Measurement of Residence Time Distributions Of Coal Particles in a Pressurized Fluidized Bed Gasifier (PFBG) using Radio Tracer Technique

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Abstract

A pressurized fluidized bed gasifier (PFBG) system of an integrated coal gasification and combined cycle (ICGCC) plant is designed to behave as a well-mixed flow system for coal; and any deviation from the well-mixed flow condition will deteriorate the performance and efficiency of the gasification system. This paper describes a radiotracer investigation carried out to measure RTDs of coal particles in a pilot-scale PFBG with objectives to determine mean residence time (MRT) of coal/ash particles in the gasifier and estimate degree of mixing at different operating and process conditions. Lanthanum-140 labeled coal (100 gm) was used as a radiotracer. The tracer was instantaneously injected into the coal feed line and monitored at ash and gas outlets of the gasifier using collimated scintillation detectors. The measured RTD was used to determine mean residence time (MRT) of coal particles within the system and simulated using fractional tank-in-series model. The results of simulation indicated that the system behaved as a well-mixed system with undesired bypassing of a small fraction coal particle from the system. The results of the study were used to improve the design of the gasifier and optimize the system.

Keywords: Pressurized fluidized bed gasifier, residence time distribution, radiotracer, Lanthanum-140, tanks-in-series model, bypassing

1. Introduction

Integrated coal gasification and combined cycle (ICGCC) is one of the most promising advanced clean coal technologies wherein coal is converted into low calorific value (CV) gaseous fuel in a pressurized fluidized bed gasifier (PFBG) and combusted in a gas turbine combustor of combined cycle plant. The gasifier involves flow of two different phases i.e. solid (coal) and gas (mixture of air and steam) and knowledge of dynamics of these two phases is important to assess the performance of the system as well as for scale up of the process. The concept of measurement and analysis of residence time distribution (RTD) is often used to investigate dynamics of flowing phases in industrial process system [1]. The analysis provides vital information such as mean residence time (MRT) of process fluid, degree of axial mixing and abnormality in flow behavior, if any. Radiotracer techniques are widely used to measure RTD of process material in pilot-scale as well as the full scale industrial systems because of their high detection sensitivity, online detection, physicochemical compatibility, availability of wide range of suitable tracers, limited memory effect and utility in harsh industrial environment [2,3,4,5,6,7].

The Research and Development Division of M/s Bharat Heavy Electricals Limited, Hyderabad, India has designed, fabricated and commissioned a pilot-scale of the PFBG system to study the feasibility of coal gasification/combustion process for power generation. It was required to investigate flow dynamics of coal particles in the system to evaluate its performance and scale up of the process. Pant et al. [8] carried out a series of radiotracer investigations in this system to evaluate feasibility of using radiotracer technique for measurement of RTD of the coal particles in the PFBG at cold as well as hot conditions [8]. They reported use of two different radiotracers i.e. gold-198 and lanthanum-140 and concluded that both are equally suitable to trace the coal particles in gasifiers. They used fractional tanks-in-series model to simulate the measured RTD and found that the system behaved as a well-mixed system with bypassing of some of the coal particles from the system. The paper describes some of the preliminary results of the RTD measurement carried out in the same PFBG system as described by Pant et al. (2009).

2. Experimental

2.1. Pressurized fluidized bed gasifier (PFBG)

The schematic diagram of the pilot-scale PFBG system is shown in Fig.1 [8]. The system consists of various subsystems such as gasifier, coal feeding system, combustor, air compressor, steam supply system, gravity recycle system, gas cleaning and cooling system and ash extraction system. The gasifier is designed for gasifying 50 kg/hr of sub bituminous coal has an internal diameter of 200 mm and consists of air plenum, distributor assembly and freeboard section. An air compressor supplies the fluidizing air required for the process. The steam required for the process is supplied by a steam generating system and passed to the fluidizing bed through fluidizing air supply line. The combustor assembly is directly coupled with the air plenum of the gasifier. The gasifier is designed to operate at 3 atmospheric pressure and 1000°C temperature. The air-plenum acts as a...
header for the fluidizing media i.e. air/steam and also distribute the same uniformly into the gasifier by means of a conical distributor attached to it. The freeboard section is slightly conical with 200 mm diameter at the bottom and 250 mm diameter at the top. Gasifier and free board sections were provided with a number of view ports and tappings for temperature and pressure measurements.

Initially the gasifier is filled with a known quantity of coal particles (50 microns-4 mm). Subsequently, coal and fluidizing air/steam are fed to the gasifier through the respective feeding systems. During the fluidization process, the gasification and combustion of coal occurs and various gases such as carbon dioxide, carbon monoxide, hydrogen, methane etc. are produced. During the combustion process, the temperature of the fluidized bed ranges from 900-1000 °C. The mixture of gases flows upward in the freeboard section of the gasifier and passes through the cyclone system, where the fine coal particles are separated. The separated fines are fed back to the gasifier using recycle system while the gaseous mixture is fed to the wet gas cleaning system. The cleaned gaseous mixture is used as a fuel gas for power generation and various other applications.

