Efficiency of an integrated process of electrooxidation and anaerobic digestion of waste activated sludge

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ABSTRACT

Background: Most of the organic content of waste activated sludge (WAS) comprises microbial cells hard to degrade, which must be pre-treated for the energy recovery by anaerobic digestion (AD). Electrooxidation pre-treatment (EOP) with a boron-doped diamond (BDD) electrode have been considered a promising novel technology that increase hydrolysis rate, by the disintegrating cell walls from WAS. Although electrochemical oxidation could efficiently solubilise organic substances of macromolecules, limited reports are available on EOP of WAS for improving AD. In this endeavour, the mathematical optimization study and the energy analysis of the effect of current density (CD) during EOP and the initial total solids concentration [TS] from WAS on methane (CH₄) production by AD was investigated.

Results: In the present work, biogas production from WAS conversion are comprehensively affected by CD and [TS]. The highest COD and VS removal by 60 and 39% respectively, were achieved with WAS at 3% of [TS] pre-treated at CD of 24.1 mA/cm², and with a maximum CH₄ production of 305 N-L/kg VS and a positive energy balance of 0.83 kWh/kg VS. Therefore, the low current densities used in boron-doped diamond (BDD) electrode are adequate to produce the strong oxidant ●OH radical on the electrode surface, allow the oxidation of organic compounds that favours the solubilization of COD from WAS.

Conclusions: The improvement of VS removal indicates that EOP help to disintegrating cell walls from WAS. This allows a decomposition reaction that leads to biodegrade more compounds during AD. The energy balance was positive, suggesting that even without any optimization the energy
used as electricity could be approximately recovered as energy from the increased methane
production. However, this kind of analysis have not been sufficiently studied so far, therefore, is
important to understand how critical parameters can influence the pre-treatment and AD
performances. The current study highlights that the mathematical optimization and energy analysis
can get the whole process more convenient and feasible.

**Keywords:** Anaerobic digestion; current density; electrooxidation pre-treatment; energy analysis;
waste activated sludge.

**BACKGROUND**

The activated sludge process is currently one the most widely used biological wastewater treatment
in Latin America, especially for municipal effluences [1,2]. In municipal WWTP, the removal of
biodegradable compounds by conventional biological aerobic systems are generating a larger
amount of waste activated sludge (WAS). In last decades, most widely applied practices for sewage
sludge disposal are land application (use as a fertilizer in agricultural field), incineration and
confinement in landfill. However, the contamination of this waste by pathogens, heavy metals,
polycyclic aromatic hydrocarbons, polychlorinated biphenyl or dioxins, limits their harnessing.

Therefore, the management of excess WAS (treatment and disposal) represent an issue of concern
and most challenging task for the wastewater treatment sector.

Additionally, activated sludge WWTPs are swiftly becoming a high-cost item on municipal budgets
amidst the rising electricity tariffs and by increasing of the CO₂ indirect emissions being attributed
to mainly to higher energy consumption and sludge production [3]. Aerobic processes convert a
substantial part (about 50 – 60 %) of the wastewater pollution into sludge, without considering
primary suspended solids removal [4]. WAS is the excess biomass from suspended-growth aerobic
wastewater treatment systems. Most of the organic content of WAS consists of microbial cells.
These are hard to degrade as their cell wall and membrane are composed of complex organic
materials such as peptidoglycan, teichoic acids, and polysaccharides that are not readily
biodegradable and they serve as a protective cover to resist osmotic lysis [5,6]. For these reasons, the use of WAS as renewable source of energy has scarcely been studied at all [7].

One of the most commonly used sludge biological treatment processes is anaerobic digestion (AD); it is estimated that 70% of the sludge are stabilized by this method [8]. This process has a major advantage as biogas is produced, for use as an energy source and could play a central role in the interconnected biofuels infrastructures of the future [9]. However, most of the organic content of WAS comprises microbial cells which significantly reduce the hydrolysis rate [10,11]. In order to enhance the efficiency of anaerobic digestion of WAS, the rate of hydrolysis needs to be increased applying pre-treatments previously.

