ÁREA TEMÁTICA: Área 3: Reciclagem

SIMULATION TOOLS FOR THE ASSESSMENT OF ADVANCED THERMAL TREATMENT OF MSW – GASIFICATION CODE

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ABSTRACT

The landfill near downtown Brasilia was shut down some years ago. The amount of waste that has been landfilled is in excess of 19,000,000 tons. Strategic studies are being carried out for the energetic utilization of the stored energy through landfill mining. Advanced conversions technologies for municipal solid waste are based on either, pyrolysis or gasification. In this work we present a novel thermochemical equilibrium model to estimate gasification performance of MSW. In most instances, though, the producer gas has to be cleaned for particle matter and tar extraction. Based on that, we proposed a two-step conversion pathway, based on staged gasification in two stand-alone equipment. Carbonization of MSW in a pyrolysis reactor followed by gasification of the derived charcoal for low tar producer gas. Code predictions confirmed the improved quality of the syngas when charcoal is used as the feedstock. Low temperature steam gasification of the derived charcoal further increased the quality of the syngas for Fisher-Tropsch synthesis and heat engine applications by virtue of its high content of hydrogen and carbon monoxide.

Keywords: Environmental remediation; Brasília landfill; Gasification modeling, landfill mining.

INTRODUCTION

The Paris Agreement, signed by the majority of the countries, aimed to reduce greenhouse gases to achieve global mean temperature 2.0 °C below that of pre-industrial era (MMA, 2020). In a regional context, 18 countries from South America stablished specific goals in order to increase the share of renewable energies sources to their electricity matrix (Washburn e Romero, 2019). Brazil, along with Mexico, Chile and Uruguay are leading this course since 2010. As for Brazil, electricity production is mostly renewable. Regardless of such favorable characteristic, the country has set a 35% decrease in its emissions before the year 2026, based on the levels of 2005. In terms of electricity the proposed target for 2023 suggested an 86% nationwide production from renewable sources, in particular that from agricultural waste, which, for the 2015 figures amounted 50 million of TOE. The contribution of the sector may reach 165 million of TOE by the 2050 (EPE, 2018). Converting this huge amount of primary energy into electricity claims advanced thermal treatments, such as pyrolysis and gasification of biomass and Municipal Solid Waste (MSW). These feedstocks, however, should be employed when solar and wind are less competitive in terms of US$/kW, such as for the production of biofuels, chemicals and alternative materials (IEA Bioenergy, 2015). MSW and biomass are readily available for most of the time in a year cycle, as opposed to wind and solar, and would help in stabilizing a multisource electricity production system.

Steam gasification of carbonaceous materials produces a syngas with increased calorific value and high concentrations of hydrogen and carbon monoxide mostly due to the absence of nitrogen (Bartocci et al., 2018). Syngas with such characteristics is a valuable feedstock in the production of hydrocarbons from Fisher-Tropsch synthesis. The absence of nitrogen in the fuel gas also allows the implementation of carbon capture and storage technologies in integrated gasification gas turbine/engines cycles (Krieger et al., 2015).

Tar concentration in producer gas for engine cycle applications is limited to about 100 mg/Nm3 (Milne et al., 1998). Staged gasification technologies have been proposed to circumvent complex gas cleaning systems for biomass gasification (Bui et al., 1994; Bhattacharya et al., 1999; Jaojaruek et al., 2011; Gómez-Barea et al., 2013). In a recent study our research group proposed a different type of staged gasification technology for low tar producer gas (Miranda et al, 2020). The system is based on specific processes taking place in two standalone reactors. First, the biomass or MSW is thermally degraded in a carbonization reactor. In a following process, gasification of the derived charcoal occurs in a single stage, regardless of the reactor's technology, fixed bed, fluidized bed or any other system configuration. By such means, the producer gas is virtually absent of heavy condensable hydrocarbons (tar). Straightforwardly, the downstream gas cleaning system can be of low complexity and cost. Figure 1 shows the two-step biomass and MSW conversion plant for low tar gas production. As it can be seen, both processes take place in two specialized plants, each having the highest level of maturity (TRL = 9) in terms of technology. As such, the system may be integrated based on off-the-shelf components.

