INNOVATIVE BIOGEOCHEMICAL SOIL COVER TO MITIGATE LANDFILL GAS EMISSIONS

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Abstract

The municipal solid waste (MSW) in landfills undergoes anaerobic decomposition to produce landfill gas (LFG), which predominantly consists of methane (CH_4) and carbon dioxide (CO_2). Fugitive LFG emissions which are otherwise not targeted by gas collection system escape into the atmosphere, forming one of the largest anthropogenic sources of CH_4 and CO_2 emissions in the United States. The landfill cover soil plays an important role in mitigating the LFG emissions by microbial oxidation of CH_4 to CO_2 thereby reducing the CH_4 emissions to atmosphere. Several researchers have investigated the addition of organic amendments to the cover soils in order to enhance microbial oxidation of CH_4 in landfill covers. In recent years, biochar as an organic amendment has shown promise in enhanced microbial oxidation due to its inert/stable chemical nature to degradation, high surface area, high internal porosity, and high moisture holding capacity. However, in all these efforts there is no regard given to the CO_2 that still escapes into the atmosphere in undesirable amounts. Steel slag, a product from steel making industry, due to its high alkaline buffering capacity, high carbonation potential, and its unique cementitious properties has found numerous applications in civil and environmental engineering. But, until now there has been no study on the potential use of steel slag in landfill covers to sequester the CO_2 emissions. Ongoing research study, funded by the U.S. National Science Foundation, explores the use of BOF steel slag in conjunction with biochar amended cover soil so as to first convert CH_4 to CO_2 by microbial oxidation and thereafter sequester the resulting CO_2 from CH_4 oxidation and the prevailing CO_2 from anaerobic decomposition together by steel slag, thereby significantly mitigating the LFG emissions from landfills. In this paper, a review on the current applications and carbon sequestration mechanisms of BOF steel slag is presented. Finally, the proposed concept of the biogeochemical soil cover for mitigation of LFG emissions and some of the results from a preliminary investigation indicating the CO_2 sequestration potential by steel slag are discussed.

Keywords: MSW landfills, landfill cover; landfill gas; bio-geochemistry; BOF steel slag; biochar; carbonation

1. INTRODUCTION

The global greenhouse gas (GHG) emissions including the methane (CH_4) and carbon dioxide (CO_2) have been rapidly increasing due to population growth and the corresponding energy and resource consumption across the globe. This has contributed significantly to global climate change. Although landfilling of waste is considered an unsustainable option, it still remains the primary waste management technique in the US and many other countries. According to United States Environmental Protection Agency (USEPA) out of the 258 million metric tons of MSW generated in 2014, about 136 million metric tons of MSW was landfilled (USEPA 2016). The MSW placed in landfills undergoes anaerobic decomposition producing the landfill gas (LFG) which predominantly consists of CH_4 and CO_2. MSW landfills are regarded one of the largest anthropogenic sources of CH_4 and CO_2 emissions into the atmosphere in U.S. In this regard, the U.S. regulations mandate the installation of active gas extraction systems to substantially minimize the emission of LFG and other non-methane organic compounds into the atmosphere. However, fugitive emissions persist as they are otherwise not targeted by the gas extraction systems.

Several researchers have demonstrated the limited oxidation of CH_4 into CO_2 in the soil naturally due to methanotrophic bacteria in the landfill cover soils (Whalen et al. 1990; Hilger et al. 2000). In order to enhance the microbial oxidation of CH_4 in landfill cover, biocovers were introduced that involved amending the cover soil with organic material to promote the microbial growth. Several organic amendments such as compost, manure, biosolids, and digested sludge which could be used to enhance microbial activity, had several limitations. Compost or sewage sludge, if it is not substantially degraded, can undergo anaerobic decomposition producing CH_4 and CO_2 itself, thus exacerbating emissions.

Recent lab-scale investigations and field pilot tests have demonstrated that biochar shows good performance as an organic amendment for enhanced microbial oxidation of CH_4 in landfill covers (Yargicoglu and Reddy, 2017a, b, 2018). Biochar is a solid byproduct derived from gasification or pyrolysis of biomass under anoxic or low oxygen conditions (Reddy et al., 2014). Biochar with its high porosity and large specific surface provides favorable environment for the methanotrophs to proliferate and thrive in the cover soil thereby facilitating the CH_4 oxidation (Yargicoglu et al., 2015). Although the biochar-amended soil cover system mitigates the CH_4 emissions into the atmosphere, it can only oxidize CH_4 into CO_2, leading to continued emission of CO_2 into the atmosphere in undesirable amounts. Hence, it is desirable to develop a cover system that can minimize both the CH_4 and CO_2 emissions into the atmosphere.

