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Particulate Emission from the Gasification and Pyrolysis of Biomass: Concentration, Size Distributions, Respiratory Deposition-based Control Measure Evaluation

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Abstract

Gasification and pyrolysis technologies have been widely employed to produce fuels and chemicals from solid wastes. Rare studies have been conducted to compare the particulate emissions from gasification and pyrolysis, and relevant inhalation exposure assessment is still lacking. In this work, we characterized the particles emitted from the gasification and pyrolysis experiments under different temperatures (500, 600, and 700 °C). The collection efficiencies of existing cyclones were compared based on particle respiratory deposition. Sensitivity analysis was conducted to identify the most effective design parameters. The particles emitted from both gasification and pyrolysis process are mainly in the size range 0.25-1.0 μm and 1.0-2.5 μm. Particle respiratory deposition modelling showed that most particles penetrate deeply into the last stage of the respiratory system. At the nasal breathing mode, particles with sizes ranging from 0.25 to 1.0 μm account for around 91%, 74%, 76%, 90%, 84%, and 79% of the total number of particles that deposit onto the last stage in the cases of 500 °C gasification, 600 °C gasification, 700 °C gasification, 500 °C pyrolysis, 600 °C pyrolysis, and 700 °C pyrolysis, respectively. At the oral breathing mode, particles with sizes ranging from 0.25 to 1.0 μm account for around 92%, 77%, 79%, 91%, 86%, and 81% of the total number of particles that deposit onto the last stage in the six cases, respectively. Sensitivity analysis showed that the particle removal efficiency was found to be most sensitive to the cyclone vortex finder diameter ($D_0$). This work could potentially serve as the basis for proposing health protective measures against the particulate pollution from gasification and pyrolysis technologies.

Keywords: Gasification; Pyrolysis; Biomass Waste; Particle Emission; Control Measure

Capsule
A particle respiratory deposition-based cyclone design scheme is proposed for health protective measures against the particulate pollution from gasification and pyrolysis technologies.

1 Introduction

Application of waste biomass for energy production is receiving increasing attention because of the renewability and carbon neutrality features of biomass. Among the plethora of biomass-to-energy technologies, gasification and pyrolysis are considered as two of the most efficient ways to produce fuels and chemicals from biomass (Ong et al., 2015). During the gasification process, carbonaceous feedstock is converted into fuel gas, hydrocarbons, and a small amount of solid residue (i.e. biochar and ash) in an oxygen-deficient environment (You et al., 2017b).

Pyrolysis is a thermochemical decomposition of organic material in the absence of oxygen and its major products include pyrolytic oil and biochar (Kan et al., 2016).

Biomass gasification and pyrolysis are not emission-free technologies. Thermochemical treatment of biomass was found to be one of the most important sources of PM$_{2.5}$ (particles of an aerodynamic diameter smaller than 2.5 $\mu$m) in both developed and developing countries (Glasius et al., 2006; Johnson, 2016; Saarikoski et al., 2008; Ward and Lange, 2010; Zhang et al., 2010). Several previous works have been devoted to characterizing the particulate emissions of gasification and pyrolysis. For example, the effects of feedstock moisture content on the PM$_{2.5}$ emission of a semi-gasified cookstove were investigated and PM$_{2.5}$ emission factors were found to decrease with the increase of moisture content (Huangfu et al., 2014). The particulate emissions at three locations of a fixed bed, downdraft gasifier were measured to determine the particle removal effectiveness of packed-bed filter (Hamilton et al., 2014). In the study, particle mass concentrations (PMCs) were found to be approximately 75 mg/Nm$^3$ in the pre-filtered producer gas and were reduced by 99% by the packed-bed filter to
about 1 mg/Nm\(^3\). A nine-stage cascade impactor was used to investigate the size distribution of particles from the gasification of municipal solid waste (MSW) (Shiota et al., 2017). The results showed that most of the particles were fine particles. PM\(_{2.5}\) emission from gasification and melting of MSW accounted for 2.5% of total PM\(_{2.5}\) emissions from all sorts of thermal treatment of MSW in Japan. Trace metals emission and size distribution of PM generated from biomass gasification were reported in several studies (Min et al., 2016; Nzihou and Stanmore, 2013; Pudasainee et al., 2014). The emission of potassium- and sodium-containing compounds during rapid birchwood pyrolysis was also studied (Davidsson et al., 2002). The results showed that the alkali emission per unit mass for small particles was higher than that for large ones and this tendency increased with temperature. A systematic study was conducted to characterize the emission of trace elements (e.g., As, Cu, Cr, Ni, V, Co, Cd, and Pb) generated from a two-stage pyrolysis/combustion reactor system (Liaw et al., 2016). The study indicated that particles produced from the combustion of volatiles produced in situ from the pyrolysis stage were smaller than 1 \(\mu\)m, and had a uni-modal distribution with a fine mode diameter of 0.043 \(\mu\)m.