The burnt coal i.e ash is generated in the process is extracted from the bottom of the gasifier at regular intervals.

Properties of the coal used as fluidizing material are given in Table 1. The fluidization phenomenon of gas-solid systems depends very much on the particle characteristics. Geldart [9] was the first to classify the behavior of the solids fluidized by gases into four distinct groups, namely A, B, C and D, characterized by the density difference between the particle and the fluidizing medium and mean particle size. According to the Geldart classification of fluidizing particles, coarser coal particles are classified as Type D particles, whereas the fine coal particle can be classified as Group B particles.

Table 1. Properties of coal particles used in experiments

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bulk Density</td>
<td>815 Kg/m³</td>
</tr>
<tr>
<td>Particle density</td>
<td>1680 Kg/m³</td>
</tr>
<tr>
<td>Average particle size</td>
<td>1.86 mm</td>
</tr>
<tr>
<td>Composition of coal Content</td>
<td>Caron (C): 38.3% wt,</td>
</tr>
<tr>
<td></td>
<td>Hydrogen (H₂): 2.4 % wt,</td>
</tr>
<tr>
<td></td>
<td>Sulphur (S): 0.3 % wt</td>
</tr>
<tr>
<td></td>
<td>Nitrogen (N₂): 0.71 % wt,</td>
</tr>
<tr>
<td></td>
<td>Oxygen (O₂): 9.6% wt.</td>
</tr>
<tr>
<td>Type of particle according to Geldart Classification</td>
<td>Group D</td>
</tr>
</tbody>
</table>

2.2. Residence time distribution measurements

Radiotracer technique was employed to measure the RTD of the coal in the gasifier. The technique involves the instantaneous injection of a suitable radiotracer into the process stream and monitoring its movement at strategically important locations, using scintillation detectors. A series of four different RTD runs were carried out at different process and operating conditions as shown in Table 2. All the experiments were carried out at atmospheric pressure. The temperature of the bed during the hot run ranged from 900-1000 °C. For the present study, lanthanum-140 radioisotope (gamma energies: 1.16 (95%), 0.92 (10%), 0.82 (27%), 2.54 (4%), half-life: 40 hours) as lanthanum chloride was selected to be used as a tracer, as it has strong affinity to get adsorbed on solid particles. Lanthanum-140 was produced by irradiating lanthanum oxide powder (La₂O₃) in DHURVA reactor at Bhabha Atomic Research Centre, Trombay Mumbai. The irradiated target was processed to produce lanthanum chloride (LaCl₃). About 1 mCi (37 MBq) activity of lanthanum-140 was taken from mother solution and diluted in about 300 ml of distilled water. About 100 gm of coal was poured into the diluted solution and stirred for about 5 minutes using a glass rod. The coal soaked in the radioactive solution was left for about half an hour. After half an hour, the radioactive solution was decanned and the coal particles were dried using an electrical heater. Based on the previous studies, it was observed that more than 60-70% of the initial lanthanum activity gets adsorbed on the coal particles. So it was assumed that about 600-700 micro curie (220-260 MBq) activity might have got adsorbed on the coal particles.
(100 gms) and was used as tracer in each run. The lanthanum-140 labeled coal was instantaneously injected into the coal feed line using a specially fabricated injection arrangement. The injection point is shown in Fig. 1. The tracer was injected after the system was stabilized and attained a steady state condition. The movement of the tracer in the gasifier was monitored using seven different collimated scintillation detectors (D1 to D7) mounted at four different locations as shown in the Fig.1. The detectors were connected to corresponding channels of a common computer controlled data acquisition system (DAS) set to record tracer concentration data at an interval of every one minute. The tracer concentration versus time data acquired was saved in the computer for further analysis.

