Process Configuration of Combined Ozonolysis and Anaerobic Digestion for Wastewater Treatment.

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Abstract

Industrial activities and increased human population have resulted in wastewaters not entirely amenable to conventional treatment methods. Anaerobic digestion (AD) can treat such wastewaters with the advantage of bioresource recovery. However, the presence of solids and recalcitrant compounds in most wastewaters may affect the AD process. Thus, combining AD with advanced oxidation processes is necessary. This study combined ozonolysis with AD to treat waste activated sludge (WAS) and distillery wastewater (DWW). When applied as a pretreatment, ozonolysis caused the rigid cell walls in WAS to rupture and solubilised the extracellular polymeric substances (EPS), leading to increased biodegradability. For the DWW, ozonolysis pretreatment reduced the biorecalcitrant aromatic compounds to simple aliphatic compounds, thereby increasing biodegradability. In the ensuing AD process, the WAS pretreatment improved TSS and COD reductions and a 230% increase in cumulative biogas production. For the DWW, the ozonolysis pretreatment did not significantly impact COD reduction or biogas production; however, when applied as a posttreatment, ozonolysis effectively removed the biorecalcitrant colour of the anaerobically digested effluent and solubilised the TSS washed out from the AD unit. Therefore, the AD-ozonolysis process configuration is substrate specific; ozonolysis is best applied as a pre-AD and post-AD for WAS and DWW, respectively.

Keywords: Ozonolysis, Pretreatment, Posttreatment, Solubilisation, Biogas.

1. Introduction

Activated sludge is the flocculent culture of organisms developed in aeration tanks due to wastewater treatment under controlled conditions (Ouyang et al., 2020). Industrial wastewater or sewage is treated such that biological floc is formed from organisms in the water by bubbling atmospheric air, thus reducing the sewage's organic content (Skouteris et al., 2020). Once this water has been treated, the overflow of the mixed liquor is sent into settling tanks, and the treated liquor is released for further treatment before discharge (Singh Asiwal et al., 2016). The excess sludge that accumulates is called waste activated sludge (WAS) which is ultimately removed from the treatment process and stored in storage tanks away from the primary treatment process. The generated WAS is sometimes subjected to further treatment by aerobic or anaerobic digestion before disposal (Ozgun, 2019; Otieno et al., 2019). WAS management and treatment account for up to 60% of the total costs incurred by wastewater treatment plants (Otieno et al., 2019). Anaerobic digestion (AD) of WAS for solids reduction, energy recovery, and stabilisation has long been considered (Silvestre et al., 2015). However, the unique characteristics of WAS restrict the AD process by slowing digestion, resulting in low solids reduction and low biogas production (Liang et al., 2021). It is thus essential to introduce a pretreatment process to enhance the biodegradability of WAS, thereby improving biogas production and solids reduction (Otieno et al., 2019).

Alcohol distilleries are one of the leading environmental polluters, as approximately 88% of the raw material (mainly molasses) used in production ends up in wastewater (Shivajirao, 2012; Mabuza et al., 2017). Distillery wastewater (DWW) is characterised by a dark colour, bad smell, and a high organic load as indicated by a biochemical oxygen demand (BOD) of

45 000-60 000 and chemical oxygen demand (COD) of 70 000-120 000 mg/L (Apollo and Aoyi, 2016; Otieno et al., 2017). Discharging the DWW into receiving streams such as rivers and lakes can lead to eutrophication and hinder photosynthesis by aquatic plants (Otieno et al. 2016; Navgire et al. 2012). The anaerobic digestion method is often the preferred first step in treating DWW. However, the biorecalcitrant colour causing melanoidin compounds hinders the rate determining hydrolysis step. Moreover, during AD, the colour-causing melanoidin compounds repolymerises easily, thereby intensifying the colour of the AD effluent (Mabuza et al., 2017).