A number of different pre-treatment operations and processes have been proposed including biological, chemical, enzymatic, thermal and mechanical [11–13]. WAS pre-treatments offer the following advantages: (a) enhance cell lysis, (b) more bioavailable organic matter can be transformed into biogas, (c) the mass is further reduced and (d) minimal pollution through unpleasant odours. As a result, pre-treatment may stabilize better the WAS and increase more than 50% the methane (CH₄) produced, reaching 0.31 m³ of CH₄ per kilogram of total sludge eliminated (equivalent to 3.41 kilowatt-hours), succeeded in recapturing between 60-100% of the energy demand, depending on type of treatment technology applied [14].

Not long ago, the use of sludge EOP has been explored as a field of interest, considering the high oxidation capacity of chemical species formed at different electrode surfaces (for example physisorbed *OH radicals or homogenous species formation like HClO) [15]. Likely, the following simplified reactions at non-active anodes may take place for the electrooxidation of most organic components in WAS:

\[
R_{\text{WAS organic compounds}} + M(\cdot \text{OH}) + H^+ + e^- \rightarrow M + ROx_{\text{solubilized COD}} + H_2O \quad \text{(Eq. 1)}
\]

Electrooxidation process transfer of organic material from the particulate matter of the WAS to the soluble fraction (facilitating biogas formation). This is because high capacity of electrochemical
hydrolysis is provided by short-lived and energy rich free radicals that carry out disintegrating microbial cell walls. As recently proposed by Pérez-Rodríguez et al. [16], hydrolysis rate can be improved if the critical engineering aspects of reactors used for this purpose are identified. The primary drawback of EOP is associated to high-energy consumptions, due to operating inefficiencies [17].

Although electrochemical oxidation could efficiently decompose the organic substances of macromolecules to smaller ones, limited reports are available on EOP of WAS for improving CH₄ production. Despite the boron-doped diamond (BDD) electrode has been widely reported as one of the most stable materials for electrochemical applications [18], more research is needed on its implementation as a WAS pre-treatment for the successive AD. In order to reduce the pre-treatment associated energy consumption, this study proposes a novel approach using electrooxidation with a BDD electrode for improving BMP and substantially increase volatile solids (VS) removal during the mesophilic anaerobic stabilisation. For this purpose, the effect of current density during EOP and the initial total solids concentration on CH₄ production by AD of WAS was assessed. In addition, a mathematical optimization study and the energy analysis of the whole process as a function of these critical parameters is presented.

RESULTS AND DISCUSSION

Current density effect of electrooxidation pre-treatment

The kinetics of the AD of WAS is distinct from those for primary sludge, because WAS is inherently less biodegradable and sludge pre-treatment is suggested [19]. From experimental factorial design results, an improvement of the measured parameters like solubilised COD and of course, biogas production could be observed, when an increase occurs in the anodic current density. This is likely attributed to the fact that the current densities used in BDD electrode are adequate to produce the strong oxidant *OH radical on the electrode surface, allowing the oxidation of organic compounds that favours the solubilisation of COD from WAS [15,20]. In addition, formation of
homogeneous strong oxidants at low current densities improves the conversion of organic matter into soluble COD, with no noticeable chemical degradation during the EOP [21,22].

**VS reduction and COD removal**

VS removal efficiency of at least 38% is considered as an indicator of proper sewage sludge stabilization, according to the Report for Control of Pathogens and Vector Attraction in Sewage Sludge [23]. The maximum VS removal efficiencies were reached with low TS concentrations (1 and 2 %) and with current densities of 21.4 and 28.6 mA/cm², which were between 47 and 42% respectively. Even though VS removal efficiency did not meet the U.S. EPA standard at the higher solids concentration (3%), EOP increased this indicator to 35% if compared to the control value (17.6%), under the same conditions (Figure 1a). In all cases, VS removals were higher for the WAS pre-treated, if compared with the 14.2±1.2 % removal obtained with the unpre-treated WAS.

*Figure 1*

The effects of TS in WAS and current density during electrooxidation pre-treatment shown a decreased intensively in the VS removal efficiency with increase in TS concentration, providing a low level of solids destruction (<38% of the original WAS) loaded to each assay.