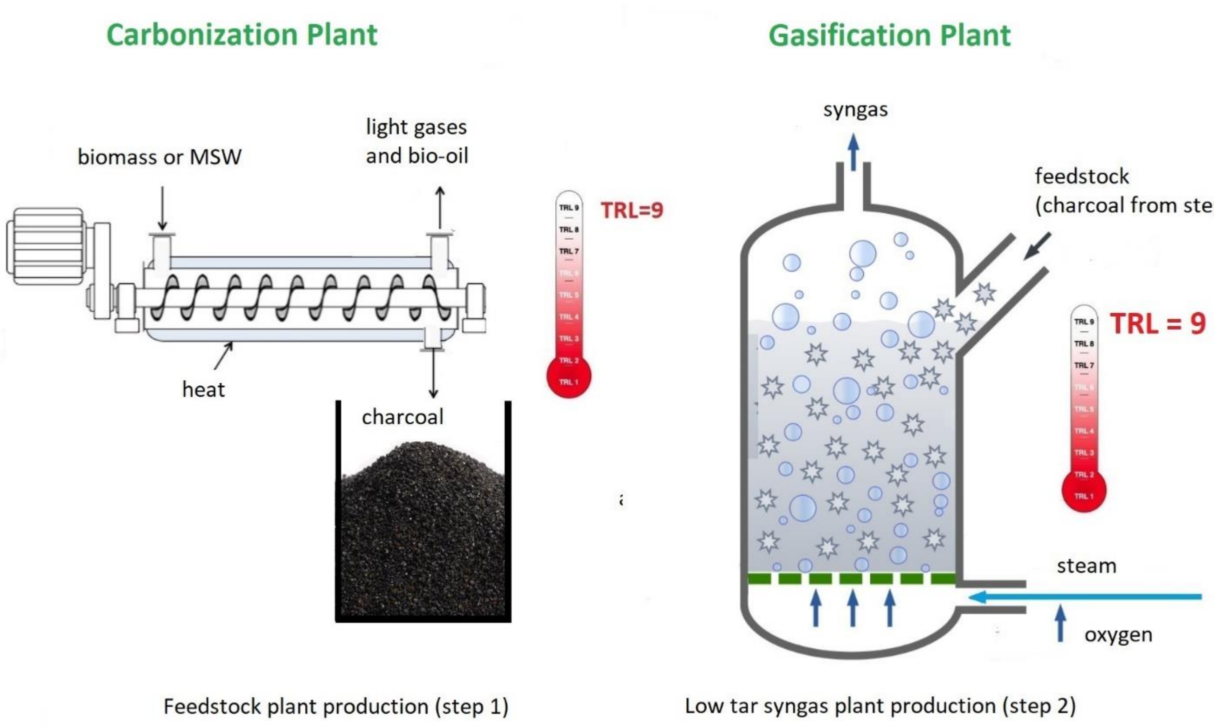

Figure 1: Two-step conversion of carbonaceous feedstock for low tar gas production.

Another interesting feature of the proposed thermal conversion process lies on the possibility of using linear Fresnel systems to produce saturated steam for the carbonization plant. The superheating of the steam would take place in the heat recovery exchanger placed before the gasifier. Steam gasification produces a syngas with improved heating value and suitable for Fischer-Tropsch synthesis.

Following the proposed thermal conversion pathway, our research group is currently developing enhanced simulations tools to assess the performance of integrated MSW gasification gas engine cycle and cogeneration plants. This paper thus presents a novel thermal equilibrium model for gasification reactions of carbonaceous feedstocks. The present model will be further coupled to a pyrolysis and heat engine models applied to advanced thermal treatment of municipal solid waste for large scale electricity production.

CARBONIZATION OF BIOMASS AND MSW

Thermal degradation of biomass and MSW produce bio-oil, charcoal and light gases, at different proportions, depending on process parameters such as the heating rate, maximum temperature, holding time and the composition of feedstock. To some extent, the carbonization process can be designed to produced charcoal with prescribed levels of H/C and O/C ratios, as shown in Fig. 2. The low concentrations of hydrogen and oxygen in charcoal reduces the formation of tar throughout the gasification process. Also, charcoal has improved storage characteristics and much higher heating value and energy density than the original feedstock, whether biomass or MSW. The region which characterizes the MSW in Fig. 2 was taken from Pohl et al. (2018).