Integrating AD with advanced oxidation processes (AOPs) such as ozonolysis is important for effective wastewater treatment. For DWW, ozonolysis pretreatment can break down the colour-causing biorecalcitrant polymeric high molecular weight (HMW) organic compounds into biodegradable low molecular weight (LMW) compounds, thus improving substrate biodegradability (Santos et al., 2013; Amaral-Silva et al., 2016). For WAS, ozonolysis has been an ideal pretreatment method for solubilising the solids, leading to improved biodegradability (Otieno et al., 2019). Additionally, the advantages of integrating ozonolysis as a pretreatment process are that it generates low inhibitory compounds and operates at ambient temperature and pressure. The ozone can be generated on-site and utilised directly, avoiding chemical supply and storage issues (Otieno et al., 2019). Alternatively, ozonolysis can be applied as a posttreatment to eliminate recalcitrant compounds that have escaped the AD process (Otieno et al., 2019). This study investigated the application of an integrated AD-AOP system to treat WAS and DWW. Ozonolysis was applied as a pre- or post-treatment to AD to determine the best system configuration. The integrated system was evaluated based on biodegradability enhancement, colour and COD reductions, and sludge solubilisation to determine the best process configuration.

2. Methodology

2.1. Materials

Sodium thiosulphate, phosphoric acid, methanol, potassium iodide (KI), sulphuric acid, silver sulfate, hydrochloric acid, sodium hydroxide, potassium dichromate, and starch were all obtained from Merck Limited in South Africa. All the sourced chemicals (analytical grade) were used as received.

2.2. Distillery wastewater and waste activated sludge

The waste activated sludge (WAS) used in the current study was obtained from the secondary settling tank of a wastewater treatment (WWT) plant in a local municipality, Vanderbijlpark, South Africa. Distillery wastewater (DWW) was collected from a molasses-based alcohol distillation plant in Durban, South Africa, and stored at 4 °C until used. The characteristics of the WAS and DWW are given in Tables 1 and 2, respectively.

2.3. Ozonolysis pretreatment process for WAS and DWW

The WAS and DWW substrates were pretreated in a 5 L fluidised ozone reactor to improve sludge solubilisation and enhance biodegradability before AD. Briefly, the wastewater substrate was transferred to the ozone reactor and then subjected to ozonolysis at a constant ozone dosage (45 mg/L/min) for one hour. The ozone gas from an ozone generator was bubbled through the substrate contained in the reactor via a gas diffuser placed at the bottom. The schematic representation of the ozonolysis set-up is given in Figure 1a.

2.4. Anaerobic digestion of WAS and DWW

Anaerobic digestion of the two wastewater substrates was carried out in two separate UASB reactors. For the DWW, the AD reactor (Figure 1.1b) of 2L working volume was operated at an optimum organic loading rate (OLR) after a successful digester start-up. The digester was operated stepwise during start-up from an OLR of 1.2 until the optimum OLR of 15 kg

COD/M³/day was attained. For the start-up, the UASB digester was inoculated with active anaerobic sludge granules obtained from a digester treating breweries wastewater. Distillery wastewater was added in small amounts, increasing the amount added whenever reactor stability had been attained, as indicated by near constant COD reduction and biogas production. The start-up period lasted for 33 days. Afterwards, the digester was operated semi-continuously from day 34 to 54 while feeding non-pretreated DWW and from day 55 to 74 with ozone pretreated DWW.

For the anaerobic digestion of WAS, an already active UASB reactor of 3 L working volume, of which 1 L consisted of sludge granules from an anaerobic digester treating municipal wastewater, was used. Anaerobic digestion of WAS was done in semi-batch mode with manual sampling and feeding. Each batch lasted six days (as determined by near-constant COD reduction and diminished biogas production after the 6th day), after which a new feed was introduced. The digestion temperature was maintained at 37 °C using a heating tape wrapped around the reactor. The reactor was fed raw WAS (no pretreatment) in the first three batches, while ozone pretreated WAS was fed in the subsequent three batches. Daily biogas production was monitored. Also, samples were collected and analysed for COD and TSS.



Figure 1: Schematic representation of the (a) ozonation and (b) anaerobic units.

