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Effect of Gypsum Waste Inclusion on Gasification of Municipal Solid Waste

Sustainable disposal techniques of municipal solid wastes (MSW) are essential for effective materials recovery and energy management. Synergistic incorporation of gypsum waste from the construction and demolition (C&D) sector is explored here for the low techno-economic viability of MSW in waste-to-energy facilities. Co-processing of MSW with this low-value gypsum can potentially provide improved product recovery and simultaneously furnish economic viability. To understand the impact of gypsum incorporation into MSW conversion, we tested synthetic MSW and a 1:1 mixture of MSW with gypsum from drywall wastes and compared their micro-scale mass loss kinetics along with macro-scale syngas evolution and conversion at different temperatures, and in pyrolytic and oxidizing environments. Gypsum incorporation led to increased syngas production and decreased char yields as the char was oxidized by CaSO₄. Thermogravimetric analysis (TGA) revealed two different temperature zones of gypsum interaction with MSW depending on the oxidation concentration in the environment. Adding 50% gypsum only changed the final ash yield by 10% in pyrolysis conditions suggesting the viability of gypsum incorporation. While the addition of gypsum led to delayed evolution of H₂ and CO, the cumulative yields of H₂ and CO₂ increased significantly and the yield of CO changed minimally. Additionally, hydrocarbon by-products such as CH₄ decreased by gypsum addition. The results showed improved syngas yield and uniformity, as well as operational conditions from the gypsum interaction with MSW which can help in the further development of gypsum waste incorporation. [DOI: 10.1115/1.4054825]

Keywords: combustion of waste/fluidized bed, energy conversion/systems, energy extraction of energy from its natural resource, energy from biomass, hydrogen energy, renewable energy

1 Introduction

The ever-increasing problem of municipal solid wastes (MSW) needs effective management towards achieving sustainable resource utilization. The world's population is steadily growing and with it, the waste generated is also increasing. In most countries, and especially in the United States, the amount of waste generated by the habitants is not matched by recycling and recovery efforts. This was apparent from the Environmental Protection Agency's statistics which showed that 50% of the 292 million US tons of MSW collected in 2018 was disposed in landfills while only 11.8% was processed for energy recovery and 23.6% recycled [1]. MSW components, in general, include wastes such as paper/cardboard wastes, glass, metals, plastics, rubber and textiles, wood, food waste, yard trimmings, and other miscellaneous wastes. Among all these elements, 34% of the metal components were recycled and only 11.5% ended in energy recovery facilities. Waste-to-energy (WtE) facilities such as incinerators offer alternatives to landfilling waste in the form of energy recovery. Even so, the effective value of the energy produced is low along with significantly higher costs compared to other ways of energy production. This leads to a debate on whether these energy recovery facilities are worthwhile. Furthermore, some wastes that contain valuable metals are often not separated and go through the highly oxidizing conditions of an incinerator, ending up in the ash residue in their high oxidation

states, thus making them difficult to recover. In addition, potential pollutants within the ash, such as Cl, Pb, Cd, and Hg must be stabilized to prevent their leaching into the environment before their next stage [2–4]. To recover these metals effectively, techniques such as oxygen staging can be used to control the spatial distribution of the oxidation potential (O₂ content) and mild reactor temperatures can be used to allow conversion of the carbonaceous content for energy and syngas recovery [5]. Thus, the metal content can be extracted from its residue at a lower oxidation state improving the economic value of the ash along with the recoverability of these metal atoms [6–9]. In these milder oxidation conditions, the carbonaceous materials thermodynamically favor char residue formation which lowers the energy and syngas output from the reactor.

To solve the problem of char formation, we incorporate the utilization of gypsum waste from the construction and demolition (C&D) sector to give them a new use by co-processing them with the MSW [10–14]. In 2018, 13.2 tons of gypsum waste was landfilled among the 15 tons generated by the construction and demolition sector [1]. This waste can be co-processed with the MSW stream in modified WtE facilities with gasifying conditions. In this way, the char produced will be reduced by the gypsum waste and this in turn can be converted into more valuable limestone/dolomite or quicklime residue for applications including construction, carbon sequestration, and flue gas cleaning [13,15–18]. In addition, the literature also revealed that gypsum addition can improve the ash behavior and avoid agglomeration and defluidization in biomass and MSW combustion [19–21].

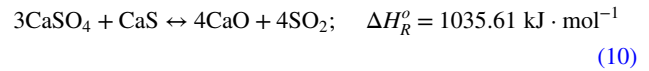
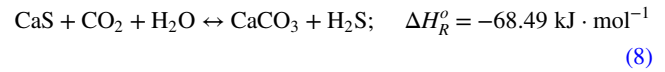
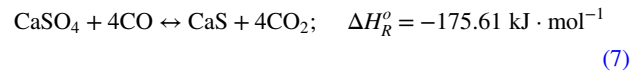
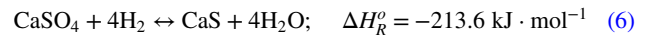
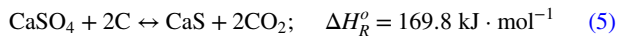
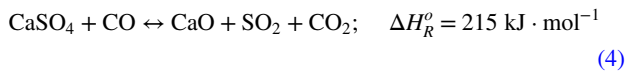
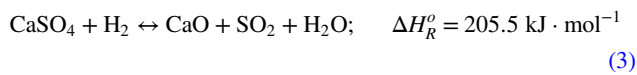
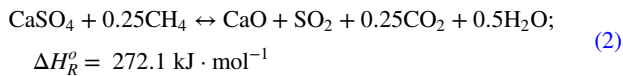
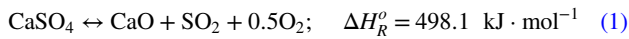
Utilizing these two wastes will contribute to sustainable and effective waste management in the residential and C&D sectors. Current literature on regeneration of CaO and SO₂ from CaSO₄ is limited to using reducing environments of CO and H₂ or mixing