Table 2. Operating and process conditions during experiments

<table>
<thead>
<tr>
<th>Run No.</th>
<th>QFeed (kg/hr)</th>
<th>QEExtraction (kg/hr)</th>
<th>HBed (m)</th>
<th>WBed (kg)</th>
<th>QAir (kg/hr)</th>
<th>QSteam (kg/hr)</th>
<th>T(oC)</th>
<th>t, (min)</th>
<th>Et (Min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>15.2</td>
<td>13.3</td>
<td>0.17</td>
<td>6.3</td>
<td>115</td>
<td>0</td>
<td>Ambient</td>
<td>30</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>7.6</td>
<td>7.2</td>
<td>0.2</td>
<td>7.6</td>
<td>115</td>
<td>0</td>
<td>Ambient</td>
<td>60</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>10.2</td>
<td>8.7</td>
<td>0.2</td>
<td>7.6</td>
<td>115</td>
<td>0</td>
<td>Ambient</td>
<td>45</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>23.3</td>
<td>7.2</td>
<td>0.2</td>
<td>12.2</td>
<td>55</td>
<td>2</td>
<td>900-1000</td>
<td>30</td>
<td></td>
</tr>
</tbody>
</table>

3. Data Analysis

The radiotracer concentration is measured in terms of counts per unit time and measured acer concentration versus time data were treated and analyzed using Residence Time Distribution software provided by International Atomic Energy Agency Vienna, Austria [10]. The normalized RTD curve was obtained using the following relation:

$$E(t) = \frac{C(t)}{\int_{0}^{\infty} C(t)dt}$$

(1)

The area under the normalized RTD function ($E(t)$) is equal to unity. Thus:

$$\int_{0}^{\infty} E(t)dt = 1$$

(2)

The treated and normalized RTD curves measured at the bottom of the gasifier during four different runs are shown in Fig. 2.

The first moment of the RTD curve was determined using the following relation:

$$\bar{t}_E = \frac{\int_{0}^{\infty} t.C(t).dt}{\int_{0}^{\infty} C(t).dt}$$

(3)

The first moment of the RTD curve gives MRT of the process material in the system. The MRTs determined from measured RTD curves for four different runs are shown in Table 3. The theoretical MRT ($\bar{t}_T$) of the material in a closed system is given as:

$$\bar{t}_T = \frac{M}{Q}$$

(4)

Where $M$: weight of bed material and $Q$: flow rate. The above equation holds good for systems with closed-closed boundaries. The freeboard section of the present experimental setup is a system with 'open-open' boundaries and thus above equation can be applied. Whereas, the fluidized bed section of the system can be approximated as 'closed-open system' and the above equation holds good for it and was used to estimate the approximate theoretical mean residence time of coarser coal particles flowing out from bottom of the system. However, the exact mean residence time is determined from residence time distribution measurement using radiotracer technique. The values of the $\bar{t}_T$ were calculated based on bed-weight and corresponding flow rates and are given in Table 2.

Table 3. Results of model simulation of residence time distribution data

<table>
<thead>
<tr>
<th>Run No.</th>
<th>QFeed (kg/hr)</th>
<th>$\bar{t}_T$ (Min)</th>
<th>$\bar{t}_E$ (Min)</th>
<th>Fractional tanks-in-series model</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>15.2</td>
<td>30.0</td>
<td>31.7</td>
<td>30.0</td>
</tr>
<tr>
<td>2</td>
<td>7.6</td>
<td>60.0</td>
<td>57.0</td>
<td>57.0</td>
</tr>
<tr>
<td>3</td>
<td>10.2</td>
<td>45.0</td>
<td>42.0</td>
<td>38.0</td>
</tr>
<tr>
<td>4</td>
<td>23.3</td>
<td>30.0</td>
<td>34.0</td>
<td>34.0</td>
</tr>
</tbody>
</table>

In order to quantify the degree of mixing and investigate the flow behavior of the gasifier, suitable mathematical models are to be used to simulate the experimentally measured RTD data. Based on the shape of the RTD curve, configuration of the gasifier and prior information available about the system, tanks-in-series was selected to simulate the experimentally obtained RTD data [11]. This model assumes that the system under investigation consists of a series of well-mixed stirred tanks each of volume $V$. The total volume of the system will be $NV$, where $N$ is number of well-mixed tanks connected in series. The physical representation of the model is shown in Fig. 2.
is called gamma function and is defined as:

\[ \Gamma(N) = \int_0^\infty e^{-x} x^{N-1} dx \]  

where \( x = V_f/V \) and \( N \) is positive and need not to be an integer. The above model is an extension of tanks-in-series model, where \( N \) need not to be an integer and is treated as an adjustable index of mixing performance. The main use of this model is to fit small deviations from the exponential distribution of a single stirred tank. If \( N < 1 \), then this implies that the system behaves as a well-mixed system with some amount of bypassing or short-circuiting of the process fluid. The tanks-in-series model has the advantage of mathematical simplicity but the parameter lacks a physical interpretation, except when it is an integer. The RTD functions of tanks-in-series model for different values of tank numbers are shown in Fig. 3.