2.5. Ozonolysis posttreatment of anaerobically digested DWW

The effluent from the UASB reactor treating DWW was diluted with water and then subjected to ozonolysis in the ozone reactor to remove the biorecalcitrant colour.

2.6. Physical and chemical analysis

Samples withdrawn at pre-determined time intervals during the AD and ozonolysis processes were analysed for pH, colour, COD, dissolved organic carbon (DOC), and BOD. The COD, DOC, pH, and BOD were determined following standard methods (Mecha et al., 2016). Aromaticity and colour were determined by UV absorption measurement, while the concentration of the cations was determined from Ion Chromatography (Navgire et al., 2012).

3. Results and Discussion

3.1. Characteristics of WAS and DWW before and after pretreatment

Tables 1 and 2 give the physicochemical characteristics of the WAS and DWW, respectively, before and after ozone pretreatment. During WAS pretreatment, the TSS was reduced by 29%

from 20.9 mg/L to 14.9 mg/L, indicating the solubilisation of the suspended solids (Gomes et al., 2013; Ariunbaatar et al., 2014). The solubilisation released the suspended COD into the aqueous phase leading to the observed increase in DOC and soluble COD from 1 700 to 2 300 mg/L and 155 to 245 mg/L, respectively. On the other hand, the total COD slightly decreased from 24 500 to 21 600 mg/L, ensuring adequate substrate retention for the ensuing anaerobic process. Through ozonolysis, the hard cell walls contained in WAS were ruptured, and the extracellular polymeric substances (EPS) solubilised, releasing the cellular contents. The increased concentration of sulphates (18 to 75 mg/L) and nitrates (0 to 115 mg/L) in the supernatant confirmed the release of cellular contents (Otieno et al., 2019). The BOD₅ of the pre-treated WAS increased from 1 520 to 3 520 mg/L contributing to an overall 2.6-fold increase in biodegradability in the BOD₅:COD ratio.

Parameter	Value		
	Before ozonolysis	After ozonolysis	
рН	6.7	6.1	
TSS (mg/L)	20.9	14.9	
COD⊤ (mg/L)	24 500	21 600	
COD _S (mg/L)	1 700	2 300	
DOC (mg/L)	155	245	
BOD₅ (mg/L)	1 520	3520	
BOD5:COD	0.06	0.16	
Sulphate (mg/L)	18	75	
Phosphate (mg/L)	67	35	

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Develop	Value			
Parameter	Before ozonoly	sis After Ozonolysis		
рН	5.55	5.01		
COD⊤ (mg/L)	15 000	14 400		
CODs (mg/L)	13 500	14 000		
DOC (mg/L)	5 580	6 200		
BOD₅ (mg/L)	7 250	8352		
BOD ₅ /COD	0.48	0.58		
Absorbance at 254 nm (a.u.)	3.85	1.75		
Absorbance at 475 nm (a.u.)	12.88	16.38		
Key: CODT – Total COD CODs –	Soluble COD D	OC – Dissolved organic carbon		

For the DWW, ozonation pretreatment increased biodegradability, as indicated by an increase of 21% in the BOD₅:COD ratio. The complex colour-causing biorecalcitrant aromatic compounds such as melanoidins are broken down to biodegradable acidic intermediates and simple aliphatic compounds through ozonation. The reduction of the aromatics contributed to the reduced absorbances at 254 nm for aromaticity (3.85 to 1.75 a.u.) and 475 nm for colour (16.38 to 12.88 a.u.). Also, the ozonation process achieved solubilisation of suspended organic solids, releasing them into the aqueous face, as shown by the reduction in total COD but with an increase in soluble COD. However, the total COD reduction was very low (15 000 to 14 400 mg/L), ensuring adequate biomass retention for the ensuing AD.

