

# Effect of particle size and doses of olivine addition on carbon dioxide sequestration during anaerobic digestion of sewage sludge at ambient and mesophilic temperatures



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## ABSTRACT

Biogas from anaerobic digestion of sludge at wastewater treatment plants consists of methane, carbon dioxide and trace contaminants which can be upgraded for utilization. Compared to current costly upgrading technologies, mineral carbonation has many benefits by using natural magnesium and calcium rich ores capable of sequestering CO<sub>2</sub>. The feasibility of olivine to sequester CO<sub>2</sub> in-situ during batch anaerobic digestion of sludge was tested for (1) ambient versus mesophilic temperatures, (2) placement of olivine in the digester, and (3) olivine particle size and concentration. Increasing the temperature, increasing the olivine surface area via increased dose and decreased particle size, and elevating the olivine in the reactor increased mineral carbonation rates during anaerobic digestion. At mesophilic temperature, the elevated 5% w/v fine olivine digester had a 17.5% reduction in CO<sub>2</sub> which equated to a 3.6% increase in methane content (%) and at ambient temperature, the same condition had a 21.7% CO<sub>2</sub> sequestration resulting in an 8.8% increase in methane content compared to the control. Response surface methodology was applied for optimization of digestion time and olivine surface area at both temperatures.

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## 1. Introduction

Anaerobic digestion (AD) is one of the most efficient and widely used technologies for the treatment of sludge from wastewater treatment plants (WWTPs) [1]. In the US, WWTPs produce approximately 6.5 million tons (dry weight) of sludge annually [2]. The total energy produced from WWTP sludge can potentially displace 441 million gallons of gasoline equivalent per year [3]. AD technology offers numerous significant advantages, such as low energy requirements [1], a reduction in pathogens and odors, and a reduction in the total solids, termed biosolids, quantity by converting part of the volatile solids (VS) fraction to biogas [4]. Biogas produced from AD of sludge is primarily composed of 50–70% methane (CH<sub>4</sub>) and 30–50% carbon dioxide (CO<sub>2</sub>), with smaller amounts of hydrogen sulfide (H<sub>2</sub>S), ammonia (NH<sub>3</sub>) and nitrogen (N<sub>2</sub>) [3,5]. However, biogas utilization requires cleanup and upgrading processes for removal of CO<sub>2</sub> and other contaminants.

The upgraded biogas can be used for the production of heat and power and/or co-generation, vehicle fuel and chemicals and injection

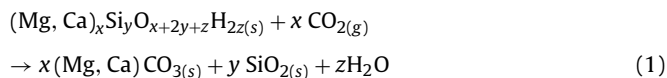
into the natural gas grid [3,5]. Only 48% of the total wastewater flow in the US is treated with AD [6] and less than 10% of WWTPs implementing AD technology utilize biogas for heat and power generation [7]. In July 2014, the US Environmental Protection Agency (USEPA) qualified biogas from landfills and anaerobic digesters as a cellulosic transportation biofuel under the new Renewable Fuel Standards (RFS2) where the biogas can generate D3 Renewable Identification Numbers (RINs) [8]. This mandate creates opportunity for US WWTPs to produce biogas as an economically-viable energy source.

The current commercial technologies for biogas upgrading and clean-up increases biogas production costs by 20–72% due to energy demand, chemical and water requirements, and causes up to 8% methane loss [9,10]. Also, CO<sub>2</sub> is usually released into the atmosphere during regeneration of the adsorbent media used in the upgrading process [5]. Mineral carbonation is an attractive alternative for the removal of CO<sub>2</sub> from biogas at WWTPs because it involves storing the CO<sub>2</sub> as stable carbonate precipitate [11–13]. Mineral carbonation, similar to a natural weathering process, utilizes calcium- and/or magnesium-rich natural ores such as wollastonite (CaSiO<sub>3</sub>), olivine (Mg<sub>2</sub>SiO<sub>4</sub>), forsterite (Mg<sub>1.82</sub>Fe<sub>0.18</sub>SiO<sub>4</sub>), and serpentine (Mg<sub>3</sub>Si<sub>2</sub>O<sub>5</sub>(OH)<sub>4</sub>) to react with CO<sub>2</sub> using the fol-

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lowing overall carbonation reaction:



[11,14]. This reaction is dependent on: (1) the rate of  $\text{CO}_2$  absorption in water in accordance with Henry's law which is responsible for carbonic acid formation ( $\text{H}_2\text{CO}_3$ ) leading to a solution pH decrease, and (2) the slow release of the silicate minerals which consumes protons and releases cations, resulting in increased pH and alkalinity leading to the precipitation of solid carbonates [11,12,15]. The mineral carbonation reaction is favored at a basic pH due to the availability of carbonate ions,  $\text{p}K_a$   $10^{-10.3}$  [11]. Conversely, the rate of cations release from the silicate material has been found to increase at acidic pH ranging from a pH of 3–6 [12,16]. The dissolution of cations also increases with increasing operating temperature and pressure, and surface area of the silicate material [11].