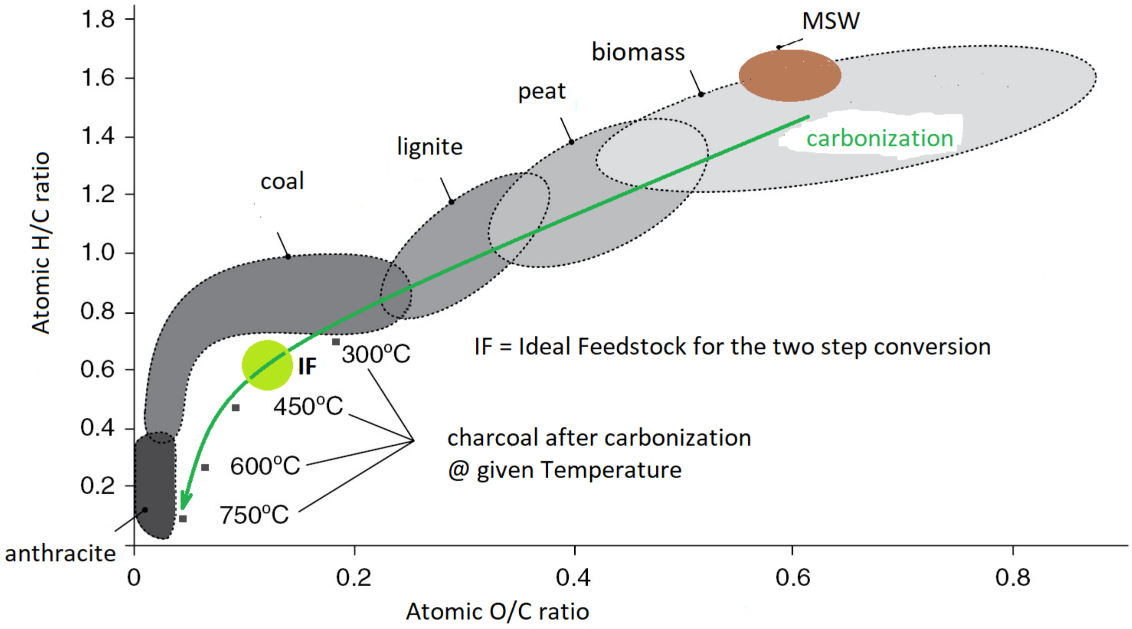

Figure 2: Van Krevelen diagram for carbonaceous feedstocks (Heidenreich et al., 2016).

As it can be seen, carbonization temperature in the range of 300-400 °C reduces the H/C and O/C atomic ratios to about 0.6 and 0,18, respectively. These levels are deemed adequate for low tar syngas production for heat engine applications and Fisher-Tropsch synthesis. In the region of 300 to 400 °C, most of the cellulose and hemicellulose in the feedstock have been thermally degraded. For the proposed conversion pathway, the thermal degradation process has to designed to maximize charcoal yields. For that, the suggested process parameters are: low heating rate and pyrolysis temperature not exceeding 500 °C at some predefined holding time. These process parameters can be obtained from TG analysis, for instance. Low heating rates and mild maximum pyrolysis temperatures are fulfilled by most of the off-the-shelf carbonization reactors available in the market. In this work, the proposed two-step conversion pathway will be applied to the solid fuel waste dropped at the “Estrutural” landfill located near downtown Brasilia. The site had been operating for 50 years and has about 19,724,915 tons of waste. Preliminary sampling from the landfill indicated the waste is comprised of 26% cellulosic material, 23% plastic, 27% water, 21% inert and 3% organics.

Charcoal production from the pyrolysis of this waste mixture would come, almost exclusively, from the cellulosic fraction. Sorum et al. (2001) reported an average of 24.7% in charcoal yields from TG analysis of the cellulosic fraction of MSW up to 450 °C maximum temperature. At 10 °C/min heating rate. Most of the feedstock degradation occurred between 250 and 400 °C and maximum weight losses in the range of 355-371 °C. As for the plastic fraction, charcoal yields are negligible. Based on these figure, we could produce about 1,266,000 tons of charcoal from the slow pyrolysis of the MSW from the “Estrutural” landfill.

