



# Intermediate ozonation to enhance biogas production in batch and continuous systems using animal dung and agricultural waste



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## ABSTRACT

Agricultural waste and animal manure (dung) pose an environmental threat in developing countries. This investigation focused on the possible use of such waste as an energy source in the form of biogas produced via anaerobic digestion (AD). The impact of single and mixed substrates on methane production under controlled batch and continuous experimental setups was considered. The study was extended to investigate the effect of substrate size and the impact of an intermediate ozonation process on enhancing the production of biogas from single and mixed substrates. Cumulative methane production (CMP), ultimate methane yield (UMY), methane production potential (MPP), methane production rate (MPR), and maximum methane production rate (MPR<sub>max</sub>) were used as performance indicators of the effectiveness of the anaerobic digestion process. CMP and MPP from mixed substrates were found to be higher than values obtained from a single substrate feeds, which may be attributed to a more balanced nutrient and organic matter found in mixed substrates. The large surface area of fine substrate influenced MPR and MPR<sub>max</sub> values in the first 30 days of digestion. In later AD stages, the effect of substrate size was negligible. The MPR<sub>max</sub> for fine substrates was  $12.3 \pm 0.3$  LkgVS<sup>-1</sup> compared to  $8.8 \pm 0.2$  LkgVS<sup>-1</sup> obtained for coarse substrate. Continuous AD with organic loading rate (OLR) of  $4 \text{ kgVSm}^{-1}\text{d}^{-1}$  showed a %AD<sub>efficiency</sub> of 62%, an average specific methane production in the range of  $98\text{--}230$  LkgVS<sup>-1</sup> and a volumetric methane production rate in the range of  $1.94\text{--}2.35 \text{ m}^3 \text{ m}^{-3}\text{d}^{-1}$ . Increasing the OLR increased the accumulation of volatile fatty acids in the system and resulted in decreased methane production. Two-stage AD with an intermediate ozonation process showed a significant increase in CMP and %AD<sub>efficiency</sub> compared with single-stage AD. The %AD<sub>efficiency</sub> for two-stage AD ranged from 63% to 83%, and for single-stage AD, it was in the range of 42.2%–64.3%. Anaerobic digestion of mixed agricultural waste improved the filtration, dewaterability, and settling ability of the final substrate, making it suitable for use as a soil fertilizer.

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## 1. Introduction

In recent years, great attention has been paid to renewable and sustainable sources of energy, manifesting itself in new governmental targets and directives worldwide (Herrmann et al., 2016; Gonzalez-Fernandez et al., 2015; Mahanty et al., 2014; Uggetti et al., 2014). Biomass is considered the fourth largest available source of renewable energy in the world (AL-Hamamre et al., 2014) and accounts for 3% of primary energy consumption in developed countries. However, biomass can contribute up to 35% of the

primary energy consumption in developing countries as reported by AL-Hamamre et al., 2014). Biogas has been of interest due to its diverse production sources and potential uses (Zhang et al., 2015; Yong et al., 2015; Regueiro et al., 2012; Kondusamy and Kalamdhad, 2014). Sources include municipal and agricultural waste, sludge, algae, animal manure (dung), and crops. The biogas can be burnt directly or converted via various processes (chemical, biochemical, and thermal) into other higher value fuels. Biogas is mainly a mixture of methane, carbon dioxide and other trace gases. The composition varies according to the source and method of production, but generally falls in the range of 40–70% methane and 15–60% carbon dioxide (Rodriguez et al., 2016). Anaerobic digestion (AD) is one of the longest known techniques, utilizing a wide variety of biodegradable material for the production of biogas

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through the activity of microorganism in the absence of oxygen (Fedailaine et al., 2015; Maamri and Amrani, 2014; Cheng and Hong, 2013).

Biogas as an energy source has proven to be an important strategy in solving the problems of energy in developing countries. By using agricultural waste and dung to produce biogas, households and communities can become more self-sufficient in terms of energy. Jordan is a none oil producing developing country that depends almost entirely on imported fuel from neighbouring countries. Given the current socioeconomic factors and the political climate in the surrounding countries, in addition to the dramatic increase in Jordan's population, the country faces serious challenges in its energy and waste management sectors. These challenges are forcing the government to look for alternative ways to supply its energy market (Abu Qdais, 2007). Reports show that around 997.25 kt of solid agricultural waste was generated in Jordan in 2011, possessing an energy content of 7.954 PJ (AL-Hamamre et al., 2014). Having said that, this waste makes no real contribution to the Jordanian energy sector. Some of the agricultural waste in Jordan is sent to landfills, added to soil, or used as animal feed, but most is incinerated in an uncontrolled manner (Abu Qdais, 2007). Al-Hamamre et al. (2014) reported that, if used, the energy availability factor for this residue could be estimated at 25% of the total energy demand of the country.