Mineral sequestration of CO_2 by carbonation using steel slag is proposed to be an effective approach to substantially alleviate CO_2 emissions into the atmosphere (Huijgen et al., 2005, 2006). Steel slag is a product generated during the steel making process. There are several studies in the literature that have investigated the potential use of steel slag for CO_2 sequestration. Most of the studies focused on CO_2 sequestration for slag pre-treatment (for pH reduction) in civil engineering applications such as its use as aggregates in concrete, base layer in roadways and pavements, railroad ballasts thereby preserving the virgin materials. There are a few studies that investigated the utilization of steel slag in landfill covers as hydraulic barrier material. However, there has been no study on the use of steel slag in landfill cover systems specifically for CO_2 sequestration. This paper discusses steel slag and its unique characteristics that aid in CO_2 sequestration. Thereafter, the geochemistry of mineral sequestration of CO_2 by carbonation is presented. A review of typical applications of steel slag leveraging the process of carbonation is also presented. Finally, the paper proposes the concept of zero emission from landfills to investigate the use of biochar and steel slag amendments to soil in the landfill cover system to sequentially mitigate both CH_4 and CO_2 emissions from landfills.

2. STEEL SLAG

Steel is one of the most used materials on earth. The global annual steel production in the year 2016 reached 1.63 billion metric tons and 78.5 million metric tons of which was produced in U.S. (World Steel Association, 2017). Steel slag is a product obtained from the iron and steel making process. It originates as a molten liquid melt while impurities are being separated from molten steel and is a complex solution of silicates and oxides that solidifies upon cooling. The amount of steel slag produced depends on both the feed and the type of furnace used, but typically 0.2 ton of steel slag is produced per ton of steel. The steel making industries in U.S. generate approximately 10-15 million tons of steel slag every year (Yildirim and Prezzi, 2011).

There are three principal types of slag based on the type of furnace used for steel production namely, (i) Basic Oxygen Furnace (BOF) Slag, (ii) Electric Arc Furnace (EAF) Slag, and (iii) Ladle Furnace

(LF) Slag. A detailed explanation of the production process in each of the furnaces and the typical chemical composition of different slag types can be found in Shi (2004), Pan et al. (2016) and National Slag Association, (2013). The composition of furnace charges, grades of steel produced, rate of slag cooling and furnace operating practices also influence the composition and properties of steel slag. The typical composition of steel slag mainly comprises of calcium silicates, calcium aluminoferrites, and fused oxides of calcium, iron, magnesium, manganese and trace heavy metals. There also exists approximately 10 to 15% free lime in steel slag based on the operating conditions employed for steel making and the steel making process. The pH of the fresh steel slag is usually similar to lime products or up to 12.5.

The steel slag generated at the steel plants is initially stockpiled at the plant and eventually sent to slag disposal sites, if there is no use or market for it. In this regard, several researchers have studied the use of steel slag in order to reduce its disposal in landfills and also preserve the natural resources. Steel slag exhibits some unique characteristics that make it suitable for its use in many civil and environmental applications. Moist or wet steel slag has a high affinity for atmospheric CO_2. The lime, Portlandite and several Ca silicates in steel slag undergo dissolution in the presence of moisture to react with CO_2 and precipitate as carbonates. This results in increased physical stability, strength and compressibility characteristics due to the binding nature of the carbonate precipitates. A detailed explanation on the carbonation mechanisms and some of the important factors influencing the process is discussed in the following section.

3. CARBONATION IN STEEL SLAG

The fundamental mechanism behind the carbonation of steel slag is to allow for the reaction between cations (Ca^{2+}, Mg^{2+}) in the presence of moisture and CO_2 to form thermodynamically stable and insoluble carbonates. BOF steel slags are ideal minerals for carbon sequestration as they are low-cost, with high CaO content (Pan et al. 2016). Various studies have demonstrated that the accelerated carbonation of steel slag is an effective technique to stabilize the alkaline mineral while simultaneously fixing the CO_2 (Pan et al. 2016; Olajire 2013).