Particulate emissions could potentially pose two adverse effects on the practical deployment of gasification and pyrolysis systems. First, particles could cause fouling, reduce system efficiency, contaminate the producer gas which affects the subsequent power generation and syngas upgrading processes (Hamilton et al., 2014; Wang et al., 2011; Woolcock and Brown, 2013). It was designated that the PM\(_{10}\) and PM\(_5\) concentrations in syngas should be lower than 50 mg/m\(^3\) and 30 mg/m\(^3\), respectively, for internal combustion engine and gas turbine, while the PM concentration should be lower than 0.02 mg/m\(^3\) for methanol synthesis (Woolcock and Brown, 2013). Second, gasification is a technology that is well suitable for decentralized application in terms of energy efficiency and economic feasibility (Buragohain et al., 2010;
You et al., 2016b). The decentralized application of the gasification technology in modern cities means the proximity of gasification systems to the public and residential communities. This imposes strict requirements on its emission control and management because elevated outdoor particle concentrations pose a potential health concern, especially for fine particles such as PM$_{2.5}$ that could penetrate deeply into the human respiratory system (Happo et al., 2013; Jalava et al., 2012; Moller and Loft, 2010; Naeher et al., 2007; You et al., 2016a). Our recent study also showed that elevated outdoor PM$_{2.5}$ may lead to increased household energy use and corresponding household carbon footprint by changing the energy utilization mode (e.g., more air-conditioner and purifier use) (You et al., 2017a).

Therefore, it is necessary to propose effective preventive and control measures towards particulate emissions from gasification and pyrolysis, which is highly contingent upon the understanding of the mechanisms and characteristics of particulate emission. Rare studies have been conducted to analyze the influences of operating conditions (e.g., gasification agents and temperature) towards time-resolved particulate emissions. In addition, possible particle formation mechanisms for gasification and pyrolysis are still lacking. In this work, we compared the particle removal efficiencies of existing cyclones with different configurations based on particle respiratory deposition. Sensitivity analysis was conducted to identify the most effective design parameters. This work could potentially serve as the basis for proposing health protective measures against the particulate pollution from gasification and pyrolysis technologies.
2 Materials and Methods

2.1 Experiments and Emission Calculation

2.1.1 System and instrumentation

To explore the fundamental mechanisms governing the particulate emission of the thermo-chemical processes, experiments were conducted in a lab-scale reactor (capacity: ca. 1g/min). A schematic diagram of the experimental setup is shown in Figure 1. The aerosol from experiments was guided into a fume hood for diluting and exhaust cooling. An aerosol spectrometer (GRIMM 1.109) was used to measure particle number concentrations (PNCs) in the fume hood. Another aerosol spectrometer was placed outside of the fume hood to measure the background PNCs. The aerosol spectrometers measured particles in the size range between 0.25 and 32 µm at an interval of 6 seconds. The duration of each sampling was about 30 min. A flask was used for tar condensation and retaining. Air and nitrogen were used as the agent for the gasification and pyrolysis experiments, respectively and the agent flow rates were fixed at 300 mL/min using a mass flow controller. 15g of feedstock was used for each experiment. For gasification experiment, the equivalence ratio (ER) was fixed as 15% during 30 minutes of experiment. \( ER = \frac{F_{\text{air}}}{F_S} \left( \frac{F_{\text{air}}}{F_S} \right)_{\text{Stoichiometric}} \) where \( F_{\text{air}} \) is the inlet air mass flow rate, \( F_S \) is the biomass feeding rate. Three temperatures (i.e. 500, 600, and 700 °C) were tested both for the gasification and pyrolysis experiments. The reactor was heated by electrical heater and the temperature was controlled by Programmable Logic Controller (PLC). Woodchips were used as the feedstock whose proximate, ultimate compositions were given in Table 1.
Figure 1. A schematic diagram of experimental setup. 1 Gas cylinder, 2 Mass flow controller, 3 Valve, 4 Gas mixer, 5 Gasifier reactor, 6 Heater, 7 Flask, 8 Aerosol spectrometer, 9 Fume hood, 10 Aerosol spectrometer.

Table 1. Feedstock composition.