3.2. Effect of ozone pretreatment on anaerobic digestion of WAS

The COD and TSS reductions during anaerobic digestion of raw WAS and ozone pretreated WAS are given in Figures 2a and b, respectively. The pre-treated WAS had better COD and TSS reductions during anaerobic digestion than the raw WAS. Ozonolysis pretreatment led to the rupture of the hard cell walls and partial solubilisation (as shown by the 29% reduction in TSS in Table 1) of the sludge, availing the cellular contents and leading to improved degradation by the microorganisms. The most significant effect of the pretreatment process on AD was observed in biogas production, as shown in Figure 3. A comparison of the biogas production profiles (Figure 3a) showed that the pretreated WAS had more than double daily biogas production than the raw WAS. The daily biogas production was higher for the pretreated WAS at the beginning of digestion and remained relatively high with continued digestion than the raw WAS, which had a significantly diminished biogas production after the first day. The pre-treated WAS had a higher cumulative biogas production of 4.75 L after six days of digestion than the raw WAS which had 1.48 L (Figure 3b). Ozonolysis pretreatment solubilised part of the organic matter, which was then easily converted to biogas. The significant increase in biogas production indicates that a major fraction of the solubilised matter was biodegradable (Kim et al., 2013; Sosnowski et al., 2008).



Figure 2; (a) COD and (b) TSS reductions during anaerobic digestion of raw (Δ) and ozone pretreated (\blacktriangle) waste activated sludge.



Figure 3; (a) Daily and (b) cumulative biogas production during anaerobic digestion of raw (a) and ozone pretreated (a) waste activated sludge.

3.3. Effect of ozone pretreatment on anaerobic digestion of DWW

The non-pre-treated DWW and the ozonated DWW were subjected to AD, and the changes in COD (Figure 4a), biogas production (Figure 4b), and colour (Figure 4c) were monitored. The COD removal during the digestion of raw DWW increased from 32% on day 34 to around

63% by the 37th day. The low COD reductions observed during the initial stages (days 34 to 36) were partly due to the acclimatisation of the microorganisms to the new feeding regime (semi-continuous feeding from batch). The highest reduction in COD averaged around 70% from the 42nd to 53rd day. To investigate the effect of pretreatment, ozone pretreated DWW was fed into the reactor from day 55 to 74 at the same OLR of 15 kg/m³/d. During this period, the average COD removal remained constant at 71%, indicating that ozone pretreatment did not significantly affect the AD process, despite the significant increase in biodegradability of the pretreated DWW (Table 2). A similar observation was made with the daily biogas production, which averaged 10 L/day for raw and ozone pretreated DWW (Figure 4b). The lack of observable differences in the anaerobic digestion of the raw and pretreated substrates could be because of the relatively high biodegradability of the raw DWW, with only 2% being biorecalcitrant (Chavan et al., 2006).



Figure 4; (a) Reduction in COD, (b) daily biogas production, and (c) daily colour reduction during anaerobic digestion of raw and ozone pretreated DWW.

A negative colour reduction indicated increased colour intensity (Figure 4c) during the anaerobic digestion of the raw and ozone pretreated DWW substrates. Under the mesophilic conditions of the UASB reactor employed, the melanoidin compounds were repolymerised into high molecular weight (usually > 5.0 kDa) long chain organics, thereby increasing the effluent's colour intensity (Liang et al., 2009; Liu et al., 2013). Ozone pretreatment was expected to eliminate the melanoidins before AD and improve colour reduction during AD. However, the increased colour intensity points to incomplete removal of the melanoidins during ozonation pretreatment. The melanoidins that remained after ozonolysis easily repolymerised during AD, increasing the colour intensity. Ozone pretreatment is, therefore,

ineffective in fully reducing the biorecalcitrant melanoidins into biodegradable compounds and ensuring complete colour removal during AD.

3.4. Ozonolysis Post-treatment of Anaerobically DWW

Table 3 shows that the DWW had a significant organic load before anaerobic digestion (BOD₅ 7,200 mg/L, COD 15,000 mg/L). The BOD:COD ratio of 0.48 indicated that the DWW substrate was highly biodegradable (a ratio of 0.4 and above is recommended). Up to 75% of the COD and 95% of the BOD5 were eliminated during biodegradation, although the colour intensity was enhanced by 40%. After AD, there was still a sizeable quantity of COD present (3560 mg/L), responsible for the biorecalcitrant component, mainly melanoidin compounds that were the source of the intense colour of the AD effluent. The effluent had a BOD5:COD ratio of 0.05, confirming the elimination of all the biodegradable organics by AD. Sludge washout from the digester was indicated by the twofold increase in the total suspended solids. To remove the colour and solubilise the solids (sludge), ozonolysis posttreatment was applied to the anaerobically digested DWW effluent.

