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Review article

Coal char temperature profile estimation using optical reflectance for a commercial-scale Sasol-Lurgi FBDB gasifier

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ABSTRACT

In the Sasol-Lurgi fixed-bed dry-bottom (FBDB) gasifier the temperature in the combustion zone should not exceed the melting point of the ash-forming minerals, causing them to melt/flow and agglomerate. Sintering of ash particles is considered desirable in Sasol-Lurgi FBDB gasification, since it promotes easy gas flow, whereas clinkering creates channeling and localized "hot spots", leading to unstable gasifier operation. Due to the counter-current mode of operation, hot ash exchanges heat with the cold incoming agent (steam and oxygen), while at the same time hot raw gas exchanges heat with cold incoming coal. This results in the ash and raw gas leaving the gasifier at relatively low temperatures compared to other types of gasifiers, which improves the thermal efficiency and lowers the steam consumption.

Vitrinite reflectance analyses were performed on a range of Sasol-Lurgi MK IV commercial-scale gasifier turn-out samples, applying ISO standards 7404-5. Average temperature profile measurements of the solid particles, successfully revealed the temperature range occurring within the various zones of the gasifier. The average (mean) temperature ranged from ca. 400 °C up to 850 °C within the pyrolysis region. In this region of the gasifier, the particle surface temperature and peak temperature showed visible evidence of heat transfer limitations occurring through lump coal when compared to the mean particle temperature. This provides some evidence of the complex radial and localized behaviour occurring within the averaged axial sample slices. In the oxidizing and combustion regimes, exothermic conditions prevail and heat transfer differences across the particles are minimized. A characteristic spike, indicative of an increase in temperature, was found in the sample taken directly above the ash-grate, seeming to indicate that agent distribution through the nozzles positioned just above the grate is not uniform, resulting in localized oxygen concentration increases with subsequent "hot-spots" and channel-burning occurring. Homogenization of the ash bed could help to optimize the agent distribution within the reactor. The surface temperature profile of the gasifier solids was thus found to be in reasonable agreement with literature, albeit that different coal types and temperature profile estimation methods were utilized.

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1. Background and introduction

Lump sized coal is used by Sasol as a feedstock to produce synthesis gas via the Sasol-Lurgi fixed-bed dry-bottom (FBDB) gasification process. Once the coal is mined, it is crushed down to less than 100 mm (typical top-size of about 65 mm) and screened to a bottom size of 5–8 mm. The coal enters the top of the gasifier through a lock-hopper system, while reactant gases (steam and oxygen) are introduced at the bottom of the gasifier. The reactant gases flow at a relatively low velocity upwards through the voids between the coal lumps. The counter-current operation results in a temperature drop in the reactor, with the result that four characteristic zones (Fig. 1) can be identified in a fixed-bed gasifier [1–4].

As the coal descends, it is first dried and de-volatilized by the heat of the rising gas. The de-volatilized coal, better known as char, then enters a gasification zone and residual char is finally burnt to ash. Ash is removed at the bottom of the gasifier by a rotating grate and lock- hopper (see Fig. 1).

Sintering of ash particles is considered desirable in Sasol-Lurgi FBDB gasification, since it promotes easy gas flow, whereas clinkering creates channeling and localized "hot spots", leading to unstable gasifier operations [5]. Encapsulation of carbon has been observed in clinkers using continuous coal scanning electron microscopy (CCSEM), giving rise to a lower carbon efficiency during gasification. The sintering and encapsulation processes are still poorly understood, requiring much further research effort, which is being investigated [6].

From an ash melting characteristic perspective, a major optimization constraint is that the Sasol-Lurgi FBDB gasification process is governed to a large extent by operating the reactor temperature below the ash fusion characteristics of the feed coal. This temperature control is via excess steam addition, which negatively impacts on the water gas shift reaction, producing carbon dioxide and hydrogen.

A study by Van Dyk [7], investigated to determine whether increasing the ash fusion temperature during gasification can be conducted with the use of additives. By increasing the ash fusion temperature, the direct effect will be that steam consumption can be decreased, which in turn will improve carbon utilization.