<table>
<thead>
<tr>
<th>Proximate analysis (dry basis, wt. %)</th>
<th></th>
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<tbody>
<tr>
<td>Moisture</td>
<td>8.2-8.5</td>
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<tr>
<td>Volatiles</td>
<td>67.8-69.2</td>
</tr>
<tr>
<td>Fixed carbon</td>
<td>16.2-17.5</td>
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<tr>
<td>Ash</td>
<td>6.2-6.3</td>
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</table>

<table>
<thead>
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<th>Ultimate analysis (wt. %)</th>
<th></th>
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<tr>
<td>Carbon</td>
<td>43.3-44.2</td>
</tr>
<tr>
<td>Hydrogen</td>
<td>5.4-6.1</td>
</tr>
<tr>
<td>Oxygen</td>
<td>41.6-42.5</td>
</tr>
<tr>
<td>Nitrogen</td>
<td>0.9-2.1</td>
</tr>
<tr>
<td>Sulfur</td>
<td>0.5-1.0</td>
</tr>
</tbody>
</table>
2.1.2 Particulate emission calculation

The particle number concentration, $PNC_{it}$ (#/L), was backward estimated from the concentration data based on the mass balance model

$$\left(f_1 C_{ot} - f_2 C_{it} - f_3 C_{it} + f_4 PNC_{it}\right) \Delta t = (C_{it(t+\Delta t)} - C_{it}) V \tag{1}$$

where $\Delta t = 6$ s is the sampling interval. $C_{ot}$ (#/L) and $C_{it}$ (#/L) are the PNC outside and inside the fume hood at the time $t$. $C_{ot}$ corresponds to the measurements of the aerosol spectrometer outside the fume hood. $C_{it(t+\Delta t)}$ (#/L) is the PNC inside the fume hood at the time $t + \Delta t$. $C_{it}$ and $C_{it(t+\Delta t)}$ correspond to the measurements of the aerosol spectrometer inside the fume hood. $V = 1.09 \times 10^3$ (L) is the volume of the fume hood space. $f_1$ (L/s), $f_2 = 300$ (L/s), $f_3 = 2 \times 10^{-2}$ (L/s), and $f_4$ (L/s) are the air flow rates entering the fume hood from outsides, exiting the fume hood, entering the aerosol spectrometer, and entering the fume hood from the reactor, respectively (Figure 1). We have $f_1 + f_4 = f_2 + f_3$. The PNC was

$$PNC_{it} = \left[\frac{(C_{it(t+\Delta t)} - C_{it}) V}{\Delta t} - f_1 C_{ot} + f_2 C_{it} + f_3 C_{it}\right] / f_4 \tag{2}$$

We do not consider particle deposition because it is negligible compared to the ventilation effect.

2.2 Gas cyclone design based on respiratory deposition modelling

Cyclones have been considered as an economical approach for particle removal in gasification systems. Different types of cyclones (e.g., Stairmand cyclone, Lapple cyclone, German Z cyclone, and Southern Research Institute (SRI) cyclones II and III) are available and differentiated in terms of their geometrical configuration. A schematic diagram of cyclone is given in Figure 2. In this work, we compared the particle removal efficiencies of existing cyclones with different configurations based on particle respiratory deposition. Sensitivity
analysis was conducted to identify the most effective design parameters. The results could be used to facilitate the design of cyclone for gasification and pyrolysis.

Figure 2. A schematic diagram of cyclone geometry and design parameters for different types of cyclones. (D: cyclone body diameter; D₀: cyclone vortex finder diameter; D₃: cyclone bottom diameter, H: height of cyclone inlet slit, L₀: height of cylindrical part of cyclone, L₃: height of conical part of cyclone, S: vortex finder length, W: width of cyclone inlet slit.)

<table>
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<tr>
<th>Common parameters</th>
<th>D/D₀</th>
<th>H/D</th>
<th>W/D</th>
<th>S/D</th>
<th>L₀/D</th>
<th>L₃/D</th>
</tr>
</thead>
<tbody>
<tr>
<td>D (mm)</td>
<td>20</td>
<td>25</td>
<td>30</td>
<td>35</td>
<td>40</td>
<td>45</td>
</tr>
</tbody>
</table>

2.2.1 Particle removal by cyclone

The removal efficiency of the cyclones could be fitted with a linear function (Lidén and Gudmundsson, 1997) as:

\[
\ln \left( \frac{E}{1-E} \right) = W₀ + W₁Z
\]

where the dimensionless parameter \( Z \) is calculated by

\[
Z = \frac{\sqrt{C_{cmd}(d_{ae})}d_{ae}}{\sqrt{C_{cmd}(d_{ae,50})}d_{ae,50}} - 1
\]

where \( d_{ae} = dₑ\left( \frac{ρ₀}{C_{cmd}(d_{ae})} \right)^{1/2} \) is the aerodynamic diameter with \( dₑ \) being the equivalent volume diameter and \( C_{cmd} \) being Cunningham correction factor, \( C_{cmd} \) is related to the particle Knudsen number \( Kn \) and is calculated by
\[ C_{cmd} = 1 + Kn(A + B \times e^{-C/Kn}) \]  

