economically feasible method for treating and reusing landfill leachate.

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1. Introduction

Landfills are the main repository for disposing of a wide array of consumer products [1]. Consequently, landfill leachates (LL) are significantly contaminated with a mixture of high-strength organic/inorganic, dissolved/suspended contaminants, among which, refractory humic substances, mainly humic acids and fulvic acids, are the primary components [2]. Despite strict rejection standards, conventional LL treatment plants are often unequipped to meet legislated norms [3]. Conventional LL treatment technologies are biological, chemical, or physical in nature, and there is a growing need for treatment methods that incorporate these three technologies [4]. Regarding this, recent physicochemical processes have been implemented for LL management and treatment.

Of particular note among physiochemical processes is the adsorption of activated carbon (AC). In addition to applications in LL treatment, AC adsorption has been tested as a post-treatment to the Fenton process and as a pretreatment to coagulation-flocculation. In fact, adsorption processes using AC are highly versatile in removing a wide range of organic and inorganic pollutants [5]. Specifically, adsorption has been extensively used to remove pollutants from water. In research by Mushtaq et al. [6], a biocomposite was prepared using *Eriobotrya japonica* seeds and Na-bentonite for the efficient adsorption of Cu(II) ions from a solution. Rashid et al. [7] also achieved high rates of nickel and zinc adsorption by using a composite of dead fungal biomass and bentonite. Furthermore, a comparative study by Babarinde et al. [8] on metal adsorption using lemon grass revealed this bioadsorbent

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Abbreviations: AOPs, advanced oxidation processes; COD, chemical oxygen demand; LL, landfill leachates; SEC, specific energy consumption; US, ultrasound.

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effective in removing Pb(III), Cd(II), and Zn(II) from aqueous solutions.

In recent years, advanced oxidation processes (AOPs) have been efficiently used to improve LL biodegradability. AOPs generate short-lived $^\bullet OH$, a non-selective, strong oxidant that degrades organic compounds. Indeed, AOPs can induce a transformation of dissolved organic carbon into carbon dioxide and mineral acids. However, ongoing research is needed to reduce the high energy demand and capital costs of most AOPs [9]. Regarding this, UV/H_2O_2, O_3, and O_3/H_2O_2 are the most frequently applied processes for generating $^\bullet OH$ in the secondary or tertiary treatment of LL [10], and favorable results can also be achieved by using solar photo-Fenton oxidation [11]. Indeed, De Torres-Socías et al. [12] obtained a high degree of LL depuration using a pretreatment process coupled to a solar photo-Fenton treatment, resulting in improved wastewater biodegradability.

In LL treatment, AOPs can improve the biodegradability of recalcitrant organic pollutants at costs comparable to conventional biological treatments [3]. For example, Cortez et al. [10] investigated LL treatment efficacy for Fenton treatment and other AOPs (O_3, O_3/OH^- and O_3/H_2O_2) at a pilot plant scale. Furthermore, high pH conditions ($pH > 8$), generate hydroxyl radicals ($^\bullet OH$) that accelerate organic matter oxidation from the complex wastewater matrix [13]. Under these conditions, target compounds are either directly oxidized by O_3 or indirectly by $^\bullet OH$. Notably, $^\bullet OH$ can react with the majority of solutes in water [14]. The ozonation process can also be improved by UV irradiation, which generates more $^\bullet OH$. Eqs. (1) and (2) represent the chemical reactions involved in ozone (O_3) actions in water under UV irradiation [15], including ozone photodecomposition in the presence of UV, a process that generates H_2O_2 and hydroxyl radicals.

$$O_3 + H_2O + h\nu \rightarrow H_2O_2 + O_2,\ h\nu < 310\,\text{nm} \tag{1}$$

$$O_3 + H_2O_2 \rightarrow HO_2^\bullet + OH^\bullet + O_2 \tag{2}$$

Then, ozone and H_2O_2 react with hydroxyl radicals to form hydroperoxyl (Eq. (3)) and superoxide radicals (Eq. (4)).

$$^\bullet OH + O_3 \rightarrow O_2 + HO_2^\bullet \tag{3}$$

$$^\bullet OH + H_2O_2 \rightarrow O_2^\bullet - + H_2O + H^+ \tag{4}$$

The $^\bullet OH$ yielded can oxidize organic pollutants and generate oxidation products (Eq. (5)).