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with carbon feedstocks such as coal and charcoal [16,22–25]. High energy and temperature needs (>1200 °C) make direct thermal decomposition of gypsum unattractive. Fuel-based reduction reactions include one-step pathways in Eqs. (2)–(4) and two-step pathways with Eqs. (5)–(7) showing the first step to form CaS and Eqs. (8)–(10) showing the second step where it is oxidized by CO₂/H₂O, O₂, or CaSO₄ [25]. Other processes such as gypsum carbonization under different conditions have also been investigated. Conditions such as hydrothermal, high pressure, or ultrasonic carbonization to create carbonates for carbon capture, and the reaction of these with additives such as NaOH, Fe oxides, and aluminosilicates to form cement and minerals for construction, medical applications, and catalytic materials were among the studies reported in the literature [13,15,18,26].

There is a significant gap in the literature on utilizing MSW-based carbon feedstock for gypsum conversion. This paper addresses such a gap using both micro-scale testing with thermogravimetric analysis (TGA) and macro-scale testing using a lab-scale experimental facility followed by a gas-chromatography analysis of the evolved syngas components from the MSW components as carbon sources and its mixture with gypsum from drywall. The various reactions involving the regeneration of CaO, CaCO₃, and SO₂ from CaSO₄ and CaS along with the respective heat of reactions are given below:



2 Methods and Materials

A synthetic MSW mixture was prepared in the lab to include various paper, plastic, and wood wastes to get a final composition as shown in Fig. 1. These mixtures were wet ground and dried to form a uniform mixture. During the pre-processing, some of the larger chunks were avoided to maintain good uniformity. For gypsum wastes, a commercial drywall board was obtained. This drywall board contained gypsum layered on both sides with cardboard. For our MSW-gypsum mixtures, the gypsum from the drywall board was separated from the cardboard content followed by grinding it with MSW to prepare MSW-gypsum mixture with a mass ratio of 1:1. The effect of gypsum on MSW conversion was investigated on micro-scale using thermogravimetric analysis and differential scanning calorimetry (TGA/DSC), and macro-scale examination using lab-scale fixed bed reactor equipped with evolved gas analysis.

TA Instruments SDT Q600 was utilized to carry out TGA of MSW, gypsum, and their 1:1 mixture to obtain the mass-loss behavior during their thermal treatment in different extents of oxidizing environments. The initial sample mass of each TGA test was 5–10 mg for all the cases to avoid transport limitations. Alumina pans were used as the sample holders wherein, the sample was first heated to 105 °C for 10 min for drying followed by heating

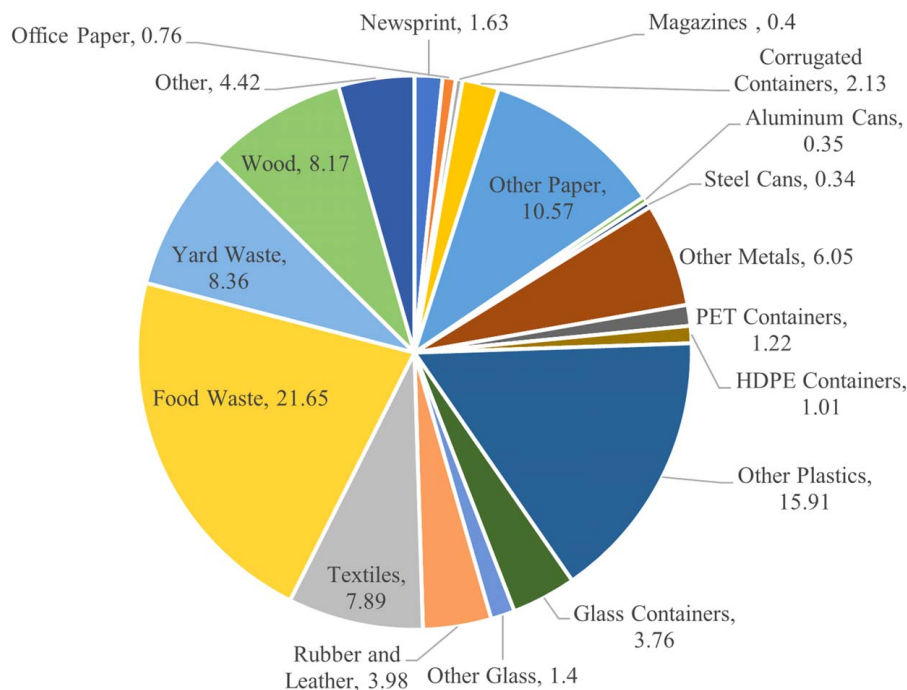


Fig. 1 Composition of synthetic MSW (in wt%)

to 1100 °C at 20 °C/min. Different oxidizing environments were tested by changing the purge gas. The purge gas concentration of O₂ was varied (0–21%) in Ar while maintaining the total purge gas flowrate to 100 sccm. Each test was conducted in triplets and their average value is reported in this paper.