(5)

Where \( Kn = \frac{2\lambda}{d_e} \), \( A = 1.257 \), \( B = 0.40 \), \( C = 1.10 \), and \( \lambda = 80 \text{ nm} \) is the particle mean free path (Friedlander, 2000). \( d_{ae,50} \) is the cut-off aerodynamic diameter and calculated by Muschelknautz's model (Hoffmann and Stein, 2002)

\[ d_{ae,50} = x_{fact,ae} \sqrt{\frac{18 S_\vartheta (0.9 Q_v)}{2\pi (\rho_g - \rho_d) v_{\theta CS}^2 (L_b + L_c - S)}} \]  

(6)

where \( L_b \) (m) is the height of cylindrical part of cyclone, \( L_c \) (mm) is the height of conical part of cyclone, \( x_{fact,ae} \) is the correction factor and it normally falls within the range of 0.9~1.4. In this study \( x_{fact,ae} \) was chosen to be 1.2. \( Q_v \) (L/min) is the volumetric flow rate. \( S \) (m) is the vortex finder length, \( \mu_g \) (kg/(m·s)) is the gas dynamic viscosity. \( v_{\theta CS} \) (m/s) is the spin velocity near the wall

\[ v_{\theta CS} = v_{\theta w} (D/D_0)/(1 + \frac{f_S f_D \nu_{\theta w} (D/D_0)}{2Q_v}) \]  

(7)

where \( f = 0.314Re^{-0.25} \) is the wall friction factor (Karagoz and Avci, 2005). \( Re = \frac{\rho_g u_{in} (D-D_0)/2}{\mu_g} \), \( D \) (m) is the cyclone body diameter, \( D_0 \) (m) is the cyclone vortex finder diameter. \( v_{\theta w} \) (m/s) is the geometrical mean rotational velocity

\[ v_{\theta w} = u_{in} R_{in} / (\alpha R_m) \]  

(8)

where \( u_{in} \) (m/s) is the average gas velocity at cyclone inlet. \( R_{in} \) (m) is the radial position of the centre of the inlet, \( R_{in} = D/2 - W/2 \). \( R_m \) (m) is the geometric mean radius. \( R_m = \sqrt{(D/2)(D_0/2)} \). \( \alpha \) is the first regression coefficient, \( \alpha = 0.04 \). \( S_f \) (m²) is the friction surface estimated by

\[ S_f = \frac{\pi}{4} [D^2 - D_0^2 + 4(DL_b + D_0 S) + (D + D_d)\sqrt{4L_c^2 + (D - D_d)^2}] \]  

(9)

with \( D_d \) (m) being the cyclone bottom diameter.
2.2.2 Respiratory deposition modelling

The cyclone design is based on particle respiratory deposition modelling. (You et al., 2017c) summarized the existing particle deposition models (Chan et al., 1980; Cheng, 2003; Cohen and Asgharian, 1990; Kim and Fisher, 1999; Kim and Iglesias, 1989; Zamankhan et al., 2006; Zhang et al., 2008). As shown in Figure S1, the airways were divided into five stages for modelling submicron particle deposition in the human respiratory system [oral (stage 1) and nasal (stage 1) airways, trachea (stage 2), bronchial airways from B1 to B6 (stage 3) and from B7 to B15 (stage 4), and the rest of airways (stage 5)]. For modelling supermicron particle deposition in the human respiratory system, the airways were divided into four stages [oral (stage 1) and nasal (stage 1) airways, trachea (stage 2), the bronchial airways from B1 to B19 (stage 3), and the rest of airways (stage 4)]. The boundary between submicron particles and supermicron particles is defined as 1 micrometer. The compiled models and parameters for particle respiratory deposition modelling is given in Table S1-S3 (please see the Supplementary Material).

In this study, six types of commonly used cyclones (i.e. C-20, C-25, C-30, C-35, C-40, and C-45) were compared in terms of their effects on mitigating particle respiratory deposition. The corresponding design parameters of the cyclones are shown in Figure 2 (Sagot et al., 2017). A case without cyclone was also considered for the purpose of comparison.

2.3 Sensitivity analysis

Parameter investigation was conducted to explore the relationships between the design parameters ($X/X_0$, where $X$ means the ratio of design parameter to cyclone diameter $D$, $X_0$ means original ratio of design parameter to cyclone diameter $D$) of cyclone geometry and the particle removal efficiency of the cyclone. Each design parameter was investigated by varying
the parameter from 50% to 150% of its nominal value while keeping the rest of parameters unchanged as nominal values.