Carbonation of alkaline solid wastes (e.g. steel slag) may occur in two ways: (a) direct carbonation (one step process), and (b) indirect carbonation (two or more step process). Moreover, the direct carbonation could be classified as dry or aqueous based on the liquid-to-solid ratio. The direct reaction of gaseous CO_2 with solid mineral or alkaline waste is the most straight forward mineral carbonation route. However, the reaction rate of dry carbonation is very slow and carbonate conversion is low. The dry carbonation of Ca silicate minerals can be generalized as in Eq. 1.

$$\text{Ca-silicate}_{(s)} + CO_{2(g)} \rightarrow CaCO_{3(s)} + SiO_{2(s)} \tag{1}$$

Aqueous carbonation is faster and results in higher carbonate formation than dry carbonation. Direct aqueous carbonation involves three coexistent mechanisms based on the minerals available. First, dissolution of lime and portlandite takes place to readily produce Ca^{2+} ions as shown in Eq. 2 and 3. Then, carbon dioxide dissolves in water resulting in an acidic environment and CO_3^{2-} ions species as shown in Eq. 4.

$$CaO+H_2O \rightarrow Ca(OH)_2 \tag{2}$$

$$Ca(OH)_2 \rightarrow Ca^{+} + 2OH^{-} \tag{3}$$

$$CO_{2(g)} + H_2O_{(l)} \rightarrow H_2CO_{3(aq)} \rightarrow 2H^{+}_{(aq)} + CO_3{}^{2-}{}_{(aq)} \tag{4}$$

Ca^{2+} leaches from the mineral matrix (larnite), facilitated by the protons present as shown in Eq. 5.

$$\text{(Ca)-silicates}_{(s)} + 2H^{+}_{(aq)} \rightarrow Ca^{2+}_{(aq)} + SiO_{2(s)} + H_2O_{(l)} \tag{5}$$

Finally, calcium carbonate precipitates as shown in Eq. 6

$$\text{(Ca)-silicates}_{(s)} + CO_3^{2-}{}_{(aq)} \rightarrow CaCO_{3(s)} \quad (6)$$

Hence, the fundamental mechanism of direct carbonation reaction in steel slag involves (1) the leaching of alkaline mineral metal from the mineral matrix (predominantly Ca), (2) dissolution of gaseous CO_2 into aqueous phase followed by conversion of carbonic acid to carbonate ions, and (3) consequent precipitation of stable carbonates from Ca silicates (Pan et al. 2016). A schematic of the direct carbonation is shown in Fig. 1.

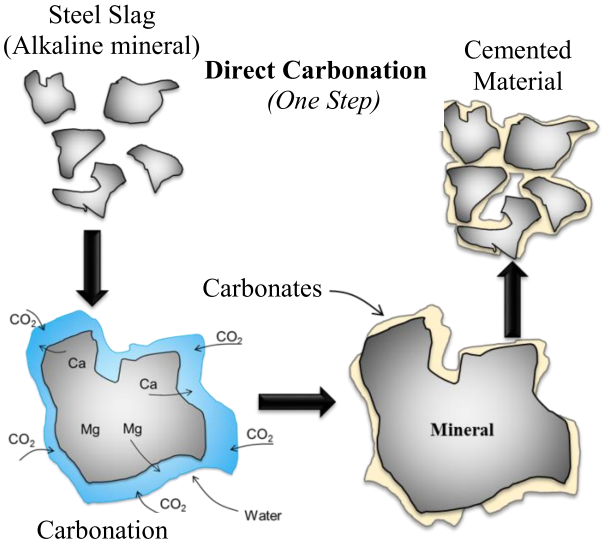

Figure 1: Direct Carbonation

Indirect carbonation is the process where reactive alkaline earth metal is first extracted from the steel slag using chemical extractants (e.g. acetic acid) and subsequently carbonated which is given by the chemical reactions in equations 7 and 8 below. This method is usually employed in commercial manufacturing of carbonates.

$$CaSiO_{3(s)} + 2\,CH_3COOH_{(aq)} \rightarrow Ca^{2+}{}_{(aq)} + 2\,CH_3COO^-{}_{(aq)} + SiO_{2(s)} + H_2O_{(aq)} \quad (7)$$

$$Ca^{2+}{}_{(aq)} + 2\,CH_3COO^-{}_{(aq)} + CO_{2\,(g)} + H_2O_{(aq)} \rightarrow CaCO_{3\,(s)} + 2\,CH_3COOH_{(aq)} \quad (8)$$