GASIFICATION MODEL

Gasification Reaction

For the present work, the gasification reaction of a carbonaceous material is given by

$$n_{fs}^{i}\left[\left(C_{X_C}H_{X_H}O_{X_O}N_{X_N}S_{X_S}\right)_{fs}+\alpha H_2O+\beta SiO_2\right]+n_{H2O}^{i}H_2O+n_{CO2}^{i}CO_2+n_{ox}^{i}(O_2+\gamma N_2)\rightarrow\ n_C^{o}C_s+$$
$$n_C^{o}C_s+n_{H2}^{o}H_2+n_{CO}^{o}CO_2+n_{CH4}^{o}CH_4+n_{H2O}^{o}H_2O+(\gamma n_{ox}^{i}+\tfrac{X_N}{2})N_2+n_{SiO2}^{o}SiO_2O \qquad (1)$$

In Eq. (1), the feedstock composition accounts for the presence of water and ash, through parameter α and β, respectively. Gasification agent is composed of a mixture of steam, carbon dioxide, oxygen and nitrogen at prescribed composition, after adjusting the stoichiometric number of moles for each substance. Both, the feedstock and specific gasification agents can be introduced into the reactor at any given temperature.

Conservation of chemical elements (C, H, O, N, S) is given by

$$n_{CO}^{o}+\ n_{CO2}^{o}+n_{CH4}^{o}+n_C^{o}-\ n_{fs}^{i}\,X_c\ -\ n_{CO2}^{i}=0 \qquad (2a)$$

$$2n_{H2}^{o}+2n_{H2O}^{o}+4n_{CH4}^{o}-\ n_{fs}^{i}X_H-2\,n_{fs}^{i}\alpha-2n_{H2O}^{i}=0 \qquad (2b)$$

$$n_{CO}^{o}+\ 2n_{CO2}^{o}+n_{H2O}^{o}-\ n_{fs}^{i}\,X_O-\ n_{fs}^{i}\alpha-\ n_{H2O}^{i}-\ 2n_{CO2}^{i}-2n_{ox}^{i}=0 \qquad (2c)$$

$$n_{N2}^{o}-\ \gamma n_{ox}^{i}-(n_{fs}^{i}\,X_N)/2=0 \qquad (2e)$$

$$n_{SiO2}^{o}-\ n_{fs}^{i}\,\beta SiO_2=0 \qquad (2f)$$

Only Eq (2a) to Eq. (2c) need integration. The output mass of nitrogen and sulfur can be obtained straightforwardly by the composition of the biomass and the gasification agent.

The as received composition of the feedstock (m_{ar}), its ash (*ASH*)) and moisture contents (*MC*), in terms of percentages, are used to infer the relative number of moles of *H_2O* and *SiO_2*, represented by α and β. For a given mass of as received feedstock (m_{fs}), α and β are estimated as follows

$$\alpha=\frac{m_{water}}{MW_{H2O}} \qquad (3)$$

$$\beta=\frac{m_{wash}}{MW_{ash}} \qquad (4)$$

We thus need to infer the mass of water from moisture and ash of the as received feedstock. The mass balance for the main components are then

$$m_{fs}\ =\ m_{ar}-m_{water}-m_{ash} \qquad (5)$$

In Eq. (5) the mass of ash and moisture is calculated from their contents (%) in the as received feedstock

$$m_{ash}\ =\ \frac{ASH}{100}\ m_{fs} \qquad (6)$$

$$m_{water}\ =\ \frac{MC}{100}\ (m_{fs}+m_{ash}) \qquad (7)$$

The output of unconverted solid carbon may be given by a predetermined efficiency (n_{CC}) of conversion such as

$$n_C^{o}\ =n_{fs}^{i}\,X_C(1-n_{CC}) \qquad (8)$$

The amount and composition of the gasification agent is given by a simplified equivalence ratio expression (Mendiburu et al., 2014), as follows

$$ER_g\ =\ \frac{m_{ox}}{m_{ox\_st}} \qquad (9)$$

For a non-prescribed gasification temperature, the equation of energy needs to be solved. Assuming gasification as a process without work and heat interactions the enthalpy of the products is equal to the enthalpy of the reactants. The enthalpy of the products is defined from the equilibrium composition of the syngas (second law). The absolute enthalpy of the reactants is made with the contribution of the sensible and formation parts of the feedstock and the gasification agent. The enthalpy of formation of the feedstock can be calculated by (Mendiburu et al., 2014)

$$\left(\bar{h}^0_{f-298}\right)_{fs} = HHV\left(MW_{fs}\right) + \left(\bar{h}^0_{f-298}\right)_{CO_2} + \frac{X_H}{2}\left(\bar{h}^0_{f-298}\right)_{H2O(l)} \qquad (10)$$

where

$$HHV = 0.3491C + 1.1783H + 0.1005S - 0.1034O - 0.0151N - 0.0211ASH \qquad (11)$$

The enthalpy of the remaining substances at any given temperature and pressure are obtained directly from the JANAF thermochemical tables inserted in the Engineering Equation Solver code.