3 Results and Discussion

3.1 Particulate emission comparison between gasification and pyrolysis

3.1.1 Particulate emission dynamics in terms of $PNC_{it}$

The variation of PNCs under different operating conditions is given in Figure 3. Generally, PNCs increase by 3.4 times with temperature increasing from 500 °C to 700 °C. Particles emitted from the gasification process are around 1.1 times more than particles emitted from the pyrolysis process. The particles emitted from both the gasification and pyrolysis process are mainly in the size range 0.25-1.0 µm, suggesting a potential exposure risk to intermediate-mode particles (particles with sizes ranging from 0.1 µm to 1.0 µm (Kohli and Mittal, 2015)). The results are consistent with the study by (Shiota et al., 2017) and most of the particles emitted from a gasification plant were found to be less than 3 µm.

The emission-initiating temperatures (the temperature where explicit emissions are observed) are around 200 and 400 °C for gasification and pyrolysis, respectively. The lower emission-initiating temperature for gasification than that for pyrolysis should be related to the fact that partial oxidation takes place in the gasification process with the existence of oxygen. The effects of temperature on particulate emissions were investigated by several researchers. Nam et al., (2010) studied the influence of ambient temperature on PM emissions from gasoline-powered vehicles. They found that in general, particulate emissions doubled for every 20 °F drop in ambient temperature. Nosek et al., (2014) analyzed the effects of primary combustion air temperature on the heat performance and particulate emissions of burning biomass. No obvious trend was observed on the particulate emissions with increasing primary combustion air temperature. However, to the best of our knowledge, none of them has reported the relationship between the initiation of particulate emission and temperature. The PNC profiles
generally exhibit a unimodal feature for the case of 600 °C for both gasification and pyrolysis, while the PNC profiles exhibit a bimodal feature for the cases of 500°C and 700 °C, which corresponds to two emission peaks for both the gasification and pyrolysis. At 500 °C, upon the initiation of the experiments, gasification or pyrolysis reactions take place and syngas is released together with fine particles, which leads to the first emission peak. As the reactions going on, solid organic material is converted into gaseous species and particle porosity increases gradually to such a level that further particle fragmentation takes place, which leads to the second emission peak. At 600 °C, porosity increases faster due to the relatively high temperature compared to the case of 500 °C, which causes the overlap of these two peaks. At 700 °C, temperature is high enough and thermal stress will cause further breakage of the ash in the gasification process. For pyrolysis, particle fragmentation rate is relatively slow without the existence of oxygen. Hence, in the gasification process the particles continue to be emitted at lower rate after reaching the first emission peak but in the pyrolysis process the second emission initiates after a short period. Real-time particulate emission from incineration of solid waste under high temperature (>850 °C) was monitored (Derrough et al., 2013) and a small secondary emission peak was also observed. However, no explanation has been provided for this phenomena in the study.
Figure 3. The variation of $n_{c_{i_{a}}}$ under different operating conditions. (a1: gasification, 500°C; a2: gasification, 600°C; a3: gasification, 700°C; b1: pyrolysis, 500°C; b2: pyrolysis, 600°C; b3: pyrolysis, 700°C).