The solid and gas temperatures present in the fixed-bed gasifier are thus of importance in the operating and control philosophy of the gasification process. A schematic representation of the solid and gas temperature profiles of a fixed-bed gasifier is given in Fig. 2 [8,9]. It will be attempted in this study to measure the solids' temperature profile occurring within the Sasol-Lurgi FBDB Mark IV gasifier from samples extracted from a gasifier turn-out campaign [4].

Sasol currently operates more than 80 MK IV gasifiers. These units can truly be seen as the "work-horses" of syngas production from coal. The demand for synthesis gas at Sasol has increased steadily over the years, resulting in continuous pressure to increase production rates of individual units [10].

In an attempt to better understand the gasification reactor within the context of the internal processes, and thereby determine an optimization path of commercial-scale Sasol-Lurgi FBDB gasifiers Glover et al. [11] conducted several gasifier dissection sampling campaigns. This work focused predominantly on quantifying the carbon behaviour during gasification. With relevance to the ash bed zone it was postulated that where thermal-channeling was observed, local steam to oxygen ratio variations may occur, possibly allowing for higher temperatures to be reached above ash melting, giving rise to clinker formation in certain areas of the gasifier.



Fig. 1. Schematic representation of a fixed-bed gasifier showing the four zones of reactivity (after hebden and stroud, 1981).



Fig. 2. Schematic representation of a fixed or moving-bed gasifier showing solid and gas temperature profiles occuring within the reactor [8,9].

Based on char optical reflectivity (reflectivity increases with increased temperature measured by means of infra-red spectroscopic techniques), calibration curves were constructed by heating samples in inert atmospheres at different temperatures and measuring the resultant changes in reflectivity [11]. The reflectivity was found to decrease in gasified chars, most likely due to a development of pore-structure. The temperature data obtained was limited to areas in the reactor where no appreciable gasification or combustion reactions took place. The two possible temperature profiles that could be constructed (Fig. 3) indicated temperature ranges of between 200 °C and 575 °C in the coal distributor, while being relatively uniform (600 °C) in the main bed (top section). The average bed temperatures in the top section of the main bed were around 777 °C, peaking to 1026 °C a short distance into the ash-zone, with a cooler zone anticipated closer to the jacket [11]. The top of the ash bed was believed to have a characteristic "W" shape which represents deviation from plug-flow.

The Glover et al. [11] findings were highly significant and a major conclusion from this work was that particle size measurements clearly showed a radial pattern, i.e. a lower fines content in the centre and at the edge of the vessel. This strongly suggested a radial distribution of permeability, and hence a radial distribution of flow; the higher flows being at the centre and near the wall. DRIFT (diffuse-reflectance infrared Fourier transform) spectra obtained by Glover et al. [11] revealed that a temperature profile, consistent with the highest flows in the centre and wall regions of the vessel existed, therefore better reaction occurred in these regions.

Krishnudu et al. [12,13] studied a 1.13 m fixed-bed pilot scale reactor in India, quenched by cutting off the supply of oxygen and steam. Sequential samples were collected by running-out the fuel bed through the ash discharge grate and a significant change in average particle size and voidage along the reactor axis was recorded. The extent of the different reaction zones in the gasifier was also revealed (Fig. 4).

The thermal history of coal chars obtained from a fixed bed gasifier is of importance in understanding gasifier performance and behaviour. Several methods of assessing thermal history have been considered [14]. The results show that Raman spectroscopy is a good method for estimating the heat treatment temperature in a thermally homogeneous coal char sample, whereas reflectance measurements provide a rapid means of characterizing thermal heterogeneity of the char samples in the temperature range 400–1000 °C. The advantages of the techniques over other char



Fig. 3. Graph showing the temperature profile in the Sasol-Lurgi MK III gasifier. temperatures are indicated in kelvin. the position of the ash bed is shown by the dotted line [11].



Fig. 4. Graph showing the predicted temperature profile of the indian pilot scale gasifier [13].

characterization methods are discussed. The use of residual volatiles yield as a temperature indicator for the char samples is questionable [14] since some of the samples taken from the gasifier are heterogeneous in nature because of mixing and flow characteristics. In this circumstance, the average value of a parameter can be misleading as the volatile matter changes only between 400– 1000 °C. Therefore additional techniques are needed which are capable of providing further information on the thermal profiles up to 2000 °C in the gasifier.