Lab-scale fixed bed reactor was utilized to observe the impact of gypsum on macro-scale effects such as syngas products and their characteristics. This experimental setup contained two electrically heated furnaces set to isothermal operation temperature to heat the incoming gases and solid samples. The incoming gas was metered using orifice flow controllers. Once the flow and desired temperature were achieved, the solid sample contained in a quartz sample holder was introduced into the reactor via a quick-release coupling and measurement time started from this point. As the sample heats and decomposes, most of the gases were sent through ice-bath to obtain dry syngas. Most of this syngas was vented out while a small portion of it was allowed to pass through the gas sampling system. This contained a filter, peristaltic pump, and gas sampling bottles to collect syngas samples at different temperatures. After the first 5 min, the syngas analysis was carried out online using Agilent 3000 micro-GC equipped with thermal conductivity detector. In this study, the gas chromatograph (GC) was calibrated for H₂, O₂, N₂, CO, CO₂, CH₄, C₂H₂, C₂H₄, C₂H₆, and C₃H₈, and using N₂ as the trace gas, the flowrates of all the syngas components were quantified. Further details of this setup can be found in our earlier papers [27–31]. The tested temperatures were 800 °C and 900 °C and the feedstock samples were 10 g of MSW and mixtures of 10 g of MSW with 10 g of gypsum. To understand the effect of O₂ concentration, these tests were conducted in two different environments—pyrolysis (2.1 slpm of N₂) and oxidizing (2.1 slpm of O₂/N₂ mixture with O₂ concentration of 10%). Each test condition was repeated at least three times and their average results are reported in this paper.

3 Results and Discussion

3.1 Mass-Loss Behavior of Municipal Solid Wastes and Municipal Solid Wastes-Gypsum Mixtures. To understand the operating conditions of the reaction of gypsum with MSW mixture, we examined the mass-loss behavior of MSW and MSW-gypsum mixtures under different oxidizing environments. All the mass-loss data will be split by temperature into two zones of low temperature (400–850 K) and high temperature (850–1300 K) to better facilitate the role of gypsum addition effects. Figure 2 shows the rate of normalized mass-loss of (a) MSW and (b) MSW-gypsum mixture with temperature for different purge gas oxygen concentrations in low temperature zone conditions. From Fig. 2(a), the decomposition of the majority of MSW components

lies between 550 K and 700 K with a peak that depended on the oxidation availability. As the O₂ concentration increased, this peak moved towards lower temperature (from 640 K to 620 K) while the peak height also increased. This is expected as increasing oxygen availability leads to improved oxidation of the solid components and this consumption of the intermediates from the solids leads to improved mass-loss. Due to the heterogeneity of the MSW samples, even after grinding, certain sporadic peaks such as the peak at 680–700 K were observed due to certain minor components. This suggests that the decomposition involving MSW needs to be further studied focusing on major and minor peaks separately based on the probability of its appearance. Considering this variability, we focus only on the major peaks in this paper.

Now, comparing Figs. 2(a) and 2(b) reveals, the presence of significant peaks beyond 650 K when MSW was mixed with gypsum. But these peaks were not observed for all the cases. While the pyrolysis (0%) case showed a very small peak at 750 K, the addition of O₂ led to peaks at lower temperature but with some variability in their peak and location. The parameters contributing to such an intermittent behavior include not only the heterogeneity of MSW but also the difficulty in controlling the solid-phase contact between MSW and gypsum. It also reveals that while gypsum addition contributed to these additional peaks between 650–750 K, further studies will be needed to understand the components involved in these peaks and the products formed.

Further, Fig. 3 shows the effect of O₂ and gypsum in the high temperature decomposition zone. Here, only MSW-gypsum decomposition is shown since MSW decomposition showed no mass-loss in this temperature zone for all the different O₂ concentrations examined here. This reveals that a significant interaction between MSW and gypsum occurs in the high temperature zone during pyrolysis, but these peaks are apparent as O₂ is added. In the absence of O₂, as the pyrolysis of MSW leads to char formation in the solid-phase, this char reacts with CaSO₄ to form CaO and CaS depending on the equilibrium conditions, contact, and product gas partial pressures. Additionally, multiple peaks are apparent during pyrolysis—one at 900–1000 K and the other peaking at ~1180 K. These peaks are not present in the oxidizing environment, because here the char formation is minimized as it burns in the presence of O₂ leaving minimal carbon for CaSO₄ to react with at high temperature.

The TGA results also provided the impact of gypsum addition on the final ash residue for different O₂ concentrations, see Fig. 4. It reveals that two parameters affecting the ash residue—the gypsum and the O₂. At low O₂ concentrations, sufficient carbon from MSW was available to reduce gypsum and thus lower ash yield. But as the O₂ concentrations increased, O₂ became the predominant oxidizer. In MSW + gypsum case, high O₂ concentrations resulted in lowered ash residue due to complete combustion. So, in gypsum incorporated conversion of MSW, higher O₂

