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Advanced Oxidation Treatment of Composting Leachate of Food Solid Waste by Ozone-Hydrogen Peroxide

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ABSTRACT
The research was conducted to investigate the efficiency and possibilities of advanced oxidation process based on ozone-hydrogen peroxide. The process was used as a post-treatment step of composting leachate utilisation. The leachate samples were collected from a typical composting plant with the aerobic biological treatment system. The samples were conditioned in a "ozone reactor" without dilution. The effectiveness of the treatment process was measured by pH values (4.0–7.0), H_2O_2 concentrations (0.5–4.0 g·dm^{-3}), ozone doses (0.5–1.5 g·h^{-1}) and reaction times (0–10 min). The highest removal efficiencies achieved were 85% and 92% for chemical oxygen demand (COD) and biochemical oxygen demand (BOD_5), respectively. The optimum process parameters occurred at pH 5.0, 2.0 g·dm^{-3} concentration of H_2O_2, and 0.75 g·h^{-1} of ozone dose. The optimal reaction time was 3 min. The O_3/H_2O_2 advanced oxidation process was found to oxidize COD and BOD_5 of the composting leachate. The oxidation reaction can be used as a feasible technique for composting leachate treatment.

Keywords: BOD_5, COD, oxidation, compost, leachate

INTRODUCTION

Food wastes (FW) are the main component of municipal solid waste (MSW). The estimates provided by Food and Agricultural Organization (2011), Carlson et al. (2012) and Zaman (2013) indicate increasing global food consumption and an over 40% increase in FW production by 2025. While analyzing the issues of the MSW economy, attention should be paid to the increase in the recycling of biodegradable waste. This process is related to the progressive reduction of storing in landfills waste that can be re-used (Wiliams, 2005).

One of the most popular FW recycling methods is composting. It belongs to the key technologies commonly used worldwide (Pitchel, 2010). Under temperate climate conditions, the composting process is divided into two stages, which in total should last for at least 8 weeks. The long duration time of the process depends on the choice of the composting method (active system with oxidation of the material or passive without oxidation) and the processed material type. On the basis of the research conducted in various countries, it was found that the first phase of the composting process should last approximately 2 weeks. Then, the material is directed to stabilization, which lasts 6–8 weeks. Currently, many studies in the field of optimization of the kinetics of the composting process in its first phase and the elimination of accompanying odorous substances are conducted globally (Lebrero et al., 2011; Yuan et al., 2015; Fernández et al., 2016; Jinyi et al., 2016; Siles et al., 2016; Yongjiang et al., 2016). In addition, in the literature on the subject there are studies on the additional hygienization of the stabilized material and the possibility of using gases generated during the process. One of the main problems associated with composting is the generation of the leachate containing material that has been extracted, suspended or dissolved in waste (Benlboukht et al., 2016; Junya et al., 2016; Lian et al., 2016; Mukesh et al., 2016).

advanced oxidation processes (AOP)are very popular in current technologies of neutralization of fluid process residues. Most commonly, they are used to eliminate color, reduce the load of organic pollutants or reduce the toxicity of water and wastewater (Jia et al., 2011; Cortez et al., 2011). The principle of AOP is based on the release of hydroxyl radicals, which accelerate the degradation of organic compounds in the aquatic environment.

The most popular AOP is the O_3/H_2O_2 system, which is used as a chemical process for purification and pre-treatment of wastewater (Maniero et al. 2008; Rosal et al., 2009; Liu et al., 2009; Quiang et al.; 2010). Tizaoui et al. (2007) obtained during the ozonization of the leachate the COD removal rate of 27% and the color reduction by 85%. Rivas et al. (2003) and Hagman et al. (2008) received at least a 20% reduction in COD using only hydrogen peroxide (H_2O_2). Using ozone along with the H_2O_2, a COD reduction of 20–50% can be achieved (Tizaoui et al., 2007; Hagman et al., 2008; Goi et al., 2009). The presented results refer to the application of the AOP system for wastewater. The leachates from the composting process are a more specific waste, the properties of which depend mainly on the oxygenation and moisture of the processed material.

The purpose of this article is to present the possibility of using the O_3/H_2O_2 oxidation system in the process of preliminary preparation of the leachate from the FW composting process for neutralization in a professional wastewater treatment plant. The effectiveness of the process was assessed by the BOD_5, COD dynamics of change and BOD_5/COD ratio.