Based on these axial particle size and bed voidage results, a partial ash segregation model was proposed [15]. A feature of this model is the advantage than an extended "hot-spot" region (Fig. 4) could be predicted in the axial direction compared with the sharp peak usually predicted by total ash segregation and shell progressive models (radial and axial positions) [16].

2. Objective of this study

The primary focus of this study is to evaluate the proposed sequential (axial) sampling "turn-out" methodology, as proposed by Krishnudu et al. [12,13], of the entire quenched Sasol-Lurgi FBDB MK IV gasifier, in order to obtain samples to accurately describe operational aspects occurring in the reaction zones within the reactor. In this paper, the commercial-scale Sasol-Lurgi FBDB MK IV solids' temperature profile results (obtained by optical reflectance measurements of the coal/char/ash samples) will be presented and discussed together with the earlier findings of Krishnudu et al. [12,13] and Glover et al. [11].

3. Experimental

3.1. Gasifier sampling methodology

A sampling plan was required for the gasifier turn-out exercise with the number of samples required, being calculated according to the specifications of a statistical model. In order to determine the temperature profile, as well as other profiles of the properties of interest, it was necessary to collect samples at different levels across the length of the gasifier. In this case, a minimum sample increment size was calculated, using the statistical design-expert software.

After, de-pressurizing the gasifier by cutting off the supply of oxygen and steam, followed by water quenching, the ash-grate speed of the reactor was controlled at minimal revolutions per hour whilst discharging the gasifier contents. This gasifier sampling procedure was similar to the method proposed earlier by Krishnudu et al. [12,13].

Each sample was further sub-sampled and contained in 200 L drums for characterization purposes. Bunt and Waanders [17] used these samples to identify the reaction zones present in the Sasol-Lurgi FBDB gasifier by means of the proximate analysis and coal/ char CO_2 reactivity measurements. This paper deals with the reflectance of vitrinite technique which was used to estimate the temperature profile of the coal/char and ash samples and will be discussed in the context of the reaction zones in the reactor.

3.2. Analytical characterization methods utilized

Each of the turn-out samples obtained were thoroughly homogenized after measuring the bulk density, riffled and split into equal quarters. Two of the representative quarters, in each case, were combined and stage-crushed to below 1 mm for analytical purposes; the other two quarters were combined for particle size determination.

3.2.1. Temperature estimation using optical reflectance

The objective of analyzing the gasifier turn-out samples was to work towards understanding the behaviour of the feed coal inside the gasifier, by considering the carbon conversion process and clarifying temperature profiles, applying petrographic techniques. Vitrinite reflectance was used for temperature estimation, with specific objectives:

- (1) preparation of a range of samples from the gasifier feed sample using a temperature treatment in a standard TGA,
- (2) determining the reflectance profiles for the controlled heattreated samples, applying standard petrographic techniques,
- (3) preparing a correlation chart of reflectance results against sample temperature to be used as a reference for the gasifier turn-out samples,
- (4) determination of the mean and maximum reflectance readings for the gasifier turn-out samples,
- (5) using the reflectance and temperature correlation to estimate the temperatures experienced by the turn-out sample, and
- (6) to correlate these petrographic findings to particle type analysis (char morphology). The char morphology results will not be discussed in this paper.

3.2.1.1. Preparation of reflectance/temperature calibration curve. The feed coal sample entering the top of the gasifier was crushed to -1 mm and eight 150 µl platinum sample pans were filled with a representative portion of the -1 mm prepared sample and loaded into a calibrated Mettler TGA. Each sample was initially treated in a nitrogen atmosphere at 30 °C for 3 min to obtain equilibrium, there after the temperature was ramped at 50 °C/min to a specific temperature. Once the designated temperature was reached, the sample was kept at the specific temperature in a nitrogen atmosphere for 10 min and subsequently cooled to room temperature. This procedure was repeated on the samples for temperatures ranging between 350 °C and 1200 °C, obtaining eight different samples at different temperatures. The individual samples were then mounted in resin and the blocks polished as for petrographic analysis (ISO 7404-2).

Vitrinite reflectance analysis (mean RoV%) was performed on the individual samples as per petrographic standard (ISO 7404-5). The results were depicted graphically against the treatment temperatures to serve as a calibration curve for all the other samples. The reflectance results obtained from the gasifier turn-out samples were compared to the temperature profile calibration curve, using the formula obtained from the linear calibration curve. The estimated temperatures found for the gasifier turn-out samples could then be compared to and contrasted with literature, as well as with other petrographic determinations.