The equation 2 was used to visualize the effects of operational parameters on VS removal under optimized conditions in the 3D graphs of Fig. 1a. ANOVA analyses shows that the variability of VS removal efficiency (VS_{RE}) for each of the treatments, in this case, the effects have a P-value of less than 0.05, indicating that they are significantly different from WAS unpre-treated with a confidence level of 95.0%, and a correlation with the following fitted model was obtained:

\[
VS_{RE}(\%) = 13.95 + 2.17[CD] + 7.29[TS] - 0.03[CD]^2 - 2.28[TS]^2 - 0.32[CD][TS] \quad \text{(Eq. 2)}
\]

Nevertheless, the highest soluble COD removal was achieved with the EOP of 28.6 mA/cm² of current density regardless of TS concentrations, which could be accounted for the disintegration and solubilisation of WAS as mentioned above (Figure 1b). On this regard, it has been reported that the formation of hidroxiradicals may be formed at 10 mA/cm², so the fact of the COD solubilisation
increases as a function of current density, indicates possible formation of other strong oxidants species, of the kind of RO₂, in bulk solution increasing the reaction between solid particles and oxidants [24]. Consequently, COD solubilisation and VS removal depend from the contact between solid particles and physisorbed radicals, and thereby a slight increasing effect on VS removal with the current density should be observed [25].

Based on the removal improvement with an increase of current density, EOP appears to be a promising pre-treatment process. In contrast to VS removal, the maximum soluble COD removal efficiencies were reached with the high TS concentration and with current densities of 21.4 and 28.6 mA/cm², which were between 52.7 and 53.4%. In all cases, COD removal were higher for the electrooxidation pre-treatments, if compared with the 18.2±0.6 % obtained with the unpre-treated WAS (Figure 1b). In this case, ANOVA analyses shows the variability of COD removal efficiency (COD_RE) of 2 effects have a P-value of less than 0.05, indicating that they are significantly different with a confidence level of 95.0%, and from the effects correlation the following fitted model was obtained:

\[
\text{COD}_\text{RE} = 16.73 + 1.00[\text{CD}] - 2.04[\text{TS}] - 0.001[\text{CD}]^2 + 0.71[\text{TS}]^2 + 0.16[\text{CD}][\text{TS}] \quad (\text{Eq. 3})
\]

These results of the Figure 1 and equations 2 and 3 allow identifying the current density and TS concentration for producing half of COD soluble and the relative VS removal efficiency for the EOP sludge was identified.

In EOP, current density of 19.3 mA/cm², flow rate of 4 L/min and treatment time of 30 min are required to large molecules contained in sludge particles and microbial cells were partially solubilized, demonstrating that, the process is controlled by mass transfer [16]. The observed result is similar to that of results reported in the literature [26]. However, the electrolysis at current density higher than 30 mA/cm² decrease of soluble COD of WAS in comparison to lower currents densities, because which might lead the acceleration of organic matter mineralization than the solubilization reaction. Thus, it is advisable to limit the current density to avoid adverse effects such as heat...
generation and higher power consumption [27]. As result of this part of study, the electrolysis treatment at current density of 28.6 mA/cm$^2$ allowed a fast WAS hydrolysis and the best degree of disintegration.

**Current density effect of the EOP on BMP**

A slow biogas generation process was observed in the initial period in all assays, which took around 10 days for 50% total biogas generated. BMP assays of electrooxidized WAS confirmed results obtained from COD solubilisation. Electrooxidation pre-treatment enhanced the methane production of WAS from 109±4 N-L CH$_4$/kg VS in unpre-treated WAS to 312±6 N-L CH$_4$/kg VS with WAS en 3% of TS and EOP to 28.6 mA/cm$^2$ (Fig. 2). This 65% increase (203 N-L CH$_4$/Kg VS), which is more than expected from soluble COD obtained theoretically and suggests that the VS disintegration and solubilisation resulted from firstly the rapid sludge disintegration during the electrooxidation pre-treatment and then organics available for slow conversion during the anaerobic digestion. A correlation between the BMP and the two experimental variables (CD and TS) may be proposed Eq. 4. ANOVA analyses shows the variability of methane production of treatments have a P-value of less than 0.05, indicating that they are significantly different with a confidence level of 95.0%.