MATERIAL AND METHODS

Leachate samples

The leachate samples were prepared during the FW composting process in the BKB 100 isothermal bioreactor for the study of organic material decomposition processes. A FW mixture consisting of vegetables (50%), fruit (25%), expired food (15%) and sawdust (10%) with a total fresh weight of 50 kg was used for the tests. The composition of the mixture was selected to obtain appropriate structural properties of compost, while ensuring proper air flow in the bioreactor, biomass moisture during the process (54–68%) and a C:N ratio above 20. In order to achieve a proper composting process and a distinct thermophilic phase, the air flow in the bioreactor was set at 0.035 $m^3 \cdot h^{-1} \cdot kg_{dw}^{-1}$ (which was 0.4 $m^3 \cdot h^{-1}$ in the composted waste mass). During the process, a total of 3.3 dm^3 leachates were created.

Experimental procedure

The advanced leachate oxidation process was carried out in a flow glass reactor. The reactor is equipped with a stirrer, by means of which it is possible to aerate the mixture with the ozone coming from the discharge generator. Through the system of independent infusions, the leachates and the hydrogen peroxide solution are infused into the reaction chamber. The AOP procedure proceeded according to the following scheme:

1. Filling the reactor with 1 dm^3 of the leachate, which was mixed applying air with a capacity of 5.0 $dm^3 \cdot min^{-1}$ over a period of 5 minutes.
2. The AOP process involved:
   a) Addition of H_2O_2 (1 $g \cdot dm^{-3}$) and ozone (1 $g \cdot h^{-1}$), reaction time 10 minutes, regulation of the reaction using 1N sodium hydroxide in the range of 4÷7 (optimization of the reaction).
   b) Differentiated H_2O_2 concentration at the optimal pH in the range of 0.5÷4 $g \cdot dm^{-3}$, ozone dose 1 $g \cdot h^{-1}$, reaction time – 10 minutes.
   c) Modification of ozone concentration in the range of 0.5÷1.5 $g \cdot h^{-1}$, reaction time 10 minutes, optimal pH and H_2O_2 concentration.
3. For each step presented in section 2, pH, COD, BOD_5, BOD_5/COD analyses were performed.

Materials and analysis

Hydrogen peroxide (30%, w/w) and sodium hydroxide solution (1 N) were of analytical reagent grade (Merck, Germany). Ozone was produced in the O3PRO30,7VW generator, equipped with a corona electrode system. The pH was measured by an Elmetron CP-511 conductometer with an EPS-1 glass electrode for the measurement of pH in the aqueous solution. COD was measured using a standard miniaturized method in airtight tubes, in accordance with the PN-ISO 15705:2005 standard. For the BOD_5 measurement, the respirometric method described in DIN EN 1899 H55 was used, which uses OxiTop Control IS (WTW, Germany) for measurements. All experiments were carried out in 5 repetitions. During testing, the temperature was maintained at 22±1°C and humidity at 55±5%.

RESULTS

The examined leachate samples were taken from the process of composting biodegradable waste in container technology. The basic leachate parameters were 36.9 g·dm^{-3} (COD), 19.7 g·dm^{-3} (BOD_5) at pH 7.32 and electrical conductivity 12.73 mS·cm^{-1}. The sample of the leachate was characterized by a black color and a sharp, irritating odor. The analyzed leachate samples have distinctively high level of oxygen demand, whereas the BOD_5/COD ratio was 0.53, which indicates a high potential of biodegradability of organic matter.

The values of COD and BOD_5 at the pH of 5.0 reached maximum, which amounted to 26% and 31%, respectively (Fig. 1). The efficiency of oxygen demand reduction fell above the pH value of 9.5 and remained stable. The Pearson correlation test showed a direct linear relationship between the pH values and COD removal efficiency ($P<0.05$, $r=0.817$) and BOD_5 ($P<0.05$, $r=0.836$).

The effect of various concentrations of H_2O_2 was examined in eight variants of concentration 0.5÷4 g·dm^{-3} with the ozonization efficiency of 1 g·h^{-1} (Fig. 2). The maximum degradation of COD and BOD_5 was 42% and 49%, respectively, at a concentration of 3.0 g·dm^{-3} H_2O_2. A further increase in the H_2O_2 concentration caused slower degradation of both COD and BOD_5. The Pearson correlation test showed a direct linear relationship between the H_2O_2 concentration and BOD_5 removal efficiency ($P<0.05$, $r=0.728$). At the same time, this test showed an inverse relationship between H_2O_2 concentration and COD removal, which was not significant ($P<0.05$, $r=-0.408$).