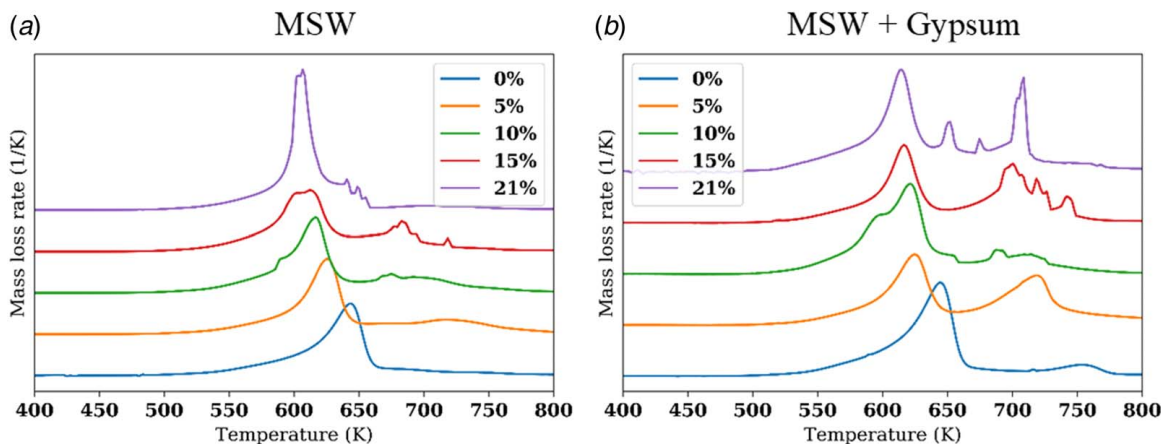


Fig. 2 Effect of O₂ concentration on the mass-loss rate of (a) MSW and (b) MSW-gypsum mixture with temperature (stacked view—baseline offset for comparison)

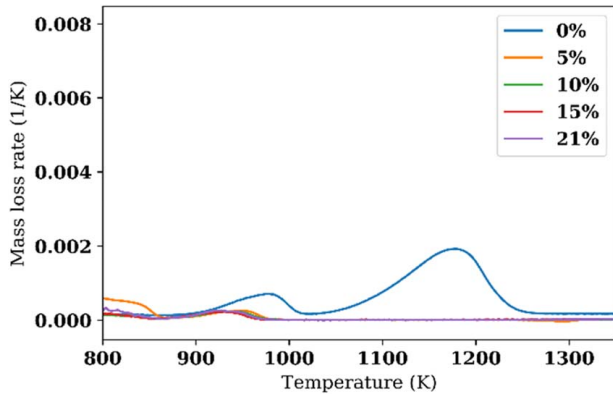


Fig. 3 Effect of O_2 concentration on the mass-loss rate of MSW-gypsum with temperature

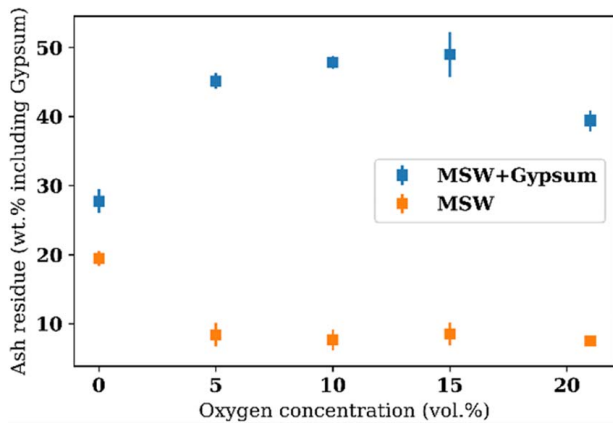


Fig. 4 Effect of gypsum addition and O_2 concentration on the MSW ash residue yield

concentrations can result in lower ash while in the pure MSW case, O_2 concentrations beyond 5% did not change the ash residue. While these results show the impact of gypsum addition on MSW incineration on a micro-scale, macro-scale studies were also carried out to reduce the heterogeneity effects of MSW sampling and allow for better sample representation.

3.2 Lab-Scale Fixed Bed Reactor Study of Gypsum Effects on Municipal Solid Wastes Conversion. MSW and

MSW-gypsum mixtures were tested in a macro-scale reactor for two isothermal cases at 800 °C and 900 °C in pyrolytic and oxidizing conditions. First, we examine the H_2 evolution from pyrolysis of these feedstocks. The source of H_2 production during pyrolysis includes hydrocarbon decomposition, cyclization, aromatization, and reforming reactions such as water gas reaction and water gas shift reaction. Figure 5 shows the impact of gypsum addition on MSW pyrolysis at (a) 800 °C and (b) 900 °C. The results reveal complete H_2 evolution in 20 min at 800 °C while it took only 15 min at 900 °C. An increase in temperature also increased H_2 flowrate peak by more than double due to the favored forward equilibrium of the above endothermic reactions. The addition of gypsum resulted in lowered and widened peaks of H_2 evolution. This could be from the heat-transfer limitations posed by gypsum addition resulting in the feedstock taking longer heating time. While the rate of H_2 decreased with gypsum addition, the cumulative yield increased by up to 20% at 900 °C. It suggests that although Eqs. (3) and (6) are possible for H_2 consumption by gypsum at these high temperatures, the downstream secondary reforming reactions reach equilibrium without gypsum interference which compensate for this consumption. This shows that gypsum addition can provide tunability in syngas generation from MSW without the need for changing operating conditions. We note that gypsum's impact on other syngas species also needs to be analyzed to understand its overall impact.