3.1.2 Particle size distributions

The size-dependent average particle number and volume concentrations of different cases are given in Figure 4a and 4b, respectively. The measured PNCs including all size ranges are $1.92 \times 10^8$, $3.90 \times 10^8$, $1.09 \times 10^9$, $1.59 \times 10^8$, $1.68 \times 10^8$, $4.57 \times 10^8$ #/L for the case of 500 °C gasification, 600 °C gasification, 700 °C gasification, 500 °C pyrolysis, 600 °C pyrolysis, and 700 °C pyrolysis, respectively. The measured particle volume concentrations (PVCs) including all size ranges are $7.95 \times 10^{-12}$, $4.80 \times 10^{-10}$, $1.10 \times 10^{-9}$, $6.05 \times 10^{-11}$, $9.98 \times 10^{-11}$, $3.15 \times 10^{-10}$ m$^3$/L for the case of 500 °C gasification, 600 °C gasification, 700 °C gasification, 500 °C pyrolysis, 600 °C pyrolysis, and 700 °C pyrolysis, respectively. Since higher operating temperature favors higher chemical reaction rates, both PNCs and PVCs increase with increasing temperature in both gasification and pyrolysis processes. In addition, gasification emits more particles compared with pyrolysis, mainly due to the factor that high-speed partial oxidation reactions take place in the gasification process, which serves to convert carbon from
solid phase to gas phase and causes more significant particle fragmentation and emission. For the case of 700°C, the number concentration of particles emitted from gasification is 170%, 110%, 161%, 772% higher than that emitted from pyrolysis for the size bins of 0.25 - 0.5, 0.5 - 1.0, 1.0 - 2.5, 2.5 - 32.0 μm, respectively. The particles with sizes ranging from 0.5 to 1.0 μm account for the biggest proportion of emitted particles in terms of number concentration, which is 70.09%, 50.84%, 49.86%, 64.41%, 63.77%, 56.37% for the cases of 500°C gasification, 600°C gasification, 700°C gasification, 500°C pyrolysis, 600°C pyrolysis, and 700°C pyrolysis, respectively. However, the particles with sizes ranging from 1.0 to 2.5 μm account for the biggest proportion of emitted particles according to their size-dependent volume concentrations, which is 39.60%, 51.94%, 58.14%, 43.08%, 53.16%, 65.29% for the cases of 500°C gasification, 600°C gasification, 700°C gasification, 500°C pyrolysis, 600°C pyrolysis, and 700°C pyrolysis, respectively. These results suggest a great concern about exposure to intermediate-accumulation-mode particles (particles with sizes ranging from 0.1 μm to 2.5 μm (Kohli and Mittal, 2015)). A bimodal particle size distribution at 700°C incineration was observed and it was found that the maximum number concentration being at about 80 nm and the minor one at 40 nm (Maguhn et al., 2003). The incineration process should be able to emit particles smaller than that from gasification and pyrolysis, because it could provide enough oxygen to convert biomass to finer particles by a higher temperature. Similarly, the PM$_{2.5}$ emitted from incineration was found to consist of about 99% of submicrometer sized particles and about 65% of ultrafine particles were PM$_{0.1}$ (Buonanno et al., 2009). The normalized particle number size distributions in dN/dlogDp and particle volume size distributions in dV/dlogDp are shown in Figure S2 in the Supplementary Material.
Figure 4. a: Size-dependent average PNCs under different reaction conditions. 
b: Size-dependent average PVCs under different reaction conditions.
3.1.3 Particle deposition on human respiratory system

Figure 5 shows the results of particle number deposition modes in the human respiratory system. The particle number size distributions (PNSDs) are calculated by assuming that all the emitted particles are directly inhaled by human respiratory system. It was found that most particles could penetrate deeply into the last stage of the respiratory system, i.e., the 5th and 4th stages for submicron particles and supermicron particles, respectively. At the nasal breathing mode, particles with sizes ranging from 0.25 to 1.0 µm account for around 91%, 74%, 76%, 90%, 84%, and 79% of the total number of particles that deposit onto the last stage in the cases of 500 °C gasification, 600 °C gasification, 700 °C gasification, 500 °C pyrolysis, 600 °C pyrolysis, and 700 °C pyrolysis, respectively. At the oral breathing mode, particles with sizes ranging from 0.25 to 1.0 µm account for around 92%, 77%, 79%, 91%, 86%, and 81% of the total number of particles that deposit onto the last stage in the six cases, respectively. The trend of particle deposition through nasal and oral breathing modes is roughly the same. However, significant more submicron particles deposit onto the first stage for a nasal breathing mode than an oral breathing mode, which indirectly causes around 10.8% more submicron particles to penetrate deeply to the last stage for the oral breathing mode.

Considering that the greatest health concern is related to the deposition of particles into the deep lung system (Casse et al., 2002; Ferin et al., 1990), the nasal breathing mode could potentially serve to mitigate human exposure to aerosols. The detailed data of PNSDs in the human respiratory system is listed in Table S4 in the Supplementary Material. Table S5 lists the results of particle volume distributions in the human respiratory system. By taking into account of particle volumes, particles with sizes ranging from 1.0 to 2.5 µm account for the biggest proportion of the total volume of particles that deposit onto the last stage of human respiratory system, which is different from the results of particle number deposition. At the
nasal breathing mode, particles with sizes ranging from 1.0 to 2.5 \( \mu \text{m} \) account for around 53%, 55%, 62%, 58%, 63%, and 74% of the total volume of particles that deposit onto the last stage in the cases of 500 °C gasification, 600 °C gasification, 700 °C gasification, 500 °C pyrolysis, 600 °C pyrolysis, and 700 °C pyrolysis, respectively. At the oral breathing mode, particles with sizes ranging from 1.0 to 2.5 \( \mu \text{m} \) account for around 51%, 55%, 62%, 55%, 62%, and 73% of the total volume of particles that deposit onto the last stage in the six cases, respectively.
Figure 5. Particle deposition on human respiratory system through:
Nasal airway: a1: gasification, 500°C; a2: gasification, 600°C; a3: gasification, 700°C; a4: pyrolysis, 500°C; a5: pyrolysis, 600°C; a6: pyrolysis, 700°C;
Oral airway: b1: gasification, 500°C; b2: gasification, 600°C; b3: gasification, 700°C; b4: pyrolysis, 500°C; b5: pyrolysis, 600°C; b6: pyrolysis, 700°C.