The mechanism of carbonation in steel slag can vary based on the composition of steel slag. A detailed explanation on each of the carbonation approaches is presented by Pan et al. (2016) and Olajire (2013). In this study, the concept that is of relevance is the direct carbonation of steel slag to realize CO_2 sequestration by forming stable and insoluble carbonates. The various advantages/benefits of carbonation of steel slag and its application in civil and environmental engineering is discussed briefly in the following section.

4. BENEFICIAL USES AND APPLICATIONS OF STEEL SLAG

BOF steel slag has been predominantly used as a construction material in asphalt paving, unpaved roads, agricultural lime, acid mine drainage remediation, manufacture of cement all of which can be ascribed towards its enhanced mechanical and pozzolanic properties resulting from carbonation. In addition to its use as a construction material, several researchers assessed the use of steel slag for environmental remediation applications, especially phosphorous removal (e.g., Drizo et al., 2006). BOF steel slag has been used as base and subbase layers in pavements in addition to its use as coarse aggregates in surface layers of pavements, mainly due to its high strength, high binding capacity with high abrasion and high frictional resistance. Due to their high density, strength, rough texture and durability, they can be processed as high-quality aggregates, comparable with natural aggregates.

Relevant to this study is the work of Herrmann et al. (2010), who investigated the potential use of EAF slag and Ladle slag (LS) as a barrier material in landfill cover. Surprisingly, these slags possessed very low hydraulic conductivity. Field investigation using ten lysimeters confirmed that the leachate collected at the base of the cover is well below the regulatory criterion and can potentially be used as a barrier material in landfill covers.

Diener et al. (2010) studied the stability of steel slag (mixture of EAF and LS) by performing long term laboratory tests to understand the leaching behavior, acid neutralization capacity and mineralogy of steel slag using multivariate data analysis. The researchers investigated the effect of CO_2 content in the atmosphere, relative humidity, aging time, temperature and water quality on the accelerated aging in steel slag. It was observed that the aging time and the CO_2 content in the atmosphere were the prime factors that had a significant effect on long term stability of steel slag. Mineralogical changes during the aging process showed the formation of calcite which was confirmed using XRD. A review on several other applications of steel slag can be found in Pan et al. (2016), Olajire (2013) and Yi et al. (2012).

In all of the literature on applications of steel slag, it has been used as a construction material or as a hydraulic barrier material in the final cover in landfills. However, to date, there has been no study on the use of steel slag for the sequestration of CO_2 emissions from the MSW landfills. This study proposes to utilize the carbonation and other mechanisms associated with CO_2 sequestration in steel slag and thereby mitigate the CO_2 emissions released into the atmosphere from the landfills. A new concept of “Zero Emissions Cover System” is proposed as detailed in the following section.

5. BIOGEOCHEMICAL SOIL COVER SYSTEM

There are several studies in the literature that have looked at the potential use of steel slag in carbon sequestration using the carbonation process. However, there has been no investigation on the use of steel slag as a cover soil amendment in landfills to sequester CO_2 emissions. A new research project funded by the U.S. National Science Foundation is currently in progress to fully explore the fundamentals and practical aspects of an innovative, low-cost landfill cover system consisting of steel slag in combination with biochar which can mitigate the CH_4 and CO_2 emissions from MSW landfills. In a previous investigation at UIC, biochar-amended soil cover was conceived and developed as an effective sustainable biocover for CH_4 oxidation. A systematic and comprehensive study was performed involving material characterization, batch tests, and lab-scale column experiments to investigate the adsorption and transport mechanisms associated with biochar amended cover soil systems. Microbial characterization was also performed to identify the specific methanotrophs that were involved in CH_4 oxidation. A full-scale field demonstration was also implemented to evaluate the performance of biochar-amended soil cover at a landfill exposed to variable environmental conditions. This research work advanced the knowledge on the fundamental processes and system variables responsible for enhancing the methanotrophic activity by providing a favorable environment for the methanotrophs to proliferate and thrive in the cover system. Unfortunately, such a biocover system can only microbially oxidize CH_4 into CO_2, leading to continued emission of CO_2 into the atmosphere.

