



# Biogas from anaerobic co-digestion of chrome and vegetable tannery solid waste mixture: Influence of the tanning agent and thermal pretreatment



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## ABSTRACT

Mesophilic anaerobic co-digestion of mixtures of solid wastes from tanneries containing chromium and vegetable tannins was investigated in an orthogonal array. The effect of thermal pretreatment on shavings was evaluated as well. The biogas and methane productivity in terms of VSS was assessed. The treatment efficiency was also evaluated in terms of variation of organic and inorganic load. All assays with chromium-containing sludge showed on average 19.6 ml of biogas/gVSS added, 8.15 ml of methane/gVSS added, 4.8% VSS reduction, 60.4% BOD<sub>5</sub> reduction, 55% TOC reduction, 67% IC increase and 54.6% TN increase, showing greater mineralization of the residue when compared to the assays with vegetable tannin sludge, which has proven to be toxic. The origin of shavings was not significant in any parameter due to the small proportion of this residue in assays. The thermal pre-treatment was beneficial only for assays with vegetable tannins, since there was evidence that the heating mechanism degraded part of the phenolic organic matter and consequently reduced its toxicity.

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## 1. Introduction

With rapid depletion of conventional energy sources, the need to find an alternative, preferably renewable, energy source from waste is becoming increasingly important for sustainable development as global energy demand is increasing at dramatic rate while fossil fuels reserves are rapidly depleting (Sumprasit et al., 2017; Thangamani et al., 2010). Thereby, a new alternative of re-using different types of tannery solid waste, such as in anaerobic co-digestion of solid wastes for biogas production is being developed and it is regarded as a clean energy technology that is able to convert energy directly from organic waste by microorganism (Ratanatamskul and Manpetch, 2016; Zupančič and Jemec, 2010). Furthermore, use of non-renewable energy sources also results in release of greenhouse gases leading to increased CO<sub>2</sub> concentration in the earth's atmosphere; thus impacting the climate change.

On the contrary, renewable energy sources such as biomass have much smaller life time carbon emissions (Sumprasit et al., 2017).

Secondary products from leather industries are regarded as animal wastes. Conversion of these animal wastes into fuels represents an energy recovery solution not only because of their good combustion properties, but also from the viewpoint of supply stability (Lazaroiu et al., 2017). Tanning processes are classified according to the type of tanning reagent used to bind the collagen fibers. Chromium sulfate is the most important tanning agent used (supply of 3% of the weight of the hide). Another popularly used tanning agent is vegetable tannins (supply of 30% of the weight of the hide) (Dhayalan et al., 2007). The release of these compounds to the environment is inevitable. Their presence may lead to various mechanisms that may cause damage to exposed organism, such as damage cell membrane, protein, DNA, interrupt electron transportation, or release toxic components (Nguyen et al., 2015).

The leather industry produces two major categories of waste, namely animal fat and residues containing mostly proteins, suitable for biogas conversion (Cernat et al., 2015; Lazaroiu et al., 2017). Shavings are generated in the leather processing operation (Piccin et al., 2013). During tannery wastewater treatment, a large volume of sludge is generated. On average, sludge treatment accounts for approximately half of the total cost of wastewater treatment. Thus,

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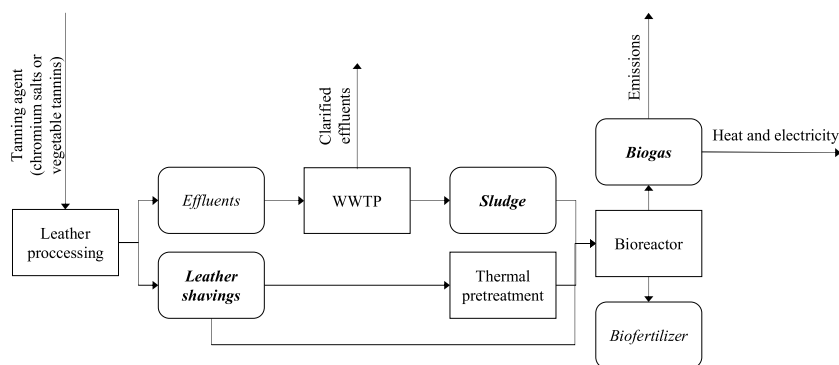


Fig. 1. Process flow sheet of the leather waste-based AD setup.

sludge management is a growing challenge (Nghiem et al., 2017). Even with all the problems involving the use of chromium in the tanning process (Fuck et al., 2011), a small amount of it remains in the residues, so they appear to be favorable for anaerobic digestion (AD). Vegetable tannins show the opposite because of their strong antimicrobial characteristics. Moreover, disposal of such tannery waste has become more difficult in view of stringent environmental pollution control standards stipulated and in the short to medium term will no longer be sustainable (Appels et al., 2008).