3.2 Comparison of cyclone removal efficiency based on respiratory modelling

Particle-removal ability of cyclone was investigated based on the results of particle deposition modelling. The removal efficiency of particle number was calculated as the following:

\[
e_n = \frac{\sum_i (PN_{a1} - PN_{ci})}{\sum_i PN_{ai}}
\]  

The removal efficiency of particle volume is calculated as the following:

\[
e_v = \frac{\sum_i (PN_{ai} \times V_{ai} - PN_{ci} \times V_{ci})}{\sum_i (PN_{ai} \times V_{ai})}
\]

where \(PN_{ai} (#/min)\) is the number of particles within size range \(i\) deposited on the human respiratory system in the case without a cyclone, \(V_{ai} (m^3/#)\) is the volume of a particle within size range \(i\) deposited on the human respiratory system in the case without a cyclone, \(PN_{ci} (#/min)\) is the number of particles within size range \(i\) deposited on the human respiratory system after the exhaust gas passing through a cyclone, \(V_{ci} (m^3/#)\) is the volume of a particle within size range \(i\) deposited on the human respiratory system after the exhaust gas passing through a cyclone. The particle removal efficiencies in different stages of human respiratory system were calculated by assuming a flowrate of 52 L/min and the results are shown in Figure 6. Compared with submicron particles, supermicron particles could be more effectively removed by the cyclones. In all the cases, the removal efficiency of submicron particle number reaches its peak at the last stage of human respiratory system, while the removal efficiency of supermicron particle number reaches its peak at the stage of Bronchial
airways. At each stage in the respiratory system, the removal efficiency of particle number decreases with the cyclone diameter ($D$) increasing from 20 mm to 45 mm. At the nasal breathing mode, the average removal efficiency of submicron particle number among all the stages of human respiratory system is 15.16%, 4.41%, 1.90%, 1.07%, 0.70% and 0.51% by cyclone C-20, C-25, C-30, C-35, C-40 and C-45, respectively, in the gasification process. The average removal efficiency of supermicron particle number among all the stages of human respiratory system is 90.13%, 68.92%, 47.56%, 28.93%, 15.65% and 8.09% by cyclone C-20, C-25, C-30, C-35, C-40 and C-45, respectively, in the gasification process. The average removal efficiency of submicron particle number among all the stages of human respiratory system is 15.83%, 4.54%, 1.94%, 1.08%, 0.71%, 0.51% by cyclone C-20, C-25, C-30, C-35, C-40 and C-45, respectively, in the pyrolysis process. The average removal efficiency of supermicron particle number among all the stages of human respiratory system is 86.35%, 58.60%, 34.71%, 18.23%, 9.07%, 4.80% by cyclone C-20, C-25, C-30, C-35, C-40 and C-45, respectively, in the pyrolysis process. Similar trends could be observed in Figure S3, which shows the removal efficiencies of particle volume in each stage of the human respiratory system as a function of cyclone diameter.
We present in Figure 7 the removal efficiencies of particle number for different cyclones as a function of the flow rate. Although the total reduced number of submicron particles associated with the gasification process is double that associated with the pyrolysis process, the removal efficiencies of particle number for different cyclones show the similar trends in the cases of both gasification and pyrolysis. For the gasification process, with the flow rate increasing from 24 to 116 L/min, the removal efficiency of particle number increases from 10.64%, 3.60%, 1.23%, 0.54%, 0.31% and 0.21% to 80.01%, 56.33%, 35.94%, 23.41%, 15.54% and 10.17% for cyclone C-20, C-25, C-30, C-35, C-40 and C-45, respectively. For the pyrolysis process, with the flow rate increasing from 24 to 116 L/min, the removal efficiency of particle number increases from 7.39%, 2.17%, 0.80%, 0.41%, 0.26% and 0.18% to 79.59%, 56.51%, 34.19%, 20.27%, 12.18% and 7.35% for cyclone C-20, C-25, C-30, C-35, C-40 and C-45, respectively. For the practical applications, C-20 has the best particle removal ability. The results suggest that increasing flowrate has a positive effect on the particle removal efficiency of the cyclone, while increasing cyclone diameter has a negative effect on the particle removal efficiency of the cyclone. Similar results were found by (Sagot et al., 2017). An increase of the particle removal efficiency with the increase of the flow rate was attributed to the stronger centrifugal force created by the higher flowrates. A reduction of the particle removal efficiency with the increase of the cyclone diameter was because of the fact that reducing
cyclone diameter could lead to an increase of the internal velocity, thus producing higher centrifugal forces and improving the particle removal efficiency. The removal efficiencies for submicron and supermicron particles were plotted separately in Figure S4 in the Supplementary Material. The removal efficiencies for supermicron particles are higher compared to submicron particles because removal efficiency generally increases with increasing particle size, which is also inherent to the calculation of cyclone efficiency.