Figure 2 shows a correlation with the following fitted model:

$$\text{BMP} \left( \frac{\text{N-L}}{\text{kg VS}} \right) = -0.99 + 13.79[\text{CD}] + 32.78[\text{TS}] - 0.30[\text{CD}]^2 - 3.66[\text{TS}]^2 - 1.38[\text{CD}][\text{TS}] \quad (\text{Eq. 4})$$

This suggest, as was mentioned before, that other phenomena occur during electrooxidation and they favour solubilisation of organic matter which then favours anaerobic digestion. As a result, the improved methane production indicates that the impact of the rate-limiting hydrolysis step could be reduced by electrooxidation pre-treatment. Results in Figure 2 show that the current density had an impact on the methane production from WAS. The methane production increased proportionally with both TS concentration and current density. This is explained by the fact that the EOP itself (in a single chamber without pH change) could disrupt cell membranes in WAS and therefore enhance biodegradation in subsequent anaerobic digestion [28].
The calculation of maximum BMP as function of CD and TS from equation 4 was evaluated using non-linear complex method [29]. Constrain values employed for this calculation are:

\[ 1 \leq [\text{TS}] \leq \text{TS}_{\text{op}}, \text{with } \text{TS}_{\text{op}} = 1 - 3.5\% \]  
(Eq. 5)

\[ \text{CD} \leq 35 \text{ mA/cm}^2 \]  
(Eq. 6)

This constrains were stablished since a low WAS particle dispersion during hydrodynamic tests was observed and the current densities recommend to produce \( \bullet \text{OH} \) radicals in BDD electrode is < 20 mA/cm\(^2\) [30]. In this study, current densities greater than 20 mA/cm\(^2\) were choose because the possible hindering of electrode area during particle-electrode interactions [31]. The effect of increasing CD and TS on BMP was evaluated with Eq. 4, resulting in a higher BMP values, as shown in Figure 2 and Table 1. The BMP depends directly on the density of applied current, finding the maximum methane production in the extreme values of TS. However, a limit would be solid concentrations higher than 3.5%, due to concentrations of solids greater than 3% reduce the useful life of the electrodes.

**Table 1**

**Energy analysis**

While methane production was significantly improved through EOP, there was also consumed as thermal and electrical energy. For industrial application of a suitable pre-treatment the energy invested in this process should be obtained as an additional methane yield. The energy consumption of the described electrooxidation process can be calculated according to the following equation [32,33]:

\[
W (\text{kW-h/kg VS}) = \frac{(V \times A \times t)}{[\text{VS}]} / 1000
\]  
(Eq. 7)

Where, \( V \) is the average supplied voltage, \( A \) is the amps, \( t \) is the operation time in hours and \([\text{VS}]\) is initial volatile solids mass in kg.

Under the best conditions (21.4 mA/cm\(^3\) and 3% of TS), the energy consumption of the EOP was 1.17 kWh/kg VS, suggesting that even without any optimization the energy used as electricity could
be approximately recovered as energy from the increased methane production (305 N-L CH₄/kg VS), which can produce about 3.43 kWh/kg VS. A summary of performance and energy outcomes for the major pre-treatments and options is given in Table 2. The calculations were based on the verified information from the various sources and solids concentration, as kg VS, were taken as the basis. A nominal VS:TS ratio of 59.2% was used. Calorific values and heat capacities have been taken from standard texts [33]. The pre-treatment methods used for the comparison were the more widely industrially applied and under the best possible conditions [32]. Considering the electrical and thermal available energy, for the sludge pre-treated by EOP, the cogeneration would produce approx. between 0.5 – 1.11 kWh/kg VS as electricity and 0.71 – 1.72 kWh/kg VS as heat, respectively (Section 2.1, supplementary material). All options for pre-treatment have substantial energy consumption, and thermal hydrolysis and ball mills have the highest energy consumption. In EOP; the low energy expenditure was due to the fact that only low current densities for short periods of time are required for WAS pre-treatment [20,26]. In this work, energy costs for agitation, pumping and heating were considered, because often the energy balance by these extras exceed substantially the energy use that spent during the pre-treatment [34].