Digestion process tends to fail, when one readily degradable organic matter is used as sole substrate without external nutrients and buffering agent. Anaerobic co-digestion was considered to reduce waste and convert to energy. The benefits of anaerobic co-digestion can improve: the dilution of toxic compound, improved balance of nutrients, increased load of biodegradable organic matter (Ratanatamskul and Manpetch, 2016). The mixture of residues of vegetable and chrome tanning and residues from different stages of the industrial process creates specific quality characteristics for treatment due to their different compositions (Mannucci et al., 2010).

Biogas has been of interest due to its diverse production sources, including industrial wastes, and potential uses, such as burnt directly or converted via various processes into other higher value fuels (Almomani et al., 2017). The composition of waste treated by AD has an effect on biogas yield and gas quality (Qiao et al., 2011). The biogas generated is mainly composed of 48–65% methane and 36–41% carbon dioxide. The methane yield is affected by many factors including the type and composition of substrate, microbial composition, temperature, moisture and bioreactor design. A number of biological transformations take place during AD, including hydrolysis, acidogenesis, acetogenesis, and finally methanogenesis. The first stage is of great importance, as complex carbohydrates, proteins, and lipids are broken down by the enzymes provided by the bacteria. In the methanogenesis stage, slow growing bacteria carry out the methanogenic transformation of waste into biogas (Almomani et al., 2017). There are some components in waste (metals and some recalcitrant organic compounds, often toxic) that do not break down and therefore become concentrated in the residue (Khalid et al., 2011). The remaining fraction in the digester, the digestate, can be further treated and processed, or can be used directly as a fertilizer (De Meester et al., 2012).

The application of AD to biosolids (biomass) is often limited by very long retention times and the low overall degradation efficiency of the organic dry solids. Various disintegration methods have hence been studied as a pretreatment: these methods disrupt cell walls which results in lysis or disintegration of sludge cells in order to promote the yield of biogas (Appels et al., 2008; Jin et al., 2015). There are different methods of pretreatment which includes physical, physiochemical, chemical and biological (Saidu et al., 2014). Thermal pretreatment is applied to improve sludge

dewaterability. It allows degradation of the sludge gel structure and release of linked water. If combined with AD, the pretreatment objective is not only to reduce the final amount of sludge to be disposed of, but also to increase methane production (Carrère et al., 2010).

After AD, the waste can be disposed of safely as its organic load is greatly reduced, besides being able to be destined for several applications, such as fertilizers and incorporation in cement. Even though the technology involved in AD is mature, there are still several parameters that can be optimized, especially with tannery residues, which can consequently improve its economics and continued relevance (Adu-Gyamfi et al., 2012). Table 1 reviews the main tannery solid waste AD studies that have been developed so far, the foci of which were the production of biogas with high methane content and treatment of the solids digested, analyzed through volatile suspended solids (VSS) variation.

A deep gap in knowledge still exists regarding the response of the AD system and its embedded microbial communities to mixed tannery waste. The objectives of this study were to improve and explore the mechanisms of AD of mixtures of the two leather industry solid residues of major concern: leather shavings and sludge from tannery wastewater treatment plants (WWTPs) by controlled co-digestion, and to compare the effect of chromium and vegetable tannins present in such waste for greater biogas production and greater waste treatment. Sludge was also used as inoculum for the treatment of waste (Khalid et al., 2011). The effect of two kinds of tanning agent (chrome and vegetable tannin) in these two types of solid waste and the use of thermal pre-treatment of leather shavings were analyzed using a factorial design. Fig. 1 summarizes the process flow sheet studied. This research attempts to add to the understanding of how tanning agents and pretreatment parameters interact to enhance biogas and methane production and organic and inorganic load variation in tannery solid waste mixed biodigestion.

## 2. Materials and methods

### 2.1. Solid waste collection and preparation

The leather shavings and sludge were obtained from two tannery factories and their respective WWTP located close to Porto Alegre, Brazil. One tannery used vegetable tannin as the tanning agent and the other used chromium salts. The samples were kept at room temperature prior to use. Thermal treatment was applied on leather shavings through autoclaving at 121 °C (1 atm) for 5 min.

### 2.2. Biodigestion experiments

The experiments were assembled to compare the three factors (sludge origin, shavings origin and thermal pretreatment

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