It is essential to develop a cover system that can minimize both the CH_4 and CO_2 emissions into the atmosphere. It is hypothesized herein that biochar and steel slag amended soil cover systems can eliminate both CH_4 and CO_2 emissions via biochar-aided methanotrophic oxidation of CH_4 into CO_2 and subsequent or simultaneous slag-aided carbonation/sequestration of all of CO_2, thus ideally eliminating the LFG emissions from the landfills. This concept of “Zero Emissions Cover” is a transformative approach to substantially mitigate, if not eliminate, the carbon footprint associated with LFG emissions. The concept of biochar and steel slag amended soil cover system to achieve zero emissions is shown schematically in Fig.2. As shown in Fig.2, the proposed biogeochemical cover system also has the potential to mitigate hydrogen sulfide (H_2S) if present in the landfill gas, due to co-disposal of construction and demolition (C&D) waste with MSW.

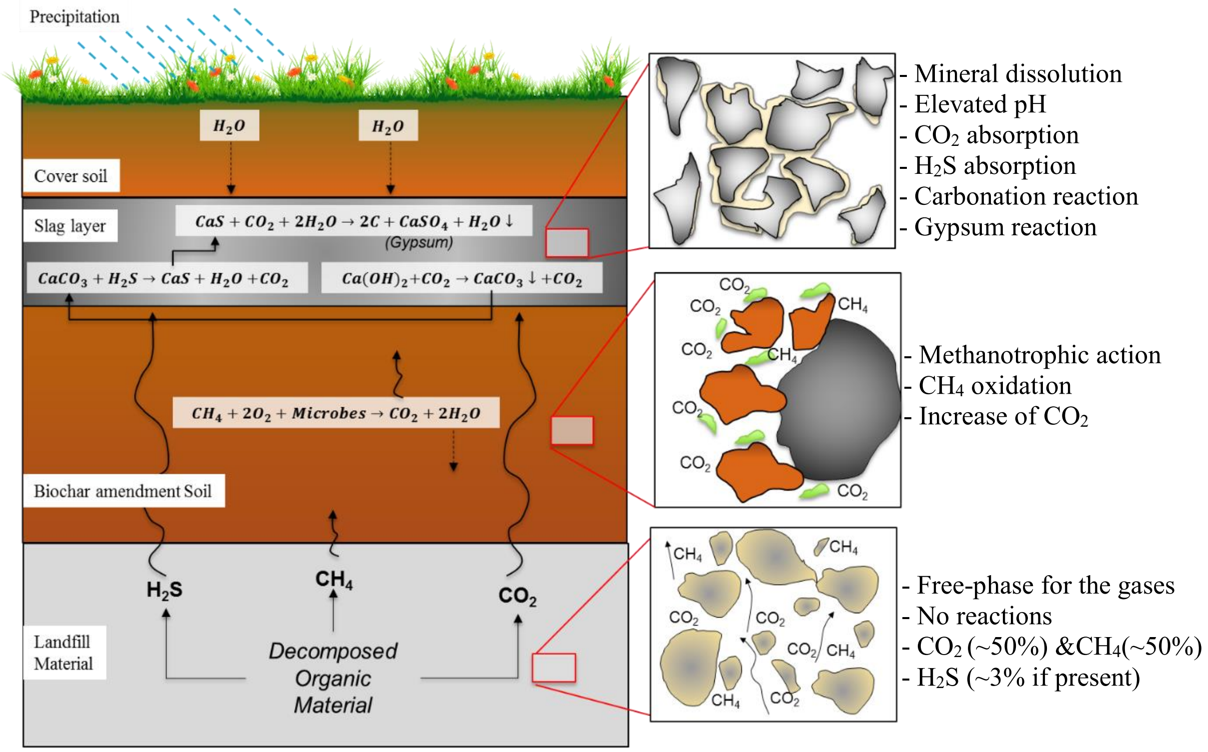

Figure 2: Concept of Biochar Steel Slag amended Soil Cover System for Zero Emissions

The combination of steel slag and biochar with their inherently distinct properties has several characteristics that are highly desirable in a landfill cover system. These include: (a) preferential adsorption of CH_4 by biochar thereby increasing its bioavailability for oxidation; (b) strong alkaline buffering capacity of steel slag which results in CO_2 absorption, and (c) increased physical stability, strength and compressibility characteristics due to the pozzolanic nature and angularity of steel slag. In addition, biochar and steel slag fines are very inexpensive, sustainable and practical to employ in field. Although there are numerous benefits identified with the use of these materials, it is of utmost importance to delineate the complex fundamental coupled biogeochemical processes that dictate the transport, adsorption, microbial oxidation, and carbonation of LFG. Moreover, it is important to assess the influence of various factors – including moisture, pH, LFG composition, loading rates, flow rates, particle size, steel slag and biochar source and composition, and several others – on the fundamental processes that occur in the proposed "zero emissions" cover system.