The impact of gypsum addition on CO evolution rate was more significant compared to H_2 evolution, see Fig. 6. Along with the delaying and widening of CO evolution peak, the cumulative yield was minimally affected by gypsum addition at 800 °C which showed a moderate decrease by 10%. Multiple reactions are possible for the reduction of CO by gypsum addition, especially in pyrolytic conditions. Equations (4) and (7) show the possible oxidation reactions of CO by gypsum. Such a delayed behavior of CO evolution could also be from excess initial pyrolytic CO reacting with gypsum via above reactions leading to CO_2 which reacts with char to form CO again. This formation of CO_2 can be seen clearly in Fig. 7. In addition to the lack of significant delay in CO_2 evolution, especially at 900 °C, addition of gypsum resulted in higher CO_2 evolution beyond the initial 5 min of devolatilization. According to the literature based on biomass and waste pyrolysis, CO_2 formation reactions such as decarboxylation from MSW decomposition occur even at moderate temperatures while H_2 and CO formation need the species to reach high temperatures. So, this clarifies the role of gypsum to result in delayed heating of MSW during the heating phase, followed by oxidizing reactions with the reducing species and carbon from the pyrolysis to form increased CO_2 . Further, Fig. 7 also shows CO_2 formation to continue with a slow rate even beyond 25 min. Based on MSW decomposition knowledge, this CO_2 is from the slow solid-phase reaction of gypsum with char residue limited by species contact, carbon

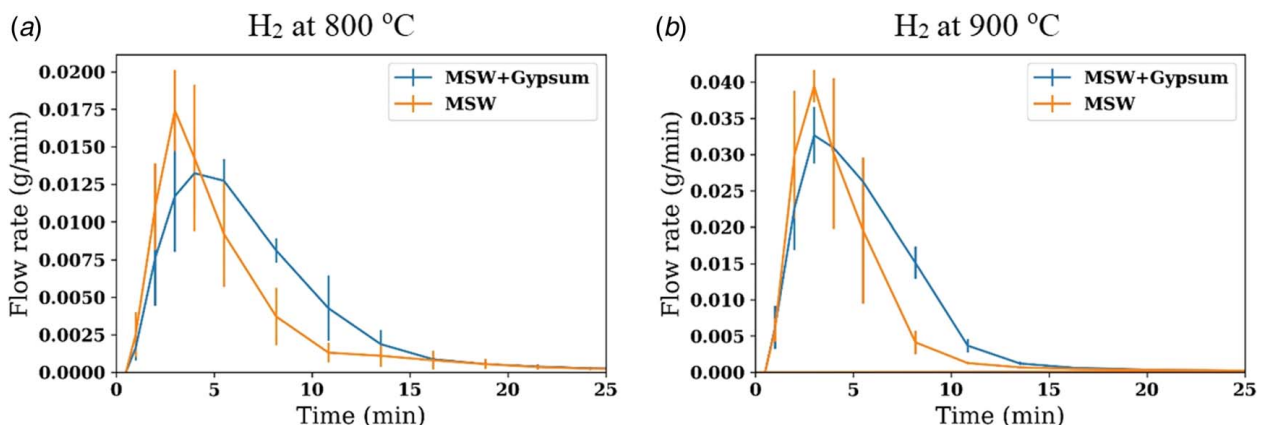


Fig. 5 Impact of gypsum addition to MSW on H_2 evolution with time during pyrolysis at (a) 800 °C and (b) 900 °C

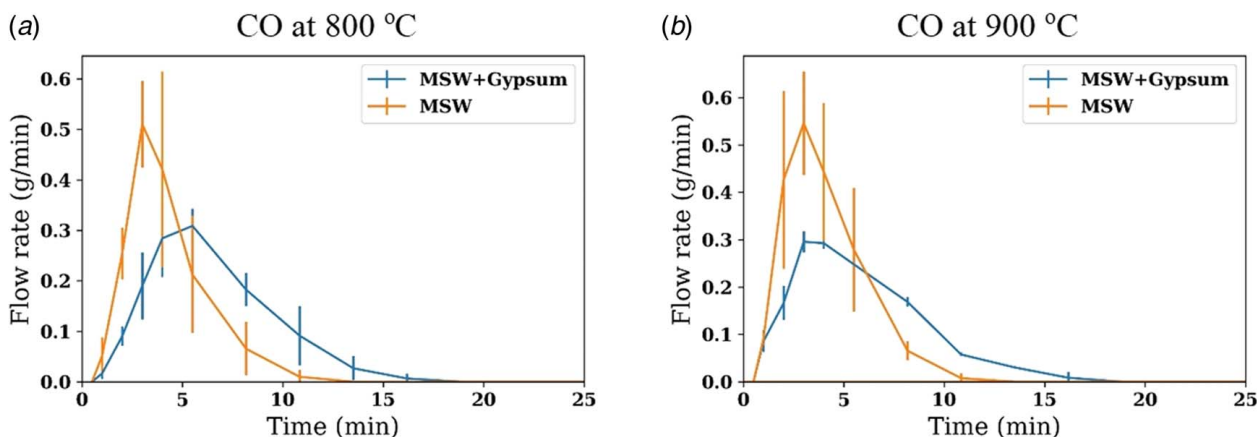


Fig. 6 Impact of gypsum addition to MSW on CO evolution with time during pyrolysis at (a) 800 °C and (b) 900 °C