The effect of the ozone dose on the removal of COD and BOD_5 in the presence of an optimized dose of H_2O_2 and pH (5.0) caused a linear increase in the degradation of oxygen demand and constant reaction time (Fig. 3). The highest efficiency of COD removal (36%) was achieved with 0.75 g·dm^{-3} ozone dose. Removal of BOD_5 achieved the highest reduction at an analogous dose of ozone and amounted to 49%. The above-mentioned results were achieved during a 10-minute reaction time.

In order to examine the effect of reaction time on the oxygen demand reduction efficiency, an experiment was carried out at variable reaction times of 0÷10 minutes in the previously optimized doses of H_2O_2 and O_3 (Fig. 4). The results showed that the increase in the reaction time linearly increases the efficiency of oxygen demand reduction. The maximum COD and BOD_5 reduction values were 63% and 69%, respectively, at 4 minutes. In the reaction time range of 6÷10 minutes, the oxygen demand reduction efficiencies did not change significantly. The results of Pearson correlation test showed a direct linear relationship between the reaction time and the reduction efficiency of BOD_5 ($P<0.05$, $r=0.992$) and COD ($P<0.05$, $r=0.998$).

The BOD_5/COD ratio during the ozonization changes in time analogous to the variability of the reduced indicators. For the untreated samples, it reached 0.53. After taking into account the optimal process conditions, the BOD_5/COD ratio amounted to 0.38.

DISCUSSION

In the carried out study, stable results were obtained at lower pH values than described in 20–21. Figure 1 shows that a more stable

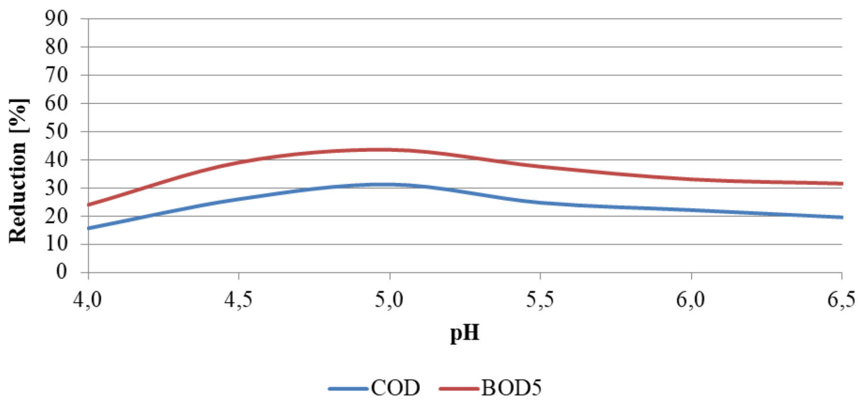

Fig. 1. Effect of the pH on COD and BOD_5 removal (H_2O_2 [1.0 g·dm^{-3}], O_3 [1.0 g·h^{-1}] and reaction time [10 min])

reduction in oxygen demand was achieved by leachate than in Cortez et al. (2011). The studies showed that the effect of using H_2O_2 positively influenced the efficiency of removing pollutants while using concentrations in the range of 1.5–2.5 $g \cdot dm^{-3}$. Cortez et al. (2011) noted the positive effect of using the O_3/H_2O_2 system in relation to the ozonation alone. In his studies, he achieved a reduction of COD from 27% to 72% depending on hydrogen peroxide concentration.

Studies by Tizaoui et al. (2007) and Rivas et al. (2003) stated that the application of excessive doses of H_2O_2 adversely affects the process of wastewater treatment. This has also been confirmed in the carried out studies as illustrated in Figure 2. Tizaouni et al. (2007) received optimal process parameters at the hydrogen peroxide concentration 2 $g \cdot dm^{-3}$. With further increase in concentration (up to 6 $g \cdot dm^{-3}$) the process efficiency decreased by 6.5%.

Ozone may participate in the generation of hydroxyl radicals. While increasing the concentration of O_3 in the studied process, the COD and BOD_5 removal rate improved (Fig. 3). Hydroxyl radicals that have not participated in wastewater treatment, together with ozone, increase the degradability of pollutants, confirming studies 24. In the study by Maniero et al. (2008) a 70% decrease in the COD value was obtained after the application of ozone at a dose of 9 $g \cdot dm^{-3}$. In analogous studies by Cortez et al. (2011) COD removal at 25% was achieved with the concentration of O_3 in the range of 63–112 $g \cdot dm^{-3}$.

Reaction time is a key indicator of the wastewater treatment process. The conducted research has shown that the use of AOP effectively reduces the time needed to remove contaminants. At the same time, the positive, the synergistic effect of using oxidants on wastewater treatment, described by 8–20, was confirmed. The achieved results of reducing the reaction time (Fig. 4), gave better results than those described in papers Lian et al. (2016) and Mukesh et al. (2016).