**Table 2**

We have therefore a variation of COD and VS removal after anaerobic digestion, and each pre-treatment gave an advantage in COD removal improvement compared to un-pre-treated sludge. The highest COD and VS removal were achieved with sludge pre-treated at 3% of TS and current density of 21.4 A m⁻², and thus the maximum methane production was achieved. EOP is feasible based on the literature and this work. The energy balance of pre-treatment sludge technology would be less than that of unpre-treated WAS as the energy consumption of the other methods are comparable while the anaerobic digestion with sludge pre-treatment is less than that of non-treated sludge. Conventional anaerobic digestion requires 18 to 25 d of sludge retention time, sometimes even 30 d, with the conditions of 20 °C temperature and 2% solid content. Most importantly, EOP
can significantly decrease anaerobic digestion time. The digestion time after EOP decreased from 30 d to 16 d as shown in the paper, in other words, electrochemical pre-treatment could reduce roughly 26% of anaerobic digestion reactor volume, which is crucial for utilities with limited site spaces.

MATERIALS AND METHODS

The experimental work was focused on BMP assays from pre-treated WAS. These were compared with results of unpre-treated WAS used as reference.

Sludge samples

Samples of WAS were collected from the Cerro de la Estrella wastewater treatment plant (WWTP), Iztapalapa, Mexico City. This facility treats 2300 L/s of municipal sewage using a conventional activated sludge process. WAS concentrations being as follow: TS (80 g/L), VS/TS fraction (59.5±11.4 %), total chemical oxygen demand (COD) (23.0±6.1 g/L), soluble COD (2.4±0.9 g/L), carbohydrates (29.0±5.3 % of VS), protein (4.3±3.2 % of VS) and oil and grease (1.0±0.9 % of VS).

Inoculum source

Anaerobic sludge used as inoculum was collected from a brewery WWTP. This was initially employed for the start-up of a seed digester fed with unpre-treated WAS. Once the seed reactor reached steady state, the resulting sludge was used as inoculum for BMP assays.

Electrooxidation pre-treatment (EOP)

A Diaclean® electrochemical cell composed by two circular electrodes and two spacers was used for the experiments. The pre-treatment assays were carried out in a single compartment of the electrochemical cell. Diamond-based material (p-Si–BDD) was used as anode and cathode. Both electrodes were circular (100 mm diameter) with a surface area of 70 cm$^2$. The relevant dimensions of the electrochemical reactor are similar to the reported by Barrios et al. [31]. The electrochemical reactor was coupled to a hydraulic system consisting of a 4 L reservoir made of glass and a peristaltic pump (JP Selecta Per-com N-M328). Tubes, valves, and accessories were made of PVC.
Sludge was stirred in the glass reservoir with an overhead mixer (stainless steel paddle area: 49 cm²) to avoid solids settling. The stirrer speed was low (100 rpm) to keep the sludge homogeneous and avoid phase separation at the reactor entrance. Power was supplied by a Delta Elektronika ES030-10, applying current densities of 14.3, 21.4 and 28.6 mA/cm² during 30 min. The temperature in the reservoir was kept constant (25 °C) with a water bath system.

**Biochemical methane potential (BMP) assays**

The anaerobic digestion for untreated (pre-treatment control) and pre-treated WAS was measured in an OxiTop® Control OC 110. BMP assays were performed with a working volume of 80 mL, in 250 mL flasks and the increase of pressure inside the headspace were stored in the OxiTop measuring head at everyday intervals automatically. BMP assays were carried out at mesophilic temperature (36±2 °C) during 16 days; initial pH was adjusted to seven and flasks shaken at 150 rpm. The amount of WAS and inoculum were calculated using a substrate/initial biomass (S/X₀) ratio of 0.5 g VS_{fed}/g VS_{biomass}. Optimization of the selected operating conditions was assessed by the response surface methodology. A 3-level full factorial design was performed (Table 3), the factors were the WAS concentration as total solids (1.0, 2.0 and 3.0 % (w/v)) and the current density of the EOP (0 as control, 14.3, 21.4 and 28.6 mA/cm²). The influence of treatments was separated into the main effects of total sludge concentration versus current densities and the interaction between these two factors. The controls used were a negative control (inoculum without substrate) to determine the endogenous production of CH₄ and a bottle with clean water at the same volume to correct pressure measurements of the system.