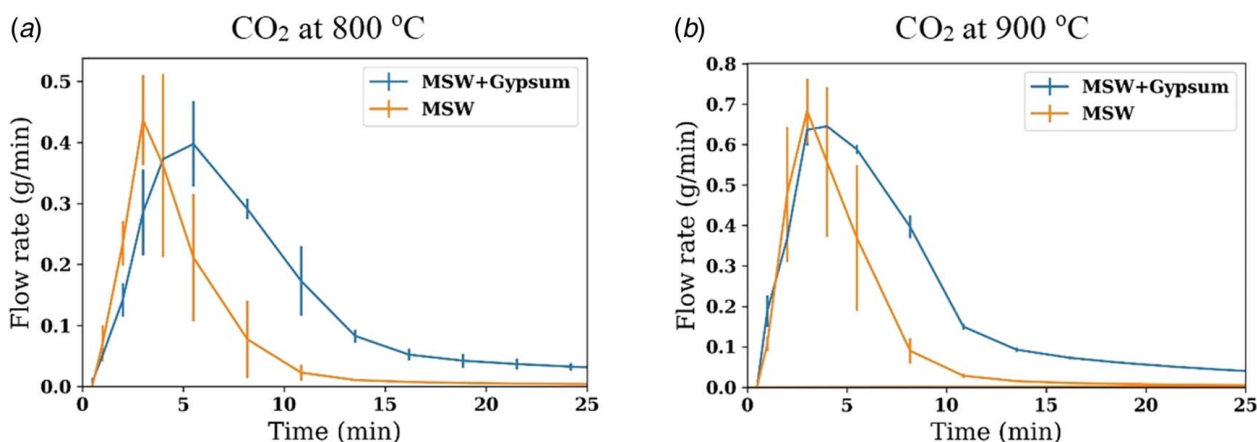


Fig. 7 Impact of gypsum addition to MSW on CO₂ evolution with time during pyrolysis at (a) 800 °C and (b) 900 °C

availability, and chemical kinetics [22]. Figure 7 also shows that increase in temperature led to sharper and higher peak owing to the improved equilibrium of these endothermic reactions. Overall, addition of gypsum almost doubled the cumulative CO₂ yield in these pyrolytic conditions depicting char consumption.

After establishing the effect of gypsum on the evolution of major syngas components in pyrolysis conditions, we can compare these results with syngas components' evolution and their variation with gypsum addition in oxidizing environment (gasifying agent:

10% O₂ and 90% N₂). From Fig. 8, we can see the delayed effect of gypsum addition similar to the pyrolysis case. But at 900 °C, such a delay was not found in H₂ evolution, and the peak heights were similar. So, in oxidizing conditions at high temperature, enhanced H₂ was observed with gypsum incorporation. Cumulatively, gypsum addition increased H₂ yield by over 70% at 900 °C in these oxidizing conditions. The reason for such a behavior is not fully clear as it needs to be derived from the behavior of other syngas components. Additionally, Fig. 8 also shows that increasing

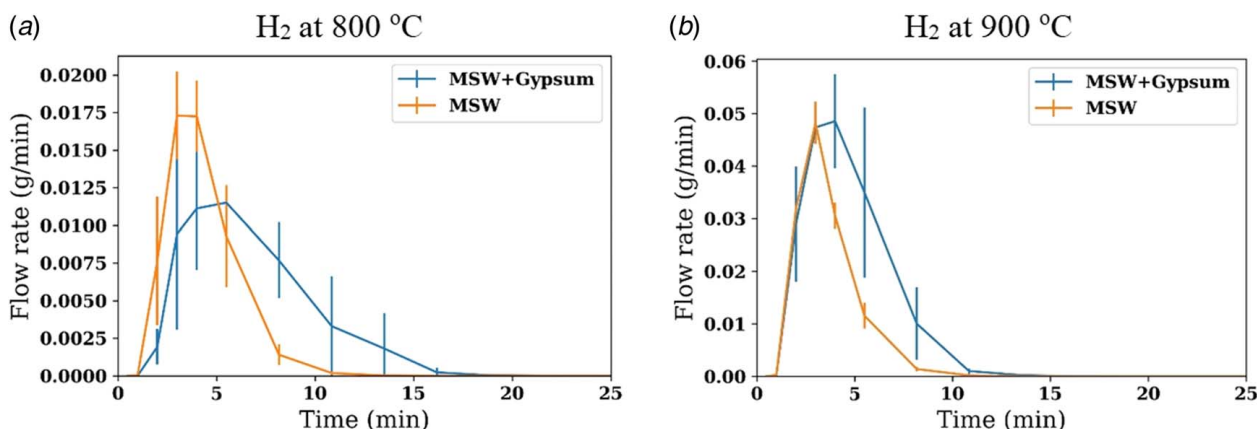


Fig. 8 Impact of gypsum addition to MSW on H₂ evolution with time during oxidation at (a) 800 °C and (b) 900 °C