3.2.1.2. Vitrinite reflectance on the gasifier turn-out samples. The gasifier turn-out samples, crushed to -1mm, were prepared as per routine preparation for petrography following ISO standards (7404-2). Vitrinite reflectance analyses were performed on a range of the samples, applying ISO standards 7404-5. Results were calculated on the following basis: (1) a mean reflectance (average temperature), (2) top 10% mean reflectance readings and, (3) maximum reflectance readings (peak temperature). The top 10% reflectance reading is assumed to be an indication of the surface temperature, since heat transfer through 100 mm particles (as is the case in the Sasol-Lurgi gasifier) is expected to show large temperature variations across particles of this size.

Difficulties were encountered with the petrographic measurement technique on samples that were located deeper in, and particularly at the bottom of the gasifier, due to the limited amounts of carbon material available for determination of reflectance. In addition, as a result of the physical alterations of the carbon particles, it was sometimes difficult to accurately recognize particles that had originated from vitrinite, as at high temperatures, vitrinite and certain forms of inertinite may appear very similar.

4. Results

The solid particulate temperature profile of the Sasol-Lurgi MK IV FBDB gasifier will be firstly presented, followed by a discussion of the earlier findings of Krishnudu et al. [12,13] and Glover et al. [11].

A graphical representation of the Sasol-Lurgi FBDB MK IV gasifier profile results will be given in the discussion and Figs. 4 and 5. Trend lines are included on the graphs in order to guide the eye. A statistical analysis of the data, with respect to the four distinctive gasification reaction zones identified, i.e. (1) drying, (2) pyrolysis, (3) reduction and (4) combustion (ash-bed) are incorporated throughout, in the form of a zone average value. These zones are integrated with error bars, showing the confidence intervals at a 95% confidence level. It should be noted that the wider the confidence interval, the less accurate the average is estimated to be for that particular zone. This could be due to significant inherent variability occurring within the zone, due to excessive chemical reaction or temperature effects for example. It could also be an indication of the actual response, due to gasification characteristics within that particular zone.



Fig. 5. Chart showing the mean optical reflectance and temperature calibration curve for the feed coal sample 32.

4.1. The Sasol-Lurgi MK IV FBDB gasifier temperature profile results

The temperature calibration curve for the gasifier feed sample is given in Fig. 5. It is clearly observed that an excellent linear correlation was obtained. This result shows that the reflectance of vitrinite (RoV) changes in a linear fashion with respect to temperature and therefore the calibration curve can be used to infer temperatures from the RoV measurements of the gasifier turn-out samples, except for the sample taken at the ash grate, due to difficulties with the determination owing to a lack of "carbon" present in the sample. It was assumed in this case that this sample had experienced a temperature of 350 °C, since this is the temperature at which the ash is discharged from the ash-lock. The actual solids temperature in this area of the gasifier is, however, possibly higher than 350 °C, since some cooling is expected to occur within the ash-lock, below the ash-grate.

It has to be taken into account that the degree of confidence when using vitrinite reflectance for solids' temperature estimation decreases towards the bottom of the gasifier as the vitrinite particles are consumed, i.e. less than 1% coal vitrinite type particles and 9% porous chars remain for reflectance analysis [4].

From the data shown in Fig. 6, it can be seen that the coal/char/ ash particles average temperature ranged from 356–383, 462–711, 828–861 and 439–942 °C in the drying, pyrolysis, reduction and combustion (ash-bed) zones respectively, indicating a significant difference between the various zones.

From Fig. 6 it can also be observed that the coal/char/ash particles surface temperature ranged from 387–617, 787–1004, 958–1031 and 452–1206 °C in the drying, pyrolysis, reduction and combustion (ash-bed) zones respectively, clearly showing the differences between the various zones. Due to the cooling effect; a wide surface temperature range of 452–1206 °C is evident in the combustion zone.

It can further be observed from Fig. 6 that the particulate peak temperature ranged from 446–940, 1051–1132, 992–1137 and 440–1360 °C in the drying, pyrolysis, reduction and combustion (ash-bed) zones, respectively. There was no significant difference in peak temperature between the pyrolysis and reduction zones observed. Due to the cooling effect, a wide surface temperature range of 440–1360 °C is evident for the combustion zone.