**Table 3**

**Analytical methods**

Total solids (TS), volatile solids (VS), fixed solids (FS), pH, total alkalinity, and soluble (CODₕ) and total (CODₗ) chemical oxygen demand and oil and grease were determined according to the Standard Methods [35]. CODₕ and CODₗ were analysed on 1:20 and 1:100 sludge dilutions,
respectively. Alkalinity ratio ($\alpha$) was determined as the quotient between partial (pH 5.75) and total alkalinity (pH 4.3). The concentration of volatile fatty acids (VFA) was measured by gas chromatography (SRI 8610-10) with flame ionization detector, N$_2$ as carrier gas using an Alltech EC-1000 column. Biogas volume was quantified by the OxiTop® system, while their composition was analysed by gas chromatography (Fisher Gas Partitioner chromatograph model 1200) with thermal conductivity detector, He as carrier gas and a Porapak Q column.

Maximization of methane production

From the experimental data obtained at different [CD] and [TS] values, the removal of COD and VS and the methane production were theoretically maximized, through a mathematical optimization analysis. To maximize the equations 2, 3 and 4, a nonlinear complex method was used. Once the objective function is determined, its derivatives are calculated and the critical point (where objective function is maximized) is obtained by means of Hessian matrices method. The methodology was implemented in Excel and mathematical details of this procedure are showed in results section. The purpose of this data treatment was to determine the conditions associated with the maximum CH$_4$ production.

Statistical analysis

Statistical analysis of the BMP assays results was carried out using STATGRAPHICS Centurion XVI version 16.1.03 software. The analysis of variance (ANOVA) test was implemented to evaluate if differences could be observed between the different current densities for each sludge concentration, after which post hoc multiple comparison was carried out by means of the Tukey HSD test at the 5% significance level. In all BMP assays, methane yields were reported as the average of replicate samples (as mean ± standard deviation).

CONCLUSIONS

The application of electrooxidation pre-treatment at different TS concentrations, for improving WAS anaerobic digestion. The effectiveness of this method was compared to unpre-treated WAS.
The highest COD and VS removal were achieved with sludge pre-treated at 3% of TS and current density of 21.4 mA/m². The maximization of biogas production indicates that the maximum degradation and methane production depends directly on the applied current density. This study shows a high prospective of electrochemical pre-treatment to be implemented with anaerobic sludge stabilization, because was produced 305 N-L CH₄/kg VS equivalent to approximately 0.77 kWh/kg VS as electricity and 1.09 kWh/kg VS as heat, respectively.

**ABBREVIATIONS**

AD: anaerobic digestion; ANOVA: Analysis of variance; BDD: boron-doped diamond; BMP: biochemical methane potential; [CD]: current density; COD: chemical oxygen demand; CODₚₑ: COD removal efficiency; CO₂: carbon dioxide; EOP: electrooxidation pre-treatment; •OH: hydroxyl radicals; N-L CH₄: normalized methane; S/X₀: Substrate/substrate/initial biomass ratio; [TS]: total solids concentration, VFA: volatile fatty acids; VS: volatile solids; VSₚₑ: VS removal efficiency; WAS: waste activated sludge; WWTP: wastewater treatment plant.

**AUTHORS’ CONTRIBUTIONS**

U-D and ME-C designed and carried out the experiments, performed the data analysis, and drafted the manuscript. J-B and A-C carried out the pre-treatment method, participated in the setup operation of assays. U-D, J-B and FF-R participated in data analysis and revised the manuscript. J-B, A-C and U-D conceived of the study, put forward the hypothesis, and gave the final approval of publication. All authors read and approved the final manuscript.

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Figure captions:

**Figure 1.** (a) VS reduction and (b) COD removal, in response to different CD and TS.

**Figure 2.** Effect of CD applied and TS on BMP under mesophilic anaerobic digestion.