The presented research results show that the proper use of the O_3/H_2O_2 oxidation system increases the biodegradability of wastewater. Similar results were obtained by Goi et al. (2009) during the leachate treatment using the Fenton reaction. In addition, during the process of contaminants oxidation, changes in the molecular structure of contaminants were achieved, as shown by the BOD_5/COD indicator. By reducing the oxygen demand, a transformation of compounds that are difficult to biodegrade into more degradable forms has been achieved, which was also described by Jinyi et al. (2016) and Rivas at al. (2003).

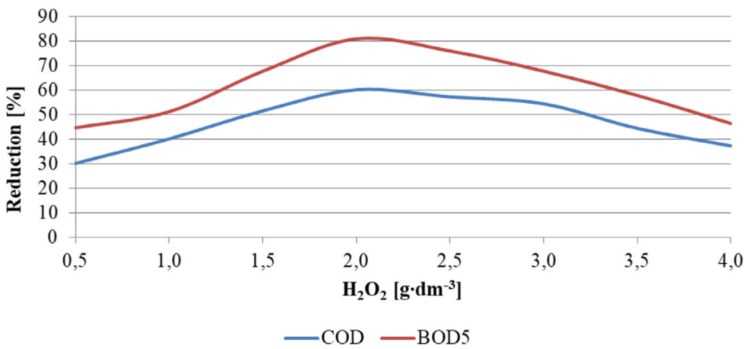

Fig. 2. Effect of the H_2O_2 on COD and BOD_5 removal (pH [5.0], O_3 [1.0 $g \cdot h^{-1}$] and reaction time [10 min])

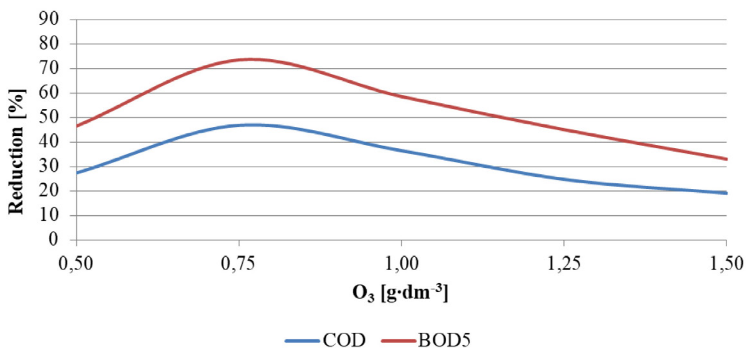

Fig. 3. Effect of the O_3 on COD and BOD_5 removal (pH [5.0], H_2O_2 [3.0 $g \cdot dm^{-3}$] and reaction time [10 min])

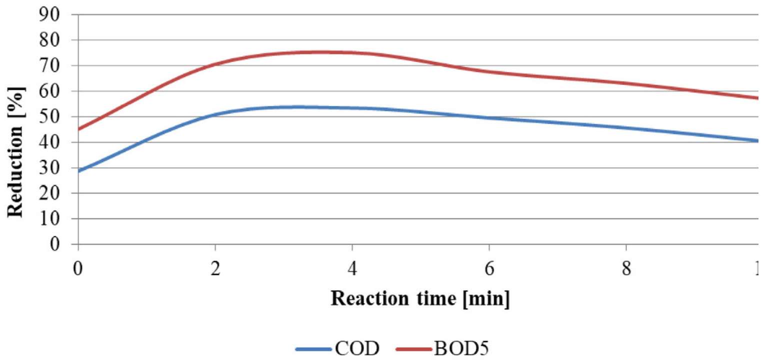

Fig. 4. Effect of the reaction time on COD and BOD_5 removal (pH [5.0], H_2O_2 [1.0 g·dm^{-3}] and O_3 [0.75 g·h^{-1}])

CONCLUSION

The results of the carried out tests clearly show that the combination of H_2O_2 and O_3 in one oxidation system is an effective way to reduce the oxygen demand during leachate treatment. The optimum process conditions occur at the pH of 5.0 and the addition of oxidants at the level of 0.75 g·dm^{-3} each. The optimal and reasonable reaction time is 4 minutes. The test results indicate the beneficial effect of AOP on process leachates, reducing the BOD_5/COD ratio by 30%, thus – lowering the oxygen demand during the treatment. The use of this reaction, due to the high biodegradability of leachates from the composting process, can be a preparatory process before transferring them to a wastewater treatment plant.

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