6. PRELIMINARY INVESTIGATION

In order to evaluate the potential for carbonation in steel slag, a preliminary experimental study was conducted at UIC to examine the ability of steel slag to sequester CO_2 at normal atmospheric conditions. A BOF steel slag sample was obtained from the Indiana Harbor East (IHE) steel plant, the cover soil from Zion Landfill, IL, and the biochar produced from gasification of wood pellets (which was used in a previous biochar study at UIC). The soil, biochar and steel slag were first characterized for their physical and geotechnical properties following relevant ASTM standards and the results are shown in Table 1.

A series of batch tests were conducted using these samples individually as well as in combinations of these materials in different proportions. The combinations of materials tested were: soil and biochar (S/BC); soil and slag (S/SL); slag and biochar (SL/BC); soil, biochar and 10% of slag by weight (S/BC/SL 10%); soil, biochar and 5% of slag by weight (S/BC/SL 5%); and soil, biochar and 2% of slag by weight (S/BC/SL 2%). The biochar content was 10% by weight in all of these combinations.

The samples were prepared in glass 125 ml vials at the same moisture content of 10% by weight and analyzed at room temperature. All the samples were tested in triplicate to ensure repeatability. The samples were injected with 12 ml of synthetic landfill gas mixture (50% CH_4 – 50% CO_2) and analyzed using a Gas Chromatograph (SRI 9300B) immediately after the injection of gas into the vial (Time 0) and after 24 hours from the injection (Time 24) according to the headspace sampling methodology. The preliminary investigation based on these batch tests showed complete depletion of CO_2 in all steel slag containing samples after 24 hours. Fig. 3 summarizes the results obtained the preliminary batch tests.

Table 1: Properties of Cover Soil, Biochar and Slag

Properties	Cover Soil	Biochar	Steel Slag
Organic Content (%)	3.10	63.7	0
Organic Carbon Content (%)	0.52	32.0	0
pH	5.3	8.7	12.3
Specific Gravity	2.65	0.81	3.04
Atterberg Limits:			
Liquid Limit (%)	31		
Plastic Limit (%)	19	Non-Plastic	Non-Plastic
Plasticity Index (%)	12		
Grain Size Distribution:			
Gravel (%)	0.0	67.8	15.78
Sand (%)	8.35	31.4	76.42
Fines (%)	91.6	0.0	7.8
Hydraulic Conductivity (cm/s)	$4.3x10^{-9}$	$1.2x10^{-2}$	$3.9x10^{-4}$