Fig. 6 shows that the first couple of samples had a similar average estimated temperature of 354 °C, which lies within the drying zone of the gasifier. Thereafter, a change in the estimated average sample solids temperature occurred, to a maximum of 1008 °C, which correlates with the first significant amount of char reported in the particle type analyses [4].



Fig. 6. Graph depicting the solids temperature profiles obtained for the Sasol-Lurgi MK IV FBDB gasifier showing average, surface and peak temperatures (°C).

At the start of "slow pyrolysis with gasification", another marked increase in estimated temperature is displayed, i.e. average temperature of 617 °C, surface temperature of 986 °C and a maximum peak temperature of 1119 °C. This finding correlates with results reported by Bunt [4] on the particle type analyses and total char results, where a significant increase of char particles were observed (from 15 vol.% to 47 vol.%) in this region of the reactor.

A third noticeable change in reflectance was determined at the end of pyrolysis and start of reduction. The mean RoV changed from 3 RoV%, to 5 RoV%, correlating to an estimated average temperature ranging from 617 °C to 837 °C, respectively. From the end of pyrolysis to the base of the gasifier, the mean reflectance value remained at around 5 RoV%, indicating this to be the maximum mean reflectance determinable from the gasifier turn-out samples. The maximum reflectance reading and the maximum estimated temperature obtained were thus determined to occur at the completion of pyrolysis. At this midpoint level in the gasifier, the estimated average temperature is 837 °C, surface temperature is 1075 °C and maximum temperature is 1175 °C.

As expected, the surface temperature trend is always higher than the average temperature, which is an indication of heat transfer limitations occurring within coarse particles. When comparing the peak temperature profile to the average and surface temperature trends, it is evident that significant temperature variations occur within the axially-derived sample layers in the gasifier, particularly in the top half of the gasifier, where endothermic conditions exist. This difference in temperature range (average, surface and peak temperature) appears to become less in the reduction zone as the char is further gasified and even less still in the oxidation zone, where exothermic conditions prevail. Within the combustion zone and ash-bed, an interesting peak is noticeable in the surface and peak temperature trends, which requires some explanation.

It was postulated that as the agents (oxygen and steam) are added counter-currently to the descending charge (within the ash-bed) and that the heat transfer between solid and gas rapidly increases the gas temperature in order to drive the reduction and pyrolysis reactions higher up in the gasifier. Reactions in this zone are highly exothermic and the rate of oxidation and combustion of the highly aromatic char proceeds rapidly. The oxygen and steam agent distributor is attached to the slowly-rotating ash-grate which extends from the base of the gasifier and the oxygen and steam agent nozzles extend a short distance above the grate. The possibility of uneven gas distribution and poor ash-bed mixing, leading to channeling and localized steam/oxygen variations, may possibly explain the high localized temperature extreme observed. At this point, the peak temperature is well above the ash fusion temperature, which gives rise to clinker formation. This observation confirms the postulation made by Glover et al. [11], from where oxygen enrichment as the likely cause of channelburning, can be considered.

Average temperature profile measurements of the solid particles, when using optical reflectance successfully, revealed the temperature range occurring within the pyrolysis zone of the Sasol-Lurgi mark IV gasifier. The average (mean) temperature ranged from ca. 400 °C up to 850 °C within this region, which is in agreement with literature-cited temperature ranges typically reported for pyrolysis [3]. In this highly endothermic region of the gasifier, the particle surface temperature and peak temperature showed visible evidence of heat transfer limitations occurring through lump coal when compared to the mean particle temperature (variations by as much as 700 °C), providing some evidence of the complex radial and localized behaviour occurring within the averaged axial sample slices. In the oxidizing and combustion regimes, exothermic conditions prevail and heat transfer differences across the particles are minimized.

4.1.1. The temperature profiles of the Indian pilot plant (Krishnudu et al. [13]) and work conducted by Glover et al. [11] on the Sasol-Lurgi MK III Sasolburg gasifier

From the Indian pilot plant data shown earlier in Fig. 4 [12,13] it is clear that the coal/char/ash temperature ranged from 355–600, 601–900, and 901–1200 °C occurring in the (1) fast devolatilisation, (2) slow devolatilisation with gasification and (3) gasification zones, respectively. The Indian pilot plant temperature profile data (Fig. 4) shows some similarity to the Sasol-Lurgi MK IV gasifier average temperature trend (Fig. 6), with a high temperature "hot-spot" measurement shown to occur for both cases near the base of the reactors. This warrants further discussion, since a different analytical technique was applied to obtain the temperature profile of the Indian coal [13].