Numerical Modeling

The composition of the gasification products is obtained through minimization of the Gibbs free energy giving by

$$g = \frac{G}{RT} = \sum_{i=1}^{N} n_i^g \left[\frac{\bar{G}^0_{f,i}}{RT} + ln\frac{n_i}{n_t} + ln\frac{P}{P_0}\right] + \sum_{i=N+1}^{L} n_i^{l,s} \ \frac{\bar{G}^0_{f,i}}{RT} \qquad (12)$$

For the syngas species concentration estimates, we follow the scheme proposed in Mendiburu et al. (2014), based on the minimization of objective function, Eq. (12), using Lagrange multipliers.
The sum of the mass conservation of every chemical element after multiplied by the Lagrange multipliers gives the following auxiliary equation

$$L = \sum_{j=1}^{K} \lambda_j \left[\sum_{i=1}^{M} \left(a_{ji} n_i\right)_P - \left(b_j\right)_R\right] = 0 \qquad (13)$$

Another auxiliary equation relates Eq. (12) and Eq. (13) through

$$H = g - L \qquad (14)$$

The minimization is obtained by the partial derivative of H in relation to the Lagrange multipliers and the number of moles of each species, that are equally null, in the gas product of the reaction.
The problem is therefore, find the product gas composition that satisfies the following equation

$$\left[\frac{\bar{G}^0_{f,i}}{RT} + ln\frac{n_i}{n_t} + ln\frac{P}{P_0}\right]_g + \left[\frac{\bar{G}^0_{f,i}}{RT}\right]_l + \left[\frac{\bar{G}^0_{f,i}}{RT}\right]_s - \frac{\partial}{\partial n_i}\left(\sum_{j=1}^{K} \lambda_i \left[\sum_{i=1}^{M} \left(a_{ji} n_i\right)_P - \left(b_j\right)_R\right]\right) = 0 \qquad (15)$$

In many thermochemical equilibrium codes, the amount of methane in the syngas is prescribed. In this work, we employed a correlation proposed in Mendiburu et al. (2014), for the number of moles of methane in the producer gas.

$$n^o_{CH4} = \frac{P_{CH4}}{100}\left[\frac{\eta_{CC} X_C + \frac{X_H}{2} + \frac{m_{water}}{MW_{H2O}} + \frac{X_N}{2} + \gamma\, n^i_{ox} - n^o_{H2O}}{1 + \frac{P_{CH4}}{50}}\right] \qquad (19)$$

The value of P_{CH4} in Eq. (19) can be seen found in Mendiburu et al. (2014).

Solution Algorithm

The main set of equations Eq. (1) to Eq. (19) was implemented in the EES code. For engineering applications, the code also solves many additional equations in order to estimate the gas composition in dry basis. The gas HHV is then compared to the biomass energy input to infer the so-called cold gas efficiency (η_g) for the thermal conversion. The total number of equations in the EES code was 164. The main input parameters are, the feedstock composition from proximate and ultimate analysis, the composition of the gasification agent and their temperature and pressure. The code can calculate product gas composition with prescribed gasification temperature or through the conservation of energy equation.

RESULTS AND DISCUSSION

The proposed model was the applied to the gasification of biomass, for validation purposes, followed by some predefined MSW charcoal compositions based on the Van Krevelen diagram showed in Fig. 2.

The ultimate analysis of the reference biomass, as received, shows 75,5% volatile matter, 11% fixed carbon 0,5% ash content and 13% moisture. As for the ultimate analysis, the composition of the reference biomass is 51% carbon, 6% hydrogen, 42.7% oxygen and 0.3% nitrogen.

Initially, gasification was set to take place at 1.0 bar (pressure) with prescribed temperature of 850 °C and 0.3 for the equivalence ratio, as conducted in the experiments of Wei et al. (2011). The gasifier was fed with biomass and gasification agent (air) at ambient temperature. Based on this input parameters, the calculated cold gas efficiency was near 27%. Dry gas composition, as estimated by the model, in volumetric basis was (in parentheses the experimental results of Wei et al.) 2.2% for methane (2.41%), 26% for carbon monoxide (21.62%), 19% for hydrogen (19.2%)), 8,6% for carbon dioxide (11.7%) diluted in nitrogen. As it can be seen, the model predicted a syngas composition in close agreement with the experimental results, from an engineer point of view. The HHV of the gas was estimated as 6648 kJ/Nm^3.