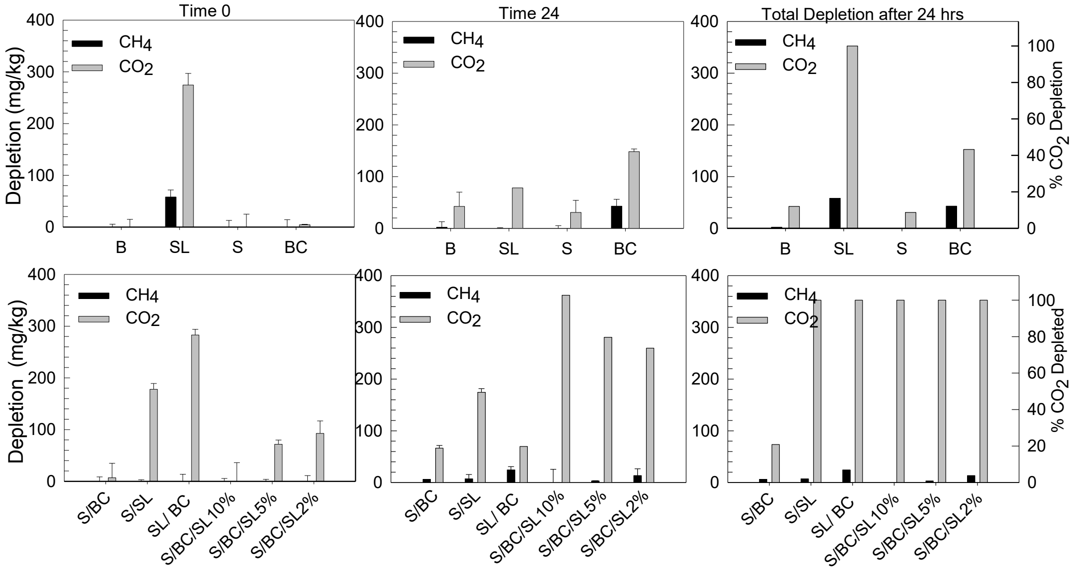

Figure 3: CO_2 and CH_4 Uptake in Samples with Soil (S), Biochar (BC) and Slag (SL) (B-Blank, S-Soil, BC-Biochar, SL-Steel Slag)

These preliminary results (SL/series results) clearly demonstrate that the CO_2 sequestration by slag can be rapid and substantial under normal landfill conditions. In addition, the CH_4 depletion in samples containing biochar and slag was observed (SL/BC data), indicating the removal of CH_4. However, the relative contributions of microbial oxidation or adsorption have not been established.

Therefore, there is still a need for extensive analysis of the carbonation potential of steel slag under various conditions including moisture, temperature, CO_2 loading, and particle size. It is also essential to investigate the effects of highly alkaline pH on the growth and activity of methanotrophs in cover soil and biochar. Some methanotrophs are found to be sensitive to pH, while others such as extremophile methanotrophs are shown to be resilient to high pH conditions (Trotsenko and Khmelenina, 2002; Saari et al., 2004; Roadcap et al., 2006). In addition, the individual mechanisms, reaction kinetics and interdependency of the microbial oxidation and carbonation must be quantified. All of these aspects are being investigated in our ongoing study.

7. ONGOING RESEARCH

It is anticipated that this innovative, low-cost, feasible and sustainable cover system will help in realizing the overarching goal of eliminating the LFG emissions from landfill into the atmosphere. In order to achieve this goal sequentially or simultaneously, it is essential to investigate the fundamental coupled biogeochemical processes involved in CO_2 sequestration by BOF steel slag and biochar/steel slag amended soils and optimize the cover system to achieve maximum CH_4 oxidation in biochar and at the same time maximize the CO_2 sequestration in steel slag. Fundamental questions that need to be addressed include: interactive geochemical changes due to the presence of slag and biochar together; survival and growth of methanotrophs; carbonation mechanisms and moisture and porewater composition impacts; clogging of pores and long-term permeability of the materials, among others.

In the ongoing research, we are focused on quantifying the physical, chemical and geotechnical characteristics of different types of slag including their leachability and surface characteristics. Thereafter, a detailed experimental investigation involving bench-scale testing and column experiments will be performed to identify the extent of carbonation in steel slag under varying environmental conditions including moisture, LFG composition, particle size, slag types among others. One of the important aspects of the conceptualized biochar-slag amended cover system is to determine the synergistic effects of having both biochar and steel slag in the cover system. It is essential to determine the extent of carbonation, microbial oxidation and the microbial activity in the cover system. Furthermore, long term column tests using large columns simulating typical landfill cover systems will be carried out to assess the simultaneous interactions of transport, adsorption, oxidation and carbonation for various simulated biochar, soil and steel slag cover systems under the influence of transient and dynamic changes in gas flow and environmental conditions. Microbial analysis of the selected tested samples will be to ensure there is adequate biological activity in the cover system for microbial oxidation of methane. Additional analysis involving the quantification of the extent of carbonation and the mineralogy of the carbonated samples will be performed using appropriate methods (SEM and XRD). Finally, based on the observations from the large-scale column experiments, few selected profiles will be tested for their performance at a landfill under realistic gas production and environmental conditions.

Acknowledgements

This project is funded by the U.S. National Science Foundation (CMMI # 1724773), which is gratefully acknowledged.

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International Confernece Protection and Restoration of the Environment XIV
Proceedings (MEMORY STICK)

ISBN: 978-960-99922-4-4