In addition to agricultural waste, dung represents a rich source of organic materials. The estimated amount of livestock manure (dung) produced annually in Jordan is reported to be around 3600 kt, with a total energy potential of 12.739 PJ (AL-Hamamre et al., 2014), rendering it a valuable source of energy in Jordan (Al Nasir and Batarseh, 2008; Jaber et al., 2015). The Jordanian government aims to increase the proportion of renewable energy to 7% by 2015 and to 10% by 2020. Accordingly, a need exists for an effective process that can treat such solid waste, reducing its hazardous effects and impact on the environment. Anaerobic digestion (AD) technology can play an essential role in reducing the amounts of ammonia and methane emitted from manure storage facilities (Gelegenis et al., 2007). Recent studies reported that the life-cycle assessment of AD was very positive when compared to other energy recovery processes, such as composting and incineration (Nallathambi Gunaseelan, 1997).

It is known that 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 (Mao et al., 2015; Kondusamy and Kalamdhad, 2014). The quality and quantity of the biogas produced from AD varies greatly and depends on a number of variables including the input substrate, digestion conditions, and reactor design. The concentration of solids, carbon to nitrogen ratio (C/N) of the substrate, temperature and pH are key factors that affect the performance of AD systems (Mahanty et al., 2014). The use of a single substrate (mono-digestion) showed low digestion performance due to a poor balance between the substrate constituents. For example, a substrate with high nitrogen or protein content can lead to low C/N ratios. Under anaerobic digestion conditions, high protein content in the substrate is known to inhibit acidogenic bacteria and methanogens due to the production of ammonia, which can lead to a reduction in methane production (Herrmann et al., 2016; Hassan et al., 2016; Nurliyana et al., 2015). On the other hand, mixed substrate methods (co-digestion), where more than one substrate is combined and used in the digestion process, is proposed by a number of researchers as an alternative cost-effective solution for reducing the adverse impact of ammonia

inhibition (Ajeej et al., 2015). It is generally considered that a value of C/N in the range of 20–30:1 is desirable for the prevention of ammonia inhibition effects and a balanced nutrient content (Ajeej et al., 2015). Various studies on improving the production of biogas from AD were conducted and reported on in literature. Some of the suggested improvements include the addition of accelerants in the form of chemical or biological additives. The additives lead to highly localized substrate concentration and favourable conditions for AD (Ajeej et al., 2015). Zhang et al. (2015) used biological additives including fungi, microbial consortia, and enzymes to improve biogas production. On the other hand, Kondusamy and Kalamdhad (2014) and Mao et al. (2015) used alkali and acid mediums in addition to other oxidative reagents and micronutrients to improve AD performance. Yuan et al. (2016) assessed the biogas production and anaerobic sludge digestion using the combination of electrochemical treatment and sodium hypochlorite. The combined processes significantly enhanced the biogas yields from sludge and shortened sludge stabilization period. Different studies have proposed the use of advanced oxidation (including ozonation) and other nonconventional pretreatments (such as sonication) with the aim of enhancing the biodegradability of the solid waste (Cheng and Hong, 2013; Chu et al., 2008; Carballa et al., 2007; Yan et al., 2009; Bougrier et al., 2006). Ozone, a strong oxidizing agent, has been proposed and used in different studies for enhanced degradation and the dewatering of sludge as reported by Michalska et al. (2012) and Mischopoulou et al. (2015). In their studies, the researchers attributed the enhancements they observed in the biogas production to the improvement of the anaerobic digestibility of the activated sludge.

Although anaerobic digestion is a widely used technology, a fundamental gap in knowledge still exists regarding the response of the AD system and its embedded microbial communities to mixed agricultural waste. Despite recent reports on co-digestion the optimal mixture and operating conditions that will maximize biogas yield without compromising the stability of the process are still unknown. In addition and to the best of our knowledge no published work has yet explored the use of ozone in enhancing biogas production from different types of solid waste, particularly under co-digestion conditions. The only available reference described the use of ultrasound and ozone for the treatment of molasses wastewater (Mischopoulou et al., 2015). Michalska et al. (2012) reported the use of Fenton reagents for the enhancement of biogas production from lignocellulose materials. The present work thus aims to evaluate the use of dung and agricultural waste as a mixed substrate (co-digestion) for the production of methane via AD. In addition, this investigation aims to explore the impact of using ozone as an intermediate chemical oxidation step between a two-stage AD on the performance of AD system and methane production. The optimization, enhancement, and effect of key operational parameters on the mesophilic co-digestion will also be evaluated in batch and continuous AD systems. The resulting improvements in dewaterability and settling ability of the digested substrate were investigated in order to determine its potential use as a soil fertilizer.

## 2. Material and methods

### 2.1. Substrate feedstock and inoculum

The substrate used in this study consisted of agricultural waste containing mainly wheat straw, cow and sheep dung, and wood dust mixed with wastewater. All materials were sourced locally from the southern part of Jordan (Karak Municipality Region; 31° 11' 5''N 35° 47' 17/45''E). The wastewater used in the substrate preparation was a secondary wastewater effluent collected from

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