Research evaluating the kinetics of the carbonation reaction of aqueous  $\text{CO}_2$  with minerals include olivine [17], wollastonite [15], serpentine [18] and forsterite [16]. Other research has investigated the effect of natural mineral addition as a microorganism immobilizer [19] and the effect of AD on dissolution kinetics [20], for reducing inhibition of ammonium rich organic sludge [21], and swine manure [22,23] or  $\text{CO}_2$  scavenging using glucose [24]. Zeolite (chemical composition 58%  $\text{SiO}_2$ , 11.94%  $\text{Al}_2\text{O}_3$ , 5.94%  $\text{CaO}$ , 4.36%  $\text{Fe}_2\text{O}_3$  and other trace components) addition to swine manure AD increased biogas production by 20%, increased methane yield by 11%, and decreased batch AD of manure time by 6 days, which was attributed to the increase in Ca/Mg ions released from the zeolite [22]. Moreover, in-situ mineral carbonation during AD will merit additional fertilizer credit of the digestate as higher content of Mg-, Ca-, or Fe-carbonate precipitates in the stabilized residues [20].

Greater than 60% of the estimated costs for mineral carbonation are associated with grinding of the silicate minerals to smaller particles [12]. Therefore, determining the optimal particle size is important for not only the process performance but also the process economics. Additional costs are associated with integration of the carbonation material into the process such as extraction of  $\text{SiO}_2$  and other inert materials from the digester requiring new digester configuration [12]. The response surface methodology (RSM) is one of the most applied methods for evaluation and optimization of multiple variables effecting the studied parameters such those reviewed by researchers dealing with various types of digestion [25–28]. Olivine was selected for these experiments because it is a magnesium-rich ore of particular interest to geochemists for natural weathering [29]. It is structured in such a way that it has a weak resistance to weathering, has a significant amount of divalent cations and thus susceptible to undergoing carbonation reactions [17].

The main objective of this study is to enhance AD of sewage sludge by in-situ  $\text{CO}_2$  sequestration using a modified version of Argonne's patented process (enhanced methane production via  $\text{CO}_2$  sequestration, US 8,247,009). This study will evaluate olivine as an efficient additive for in-situ  $\text{CO}_2$  sequestration by comparing the results of (1) ambient and mesophilic temperature AD, (2) the placement of the olivine in the bottom of the anaerobic digester vessel versus elevating the olivine in a bag/basket system, and (3) effect of total surface area based on varying olivine concentration and particle size. RSM was applied as a method for optimization of the total surface area and digestion time for both the ambient and mesophilic temperature experiment.

## 2. Materials and methods

### 2.1. Sludge samples

The sludge samples were obtained from Stickney Water Reclamation Plant (WRP) of the Metropolitan Water Reclamation District of Greater Chicago (MWRDGC) located in Stickney, IL. For the purpose of this study, 'raw sludge' refers to the sludge obtained prior to the inlet of the anaerobic digesters and 'digested sludge' refers to the sludge obtained from the effluent of the anaerobic digesters located at Stickney WRP. The raw sludge was used as the feedstock (substrate) and the digested sludge was used as the inoculum for the experiments (Table 1).

### 2.2. Experimental design and set-up

The three digester configurations used in these experiments were termed the Digester System (Fig. 1A), Challenge Technology MPA-200 Biomethane Potential Analyzer system (Springdale, Arkansas) called Challenge System for short (Fig. 1B), and Incubator System (Fig. 1C). Table 2 shows the operating conditions for the three systems. The Digester System was custom built with a working volume of 400 mL. This system operates at ambient temperature with intermediate fluid recirculation for mixing. The volume of gas produced for the Digester System was measured daily utilizing a gas collection system (Fig. 1A). During gas volume measurement the gas collection burettes are allowed to reach equilibrium with the atmosphere one at a time by moving the 1 L leveling bottle up or down until the liquid level is the same as in the burette. The volume of the gas collection solution is then recorded and the final gas volume is determined by the daily displacement of the gas collection solution. The gas collection solution contains a buffer solution with 5 wt% sulfuric acid and 15 wt% sodium sulfate in water ( $\text{pH} < 1.0$ ) per Method 2720B [30] to minimize gas solubilization into the liquid. Sulfuric acid limits carbon dioxide solubility and sodium sulfate limits solubility of methane and other gases. A time series experiment was conducted to determine the impact of this acid displacement solution on biogas quality (See Supplemental File 1 for more details). Results showed that  $\text{CH}_4/\text{CO}_2$  ratio in the standard biogas samples reduced from 65% to 53.5% in the buffer solution. This  $\text{CO}_2$  loss is considered in reported methane and carbon dioxide concentrations [31]. The percent methane was normalized to assume 100% biogas (methane and carbon dioxide only) was produced. Both Challenge System and Incubator System are identical continuously stirred digesters carried out in 650 mL Wheaton bottles. After the first experiment, the working volume was increased from 400 mL to 550 mL in an attempt to decrease the amount of headspace available for medium evaporation. The Challenge System consists of an eight-position water bath providing temperature control and agitation, an eight-channel respirometry-based unit for gas measurement, and a computer with pre-installed software for automated gas data recording. Unfortunately gas volume measurement could not be performed on the Incubator System digesters. Therefore for the second experiment utilizing the Incubator system, one of the replicate digesters was placed in the Challenge System and the remaining 1–2 digesters were placed in the Incubator System. The data collected from these two systems are averaged for the same condition resulting in duplicate/triplicate results for all data except for the gas production.

The measured gas volume was adjusted to ambient temperature and pressure (20 °C and 1 atm) each time in order to account for fluctuations in the lab [31]. For the Digester System, the gas was assumed to be dry since it was produced and measured at ambient temperature. For the Challenge System, the gas was produced at 37 °C and measured at ambient temperature which caused water

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