![Fig.3. Impulse responses of gamma function model for different tank no.](image)

The model simulated RTD curves were fitted to the experimental data using least square curve-fitting method and optimum values of the model parameters corresponding to best fit were obtained [12]. The quality of the fit is judged by choosing the model parameters to minimize the sum of the squares of the differences between the experimental, \( E(t) \) and model simulated or predicted curves, \( E_m(t, N, \tau_m) \). Thus root mean square (RMS) value is given as:

\[ \text{RMS} = \sqrt{\frac{1}{n} \sum (E_m(t, N, \tau_m) - E(t))^2} \]
\[
\text{RMS} = \left( \frac{1}{n} \int_{0}^{\infty} [E(t) - E_m(t, N, \tau_m)]^2 dt \right)^{0.5} = \text{Minimum}
\]

where, RMS is root mean square \(n\) is number of data points. The comparison of experimental and model simulated RTD curves obtained from experimental run 1 to run 4 are shown in Fig. 5 to Fig. 8.

4. Results and Discussion

In tracer test carried out at cold condition it was observed that, the tracer concentration curves were recorded by detectors D3 and D4 mounted at the inlet and outlet of the cyclone during cold conditions and it indicates an instantaneous increase in tracer concentration soon after injection and subsequently become constant. The increase in background levels at inlet and outlet of the cyclone could be due to residual tracer particles in the injection system and getting detected by detector D3 and D4. However, at hot condition i.e. temperature > 900 °C (run 4), the increase in background levels recorded by detector D3 and D4 were more than that of the cold runs. This could be due to generation of fine coal particles at hot conditions and are accumulated at the bottom outlet of the cyclone thus further increasing the background at detector D3 and D4.

The tracer concentration curves monitored by detector D1 at the coal feed inlet and by detector D2 at the distributor outlet were considered for detailed flow analysis. The tracer concentration curve monitored by detector D1 is a sharp pulse of narrow width in comparison to the curve monitored at distributor outlet by detector D2 and could be considered as an impulse for analysis. The normalized tracer concentration curves measured at the distributor outlet (D2) during the four different runs are shown in Fig. 4. Each measured RTD curve was simulated using tanks-in-series model and it was observed that the model predicted response does not fit well to the experimental data. This indicated that the simple tanks-in-series model with \(N=1\) is not a suitable model to describe the flow behavior of the coal in the gasifier. Thus an extension of tanks-in-series model as described above was used to simulate the measured RTD data and the model was found suitable to describe the dynamics of the coal particles in the gasifier. The model predicted RTD corresponding to the minimum RMS and experimentally measured RTD curves are shown in Fig. 5 to Fig. 8. The model parameters i.e. tanks number (\(N\)) and mean residence time (\(\tau_m\)) corresponding to best fit i.e. minimum RMS value are tabulated in Table 3.
5. Conclusions

Radiotracer technique was successfully applied to measure MRTs and investigate flow dynamics of coal particles in a pilot-scale PFBG. The values of the measured MRTs are close to the theoretical and model simulated MRTs.

An extension of tanks-in-series model (gamma distribution model) was used to simulate the measured RTD curves and was found suitable to describe the dynamics of coal in the gasifier. The values of model parameter i.e. tank number were found to be less than 1 (N<1) indicating bypassing or short-circulating of coal in the gasifier at all the operating and process conditions used in the experiments. The gasifier behaved as a well-mixed system with a small fraction of coal particles bypassing the system, which is highly undesired. The results of the study were used to improve the design of the system; and optimize and scale up of the process.

Acknowledgements

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6. References

7. Nomenclature

A: Amount of radiotracer (MBq)

\( t \): time variable (second)

\( C(t) \): Radiotracer concentration (Counts/unit time)

\( E(t) \): Experimental RTD curve (second\(^{-1}\))

\( E_m(t, \tau_m, N) \): Theoretical RTD curve (second\(^{-1}\))

\( \bar{E}_E \): Experimental mean residence time (second\(^{-1}\))

\( \tau_m \): Model predicted mean residence time (Min)

\( \bar{E}_T \): Theoretical mean residence time (second\(^{-1}\))

T: Temperature (\( ^\circ \)C)

M: Weight of bed material (kg)

N: Number of tanks

N: Data points

V: Volume of tank (m\(^3\))

\( V_f \): Volume of fractional tank (m\(^3\))

x: Fraction of volume (m\(^3\))

Q: Flow rate (kg/hr)

\( Q_{\text{Feed}} \): Feed flow rate of coal particles (kg/hr)

\( Q_{\text{Extraction}} \): Extraction flow rate of coal particles (kg/hr)

\( Q_{\text{Air}} \): Flow rate of air (kg/hr)

\( Q_{\text{Steam}} \): Flow rate of air steam (kg/hr)

\( H_{\text{Bed}} \): Bed height (m)

\( W_{\text{Bed}} \): Bed weight (kg)

8. Greek Symbols

\( \tau_m \): Model predicted mean residence time (Min)

\( \Gamma \): Gamma function