Parameter	Before	After	Change, %	
рН	5.5	7.45	+62	
COD (mg/L)	15000	3560	-76	
DOC (mg/L)	5800	1700	-72	
TSS (mg/L)	2.34	4.46	+110	
BOD ₅ (mg/L)	7250	190	-95	
BOD ₅ :COD	0.48	0.05	-90	
Colour/absorbance (a.u.)	3.20	4.50	+41	

Table 3: Characteristics of DWW before and after AD

The ozonolysis posttreatment of the anaerobically digested AD resulted in up to 80% colour removal and a 14% reduction in DOC. The oxidation of the melanoidin molecules and the mineralisation of biorecalcitrant organic substances led to reductions in colour and DOC. Color-causing organic compounds with aromatic rings, functional groups like OCH₃ and OH, carbon double bonds (C=C), and atoms like N, P, O, and S (negatively charged) are selectively attacked by ozone (Bar Oz et al., 2018). The attack can lead to a rapid colour disappearance but with the formation of end products such as carboxylic acids and stable intermediates. The products formed are still detectable as DOC; thus, the observed low DOC reduction of 14%. Moreover, the products and intermediates are usually refractory to further oxidation by ozone (Peña et al. 2003). Ozone can also break down, resulting in the generation of hydroxyl radicals (OH \bullet). The radicals can react (unselectively) with all organic compounds, potentially leading to total mineralisation via chain degradation reactions (Kasiri et al., 2013; Mecha et al., 2016; Wang et al., 2004).

Figures 6a and b show the changes in total (COD_T) and soluble (COD_S) COD and the BOD₅:COD ratio, respectively, during ozonolysis posttreatment of the AD effluent. Total, soluble, and suspended COD can be used to investigate the fate of sludge during ozonation. The COD_T reduced to 1250 from 1438 mg/L (13% reduction), while the COD_S was unchanged. The decrease in COD_T was attributed to the solubilisation of sludge washed out ((suspended COD) from the AD reactor. After one hour of ozonolysis, the COD_T and COD_S levels were nearly similar, indicating that up to 88% of the sludge had been solubilised. Previous investigations on the treatment of DWW with ozonolysis found low COD_T decreases within the first hour of treatment (Zeng et al., 2009; Sangave et al., 2007). The suspended COD (TSS) is dissolved during ozonolysis resulting in constant CODs, but with a reducing COD_T. After the posttreatment, the BOD₅:COD ratio of the AD effluent increased twofold (0.05 to 0.11), indicating improved biodegradability (Venkatesh et al., 2015).



Figure 5; Colour (a) and DOC (b) reductions during ozonolysis of anaerobically digested DWW.



Figure 6; Change in (a) COD_T (\circ) and COD_S (Δ) and (b) BOD_5 :COD ratio during ozonolysis of anaerobically digested DWW.

Conclusion and recommendations

An integrated AD-ozonolysis treatment system is a promising technique for treating distillery wastewater (DWW) and waste activated sludge (WAS). Ozonolysis pretreatment of WAS solubilised the sludge and led to a two-fold increase in the cumulative biogas production in the ensued anaerobic process. In the case of distillery wastewater, ozonolysis pretreatment did not significantly impact COD reduction or biogas production; however, when used for post-treatment, ozonolysis effectively removed the biorecalcitrant colour of the AD effluent and solubilised the TSS washed out from the AD unit. The configuration of the intergrade anaerobic digestion-Advanced oxidation process (AD-AOP) treatment system is thus substrate-specific. Ozonolysis should precede AD when treating WAS, while for DWW, ozonolysis should be applied as a posttreatment to AD. Kinetics and energy analyses should be determined to guide in designing an integrated AD-ozonation process for WAS and DWW treatment.

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