Figure 7. Removal efficiency of particle number of different cyclones under different air flowrate. (a: gasification, b: pyrolysis).

3.3 Sensitivity analysis

The effects of design parameters on the removal efficiency of particle number are shown in Figure 8. Both gasification and pyrolysis show the similar trends of the removal efficiency of particle number under different design parameters of the cyclone. For the gasification process, the removal efficiency of submicron particles decreases from 67.49%, 48.25%, 46.70%, and 24.77% to 3.35%, 7.06%, 6.35%, and 10.95%, as X/X₀ increases from 0.5 to 1.5 for the cases of D₀/D, H/D, W/D and S/D, respectively, which means that increasing D₀, H, W and S has a negative effect on the removal efficiency. Although the removal efficiency of submicron particles increases from 15.25%, 12.98% and 16.94% to 16.91%, 18.90%, and 16.94%, as X/X₀ increases from 0.5 to 1.5, for L₀/D, L_c/D, and L₀/D, respectively, L_b, L_c, and L₀ have
negligible effect on the cyclone removal efficiency, as they generally fall along the horizontal line. Hence, the variation of $L_b$, $L_c$, and $L_0$ may have a limited effect on changing cyclone removal efficiency. The similar trends could be observed in the removal efficiencies of supermicron particles under different design parameters. However, the removal efficiency of supermicron particles is significantly higher than that of submicron particles. Among these seven parameters, the particle removal efficiency is found to be most sensitive to $D_0$, which suggests that the most effective way of modifying the cyclone design is on the change of $D_0$. Similarly, reducing $D_0$ has the most significantly positive effect on reducing particle volume deposition in the human respiratory system, as shown in Figure S5 in the Supplementary Material. In addition, as the value of $D_0/D$ decreases, the slope of $D_0/D$ tends to remain constant for submicron particles, while the slope of $D_0/D$ tends to increase for supermicron particles. The results show that there is still a large potential of removing submicron particles from the system by decreasing $D_0/D$. However, decreasing $D_0/D$ would reach its limit of reducing deposited supermicron particles because the removal efficiency of supermicron particles is approaching 100%.
4 Conclusions

In this work, we characterized the particulate emission potential of the gasification and pyrolysis process. We compared the particle removal efficiencies of existing cyclones with different configurations based on particle respiratory deposition. Sensitivity analysis was conducted to identify the most effective design parameters. Generally, PNCs increase by 3.4 times with temperature increasing from 500 °C to 700 °C. Particles emitted from the gasification process are around 1.1 times more than particles emitted from the pyrolysis process. The particles emitted from both gasification and pyrolysis process are mainly particles within the size range 0.25-1.0 µm and particles within the size range 1.0-2.5 µm. The emission-initiating temperatures for gasification and pyrolysis were found to be around 200 and 400 °C, respectively. The PNC profiles generally exhibit a single mode feature for the case of 600 °C for both gasification and pyrolysis, while the PNC profiles exhibit a bimodal feature for the case of 500 and 700 °C, respectively. Particle respiratory deposition modelling showed that most particles penetrate deeply into the last stage of the respiratory system, i.e., the 5th and 4th stages for submicron particles and supermicron particles, respectively. To minimize total particle number deposited onto the human respiratory system, gas cyclone diameter \( D \) should be small. Sensitivity analysis showed that the cyclone collection efficiency was found to be most sensitive to \( D_0 \), which suggests that the most effective way of modifying cyclone design would be the one associated with changing \( D_0 \). The results from this study could not only provide information for setting up the emission standards for
gasifiers, but also serve as the basis for controlling the particulate pollution from gasification and pyrolysis technologies.

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**Appendix A. Supplementary Material**

Additional Data could be found in the Supplementary Material.

**References**


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• Particulate emissions of gasification and pyrolysis are compared.

• The particles from gasification and pyrolysis are mainly $\text{PM}_{0.25-1.0}$ and $\text{PM}_{1.0-2.5}$.

• Most particles penetrate deeply into the last stage of the respiratory system.

• A particle respiratory deposition-based cyclone design scheme is proposed.

• The cyclone vortex finder diameter is the most sensitive design parameter.