$$^\bullet OH + \text{Organic} \rightarrow \text{Products} \tag{5}$$

Finally, these radicals further react with hydroxyl radicals and ozone to produce more hydroxyl radicals (Eqs. (6)–(8)).

$$^\bullet OH + O_2^{\bullet -} \rightarrow OH^- + O_2 \tag{6}$$

$$O_3 + O_2^{\bullet -} \rightarrow O_3^{\bullet -} + O_2 \tag{7}$$

$$O_3^{\bullet -} + H_2O \rightarrow {}^\bullet OH + OH^- + O_2 \tag{8}$$

O_3 can also react with dissolved organic matter (DOM) present in LL to generate $^\bullet OH$ and an oxidation product, as shown in Eq. (9):

$$O_3 + \text{DOM} \rightarrow {}^\bullet OH + \text{products} \tag{9}$$

In basic conditions, OH^- acts in ozone decomposition by producing highly reactive intermediates, such as the superoxide ion and $HO_2^\bullet$ (Eq. (10)).

$$O_3 + OH^- \rightarrow O_2^{\bullet -} + HO_2^\bullet \tag{10}$$

Furthermore, the generated $HO_2^\bullet$ can produce hydrogen peroxide, oxygen, and hydroxyl radicals (Eqs. (11)–(12))

$$HO_2^\bullet + HO_2^\bullet \rightarrow H_2O_2 + O_2 \tag{11}$$

$$HO_2^\bullet + H_2O_2 \rightarrow {}^\bullet OH + H_2O + O_2 \tag{12}$$

Although O_3 can completely oxidize most organic contents in LL, its utility as a complete treatment method is limited due to high ozone doses and long treatment times [16], translating into increased energy consumption.

Among AOPs, ultrasound (US) technology is a viable process that does not require chemicals. Indeed, US technology has the potential advantages of being simpler, safer, and cleaner than conventional AOPs. This method functions by exposing liquids to high mechanical pressure waves, resulting in acoustic cavitations that induce US propagations. US disintegration arises from the formation, growth, and implosive collapse of bubbles in the corresponding liquid [17]. Ultrasonic liquid irradiation causes high-energy chemical reactions inside the bubble and in liquid surrounding the bubble, including the cleavage of H_2O molecules into $^\bullet OH$ and $H^\bullet$ radicals [18]. The reaction of these radicals with water could form H_2O_2 and other radicals, such as $HO_2^\bullet$, which can degrade stable organic contaminants [19] (Eqs. (13)–(16)).

$$H_2O + US \rightarrow H^\bullet + {}^\bullet OH \tag{13}$$

$$H^\bullet + O_2 + US \rightarrow HOO^\bullet \tag{14}$$

$$\text{Pollutants} + {}^\bullet OH + US \rightarrow \text{Degradation products} \tag{15}$$

$$\text{Pollutants} + HOO^\bullet + US \rightarrow \text{Degradation products} \tag{16}$$

Furthermore, the US process can promote not only pollutant degradation through hydroxyl radicals, but also degradation through pyrolysis, in accordance with molecular properties [20]. The inefficient conversion of energy in the US process and possible difficulties in scaling are the main problems of this technology [21]. Considering these operating costs, solar energy is increasingly in high demand for the treatment of complex wastewaters. Efforts to improve the treatment of wastewater using solar energy include maximizing the incoming solar energy dose, thereby enhancing efficacy in toxic compound elimination.

The aim of this study was to evaluate the degree of LL depuration achieved by using new adsorption processes. Specifically, two different AOPs were assessed: 1) US combined with ozonation ($US + O_3$), and 2) solar/ozonation (Solar/O_3). Both processes were evaluated without or without AC (obtained from coffee waste) pretreatment and with or without H_2O_2; meaning that eight processes total were analyzed. Comparisons were performed for both AOPs in terms of energy consumption, measured as specific energy consumption (SEC); the reagents needed to depurate a unitary mass of organic contamination, measured as chemical oxygen demand (COD); and overall cost-efficiency.

2. Materials & methods

2.1. Experimental design

The LL samples used in this study were obtained from different sectors of a Chilean landfill (16.7 ha total area) managing domestic waste. The main characteristics of raw LL samples are shown in

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