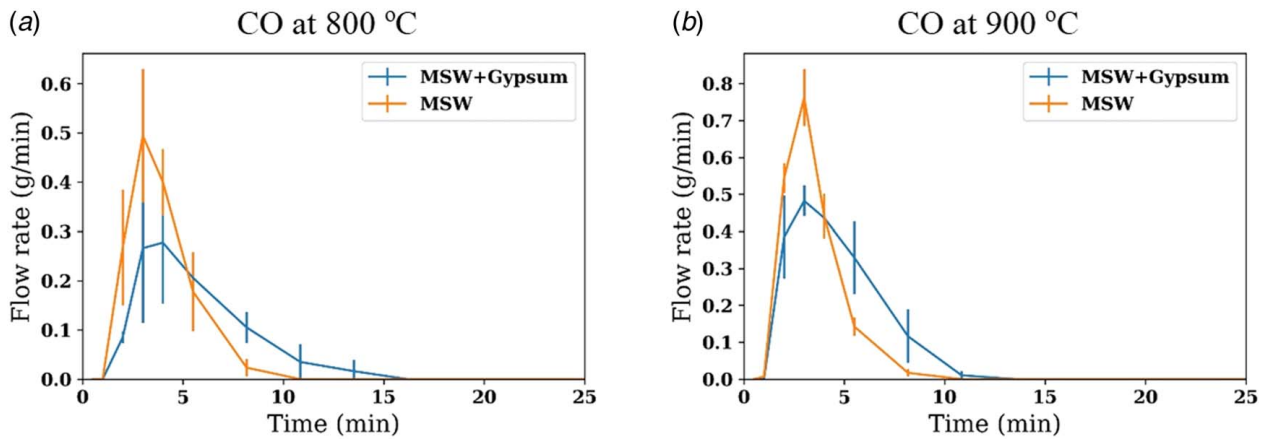


Fig. 9 Impact of gypsum addition to MSW on CO evolution with time during oxidation at (a) 800 °C and (b) 900 °C

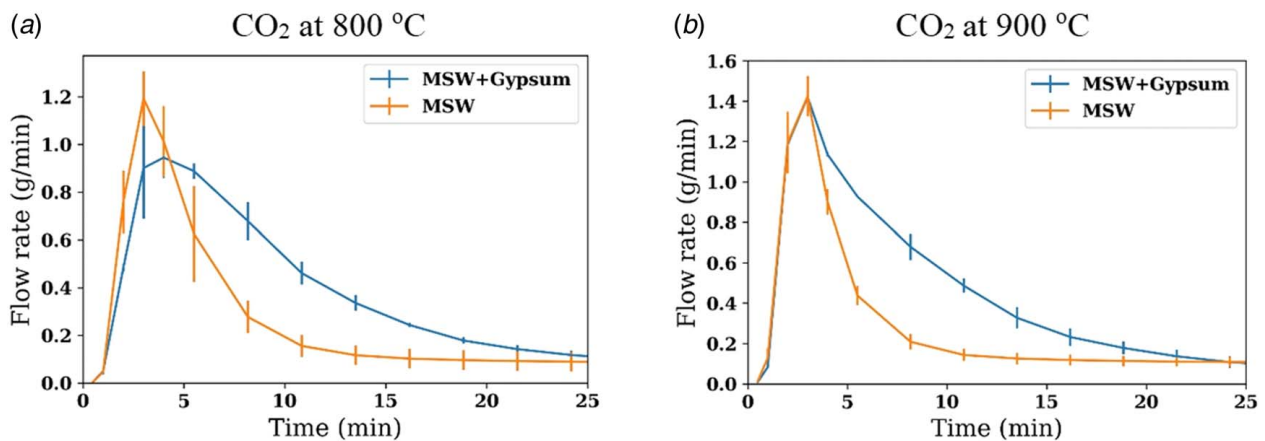


Fig. 10 Impact of gypsum addition to MSW on CO₂ evolution with time during oxidation at (a) 800 °C and (b) 900 °C

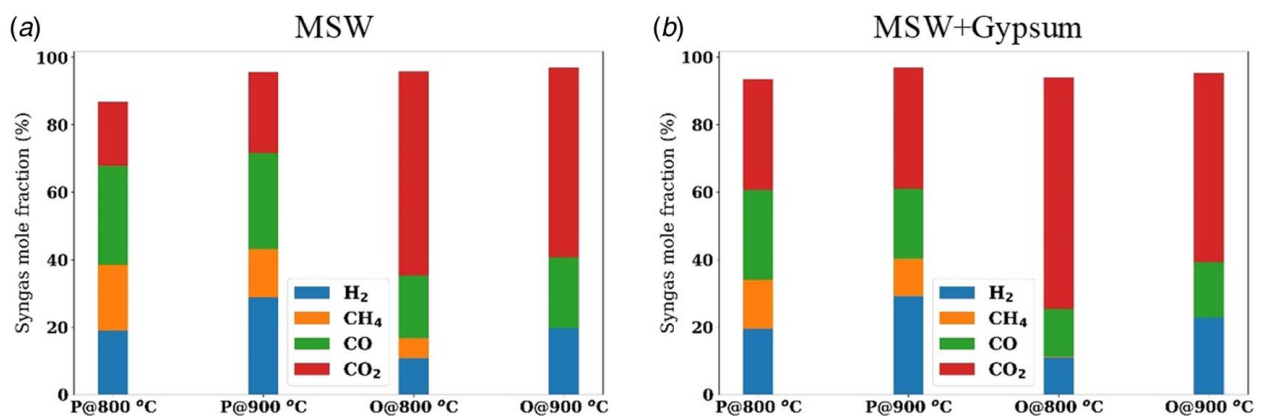


Fig. 11 Molar fractions of major components in syngas in pyrolysis and oxidation of (a) MSW and (b) MSW + gypsum at different temperatures (P: Pyrolysis and O: Oxidation)

temperature led to almost four times higher H₂ flowrate peak in MSW + gypsum gasification while completing its evolution within 10 min. Comparing Figs. 5 and 8 also reveal that the addition of 10% O₂ resulted in increased H₂ flowrate peak. This is because O₂ addition improves the energetics of devolatilization and oxidizes some of the species. This results in improved breakdown of MSW and the resultant CO₂ and H₂O species reform the hydrocarbons for improved H₂ yield.