According to van der Walt [16], Gray-King assays were done on the Indian sequential samples (temperatures ranged from 400 °C to 900 °C), and a correlation between the volatile matter content in the char fractions and the temperature of this low-temperature carbonization was established. The correlation was given as

$log \ VM = 12.823 - 4.308 log \ T$	(1)
$\log VM = 5.689 - 1.613 \log T$	(2)

where VM is the dry, ash-free volatile matter content in the char; T is the temperature in °C for -212 micron coal (Eq. (1)); and for 25–100 mm coal lumps, (Eq. (2)).

Extrapolating outside of the temperature range (>900 °C) casts some doubt on the predicted highest temperature peak value of 1200 °C occurring within the ash-bed for the Indian case. This observation is therefore in agreement with Johnson and Thomas [14], who stated that the use of residual volatiles yield as a temperature indicator for the char samples is questionable. However, the fact that this temperature peak also occurs in the ash-bed region, analogous to the Sasol-Lurgi MK IV gasifier case, where optical reflectance was correlated with controlled feed coal temperature in the range 350–1200 °C (Fig. 5), does offer some respite for the predictive method used by Krishnudu et al. [13].

One could argue that both predictive temperature techniques are incorrect, since the Sasol-Lurgi MK IV gasifier particulate peak temperature of 1400 °C is also outside the calibration range, but the particulate surface temperature above the ash grate is below 1200 °C and also has this characteristic spike profile, which seems to suggest that the increased temperature observed in this region is correct and the peak temperature result of 1400 °C should be treated with some caution. The distribution of oxygen and steam within the ash-bed appears to be a common effect for both the commercial-scale Sasol-Lurgi MK IV gasifier and the Indian cases. Further investigation, in order to fully understand the implications for Sasol-Lurgi FBDB gasification is advised.

Glover et al. [11] used an infra-red spectroscopic technique on the recovered char particles to estimate the solids temperature profile. The average bed temperatures were found not to vary significantly in the top part of the main bed (being in the order of 777 °C) and some evidence was found that the gasifier jacket water acted to cool the boundary layer. Without the interfering effects of the ash-channelling, the bed could be described as an adiabatic core with a cooler boundary layer. The temperatures in the vicinity of the char-ash interface were approximately 877 °C, peaking to 1027 °C.

From the averaged Sasol-Lurgi MK III gasifier data [11] given in Fig. 3 it can be observed that the coal/char/ash temperature significantly differed between the drying and pyrolysis zones, i.e. 383–636 and 760–872 °C, respectively. No data was supplied for the gasification and combustion zones. It can be observed from the trends in Fig. 3 that the Sasol-Lurgi MK III gasifier was operating at a higher average temperature when compared to the other

two cases (Figs. 4 and 6). This is not an unexpected result since the ash fusion temperatures of the Sasolburg coal are known to be higher than is the case of Highveld coals at Secunda where a different coal type is used. The gasifiers are purposely operated at higher temperatures (below ash-melting) resulting in the use of less excess steam and a resultant positive impact on the H to CO ratio. [7].

5. Conclusions

The Sasol-Lurgi MK IV gasifier solids average temperature profile was found to be in reasonable agreement with the work of Krishnudu et al. [12,13], albeit that two different coal types and temperature profile estimation methods were utilized, i.e. optical reflectance and residual volatile matter distribution in the case of the Sasol-Lurgi MK IV gasifier and Indian pilot plant, respectively. A characteristic spike, indicative of an increase in temperature, was found in the sample taken directly above the ash-grate in the case of the Sasol-Lurgi MK IV gasifier, seeming to indicate that agent distribution through the nozzles positioned just above the grate is not uniform, resulting in localized oxygen concentration increases with subsequent "hot-spots" and channel-burning occurring. This finding is also observed from the Indian pilot plant case study, suggesting that homogenization of the ash bed in the fixedbed gasifier could help to optimize the agent distribution within the reactor.

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