After this validation process, the code was applied to the gasification of charcoal derived from MSW in different gasification agent composition and reactor temperatures. Additionally, we also estimated the gasification performance of an MSW composition without pre-treatment, except drying prior to gasification ($CH_{1.6}H_{0.6}$). Thermal equilibrium code predictions of gasification of two charcoals derived from municipal solid pyrolysis are also shown in Table 1, in addition to the raw dried MSW.

The performance of the feedstocks after gasification was investigated based on the following input parameters:

1. Gasification with air at room temperature and steam preheated to either 600 or 900 °C, matching gasifier temperature;
2. Gasification take place at sea level pressure;
3. Steam to dry charcoal ratio, by mass, was set to 0.20;
4. Feedstock (CHxOy) enters the reactor with 3% moisture content at 25 C;
5. Optimized equivalence ratio was in the range of 0.25 to 0.35 3.0, for gasification temperatures of 600 and 900 °C;
6. Producer gas cooled to 25 °C;
7. % vol. of CH4 in the producer gas calculated with the help of Eq. (19);
8. Char losses equivalent to 1% of dry charcoal input.

Table 1: Gasification performance parameters of charcoals from the slow pyrolysis of MSW.

Analysis	$CH_{0.8}O_{0.2}$ 600 °C	900 °C	$CH_{0.5}O_{0.1}$ 600 °C	900 °C	$CH_{1.6}H_{0.6}$ 600 °C	900 °C
Gas composition (dry wt.%)						
	34.3	35.4	34.1	34.6	18.2	23.8
CO	2.3	1.5	2.1	1.7	13.2	9.0
CO_2	20.8	20.7	16.5	14.8	24.6	22.5
H_2	2.1	1.9	2.1	2.1	2.1	1.7
CH_4						
Energy parameters					5,712	6,008
LHV (kJ/Nm3)	7,337	7,382	6,623	6,675	23.5	25.3
Cold Gas Efficiency (%)	34.6	35.1	32.1	32.4		

As it can be seen from Table 1, the cold gas efficiency for MSW and their charcoals are in the range of 23 to about 35% while lower heating value, in volumetric basis, varied from 6,000 to about 7,400 kJ/Nm^3.

The results from Table 1 show that, gasification of charcoal gives improved syngas quality, in terms of heating value, for heat engine applications and higher concentrations of CO ans H2, for Fisher-Tropsch synthesis. Another relevant feature of the syngas obtained from charcoal gasification would be the very low tar concentration, as expected from the proposed two-step energy conversion system.

Improvements can be further attained by steam gasification of the derived charcoals. The use of superheated steam in combination with small amounts of oxygen, would give a high-quality gas either as energy carrier or as feedstock for the production of chemicals.

We thus performed a simulation of a steam gasification process of the carbonized MSW with whose chemical composition gives an H/C and O/C ratios of 0.7 and 0.2, respectively. From Fig. 2 we can see that such charcoal would be produced by a slow pyrolysis process in the temperature range of 350 to 400 °C.

The producer gas from steam gasification of charcoal ($CH_{0.7}O_{0.2}$) derived from MSW, at 600 °C would give a predicted composition (dry basis) of 2.07% for methane, 59.1% for carbon monoxide, 38,5 for hydrogen and about 0.5% for carbon dioxide. With this composition, the gas has LHV of 12347 kJ/Nm^3. The cold gas efficiency is about 63%.

CONCLUSION

This paper presented the preliminary results of a thermochemical equilibrium model developed to assess the gasification performance of charcoals derived from slow pyrolysis of MSW. The obtained producer gas would be absent of tarry compounds thus claiming a simple low-cost gas cleaning system prior to its utilization in any heat engine cycle. Carbonization of MSW would take place in any off-the-shelf hardware available in the market due to the low heating rate and maximum temperature requirements for the conversion plant. Likewise, the gasifier would be of a single stage and the technology would be selected mostly based on the charcoal particle size, rather that from its composition in addition to the convention rate. Gasification of charcoal from MSW produces a gas with improved composition and heating value as compared to the parent feedstock. Steam gasification of charcoal derived from MSW would further improve gas quality.

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