The effect of temperature on CO evolution during MSW oxidation was also to increase its flowrate and yield, see Fig. 9. Increase in temperature from 800 °C to 900 °C, increased the CO flowrate peak from 0.5 g/min to 0.75 g/min in the case of MSW oxidation and from 0.3 g/min to 0.48 g/min in the case of MSW + gypsum oxidation, unlike MSW pyrolysis wherein the CO flowrate peak values were minimally affected by temperature but only moved to faster (shorter) times, see Fig. 6. We can also see that the total

reaction time zone was affected in the presence of gypsum, especially at 800 °C which led to prolonged flow until 15 min. This significant influence of temperature on the CO flowrate values can be associated with its sensitivity to temperature in the presence of O₂. With increase in temperature, exothermic reactions are not favored but the Boudouard reaction is favored. So, the second step of CO oxidation is not favored and thus leading to more CO yield. In the presence of gypsum, while the kinetics were significantly varied, the total CO yield changed minimally. This resulted in delayed evolution effect as seen in MSW pyrolysis, possibly from CO consumption by gypsum during the MSW heat-up and oxidation. Further insight can be obtained by observing CO₂ evolution under these conditions, see Fig. 10. The results show significant increase in the CO₂ yield in the presence of gypsum even in oxidizing conditions. The cumulative CO₂ increased by 35–40% with the addition of gypsum. In TGA results no high temperature reaction was found between char and gypsum in oxidizing conditions, as char got consumed by the O₂ before gypsum was active. But in macro-scale examination, significant char was available to react, especially after 10 min from the start of experimentation, which resulted in the gypsum led oxidation of the char. This contrast with TGA results to reveals that the oxidation rate of char by O₂ and gypsum are of comparable rate and thus in real MSW disposal reactors, gypsum can have significant improvement in char conversion and lead to improved carbon recovery.

While incorporation of gypsum led to enhanced CO₂, in the pursuit of MSW utilization via syngas, this may mean lowered syngas quality and heating value. This can be observed from the composition of cumulative syngas yield, see Fig. 11. This shows the presence of gypsum led to improved oxidation potential of the feedstock and thus resulted in more oxidized syngas components—lowered CH₄, CO, and increased CO₂. But an interesting observation was that, even with the increased oxidation of syngas components, H₂ fraction in the syngas was unaffected by gypsum addition. It implies that the benefits of gypsum incorporation can include both improved carbon conversion, and lowered char without compromising H₂ output. Further studies are needed to quantitatively assess the energetics and economic benefits, but the results presented here showed that carbon conversion and H₂ yield can be improved from MSW gasification by gypsum addition.

4 Conclusions

Co-processing of synthetic MSW mixtures with gypsum waste from C&D sector was studied at micro and macro-scales to explore the synergistic waste disposal and energy recovery. Mass-loss kinetics and syngas components, and their evolution characteristics were obtained for different temperatures, heating rates, and O₂ concentration in the gasifying agent using micronized mixtures of MSW and 1:1 ratio of MSW and gypsum from drywall. TGA studies revealed the gypsum interaction peaks with MSW to be in two different zones—high temperature peaks (900–1300 K) and low temperature peaks (700–800 K). High temperature peaks were observed for pyrolytic conditions (no oxygen) due to gypsum-char reactions while they disappeared in the presence of O₂ as char was consumed by O₂ prior to these zones. But low temperature peaks appeared irregularly in oxidizing atmosphere which may correspond to reducing char intermediates that form in the presence of O₂ at relatively low temperatures. In pyrolytic conditions, the solid residue mass yield only increased by 10% from 1:1 MSW-gypsum mixtures compared to MSW, while in oxidizing conditions, it increased 40% of the initial feedstock (gypsum mass included). This shows the possibility of gypsum incorporation into MSW gasification without significant adverse effects of increased ash residue.

In macro-scale tests, the addition of gypsum led to delay in the evolution of H₂ and CO while the peak position of CO₂ was unaffected. Gypsum addition to MSW was found to increase cumulative H₂ yield by 20% in pyrolyzing conditions and 70% in oxidizing conditions at 900 °C. Cumulative CO yield was minimally affected

by gypsum addition, while CO₂ yield significantly increased due to gypsum's oxidation of char and CO. Overall, gypsum addition led to significant increase in the carbon conversion of MSW disposal into syngas. Increase in CO₂ led to decreased syngas quality in terms of dilution and the cumulative syngas composition revealed sustained H₂ mole fraction, while change in CO was minimal. But the syngas uniformity was significantly improved as gypsum addition lowered hydrocarbon by-products. These results reveal that gypsum can be incorporated into MSW incineration and gasification to improve syngas yield and uniformity without a significant increase in the resultant ash. These results uncover the potential for synergistic disposal of wastes from municipal and C&D sectors which can improve the techno-economic viability of energy and material recovery from MSW.

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Conflict of Interest

There are no conflicts of interest.

Data Availability Statement

The datasets generated and supporting the findings of this article are obtainable from the corresponding author upon reasonable request.

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