FUEL BED EVALUATIONS AND COAL
PROPERTIES TRANSFORMATION IN A
SASOL-LURGI FIXED BED DRY BOTTOM
GASIFIER OPERATING ON NORTH DAKOTA
LIGNITE

A Thesis Submitted to North-West University, Potchefstroom, in Fulfilment of
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By

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To my Beautiful Family:-

Koena (my wife) and Kgothatso (my daughter – Mmatlala lebesabesi, Manape masenelo sa babirwa), this one is dedicated specially to you ladies. Thanks for the support. You are my inspiration...

My late grandmother (Maite masebapale, Mohlapa ke mmamatipa).- may your good soul rest in peace...

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My inlaws - for the first time in my life do I get to have 4 parents and even more siblings. This is a priviledge...

All my friends and relatives...

“Mmirwa a šaya lekopo ga se mmirwa ke mmirwana”
DECLARATION

I, the undersigned, declare that the work contained in this thesis is my own original study and has not been submitted at any university for a degree.

S.J. Mangena

30/04/2009

Date
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"When I am asked what particular research on coal would be of most practical value to those who sell it, equally those who have to use it, I have no hesitation in saying: research on the composition of coal" Richard Vernon Wheeler
SYNOPSIS

The growth in coal consumption worldwide as well as the high oil prices in the recent past has led to the current increased interest in the application of coal gasification technologies. The Sasol-Lurgi Fixed Bed Dry Bottom (S-L FBDB) gasification technology is one such technology that has the biggest market share in the world and maintains its competitive edge particularly with regard to gasification of low grade and low rank coal. To ensure sustained competitive advantage through technology development, it is important to understand the fundamentals of the process as well as the behaviour of coals of different rank in the reactor.

The main objective of this study was to investigate the fuel bed behaviour as well as coal properties transformational behaviour in a S-L FBDB gasifier that gasifies North Dakota (ND) lignite. It was hypothesised in this study that using the FBDB gasifier sampling methodologies available in the literature, with some modifications to suit the context of this study, can help to explain the fuel bed behaviour as well as the coal properties transformational behaviour during gasification of lignite in the S-L FBDB process.

To test the hypothesis and to achieve the objectives of the study, two MK IV S-L FBDB gasifiers (i.e. “Albert” and “Bernice”) operating at the Great Plains Synfuels Plant of the Dakota Gasification Company (DGC) in the United States of America (USA) were sampled using the Turn-Out method developed by Bunt (2006) and modified in this study to suit lignite. The samples were characterised for their chemical, physical, petrographic and mineralogical properties which were then interpreted in terms of their transformation in the various reaction zones of the gasifiers.

The different reaction zones in the “Bernice” and “Albert” gasifiers were successfully identified using chemical analyses (i.e. proximate and ultimate analyses as well as Fischer tar yields). Identification of reaction zones in the S-L FBDB gasifiers operating on lignite is a first in the history of the process. In comparing Secunda GG41 gasifier operating on bituminous coal with the
DGC “Bernice” and “Albert” gasifiers operating on lignite, the reaction zones were found to be very different due to, amongst other things, the different operating philosophy, stability and coal rank. About two thirds of the reactor volume, in the case of DGC “Bernice” and “Albert” gasifiers, was found to be drying and devolatilizing the coal, leaving only about a third of the reactor volume for gasification and combustion. Nonetheless, due to the high reactivity of the lignite, more than 98% of the char/fixed carbon was consumed within a third of the remaining gasifier volume and this is a significant new finding. The fact that the entire reactor volume was utilized for drying, devolatilization, gasification and combustion with carbon conversion of >98%, makes the S-L FBDB gasifier very suitable for lignite gasification. In line with the Secunda GG41 gasifier, clear overlaps between the reaction zones were observed in the “Bernice” and “Albert” gasifiers. This therefore confirms the gradual transition from one reaction zone to another as reported in the literature.

The volatile matter in the ash from both the “Bernice” and “Albert” gasifiers was about 10% (dry basis). This volatile matter is most probably inorganic in nature given the presence, in the samples obtained from the ash bed, of calcite (CaCO₃), gypsum (CaSO₄·2H₂O) and melanterite (FeSO₄·7H₂O) which are expected to decompose during volatile matter determination at 900 °C. Using only the volatile matter, as determined by proximate analyses, to determine the pyrolysis zone position in the reactor will in the case of DGC gasifiers therefore be delusive.

As expected, most of the H, N and S were released in the pyrolysis zone of both the “Albert” and “Bernice” gasifiers. A significant increase in the reactivity of the chars from both the “Bernice” and “Albert” gasifiers was observed in the gasification zones. It is due to this increased reactivity that the char/carbon in these gasifiers were consumed within only a third of the gasifier volume. The increased reactivity is most probably due to the catalytic reactions effected by the organically bound alkali and alkaline earth metals, particularly calcium as the coal was found to be rich in this element.
Thermal fragmentation was found to be severe with the ND lignite tested. The feed coal was found to decrease in size from 3% in the feed to 90% of <6.3 mm fine particles in the drying and pyrolysis zones of both the “Albert” and “Bernice” gasifiers. This is also a new significant finding in the history of the S-L FBDB gasification process which is traditionally known to operate on coarse coal.

Mineral matter in the feed coal to the “Bernice” gasifier was mainly dominated by the organically bound calcium. The crystalline phases in the gasification and combustion zones were dominated by gehlenite and bredigite which may have formed from the transformation, at higher temperatures, of the organically bound Ca and Mg to CaO and MgO and subsequent interaction with the reactive silica and transformation products of the clays.

In the “Bernice” gasifier, a significant amount of calcite was found to be forming in the beginning of the gasification zone, towards the end of pyrolysis, and decomposing slightly in the hotter combustion zone. It is suggested that the calcite was formed from the reaction of CaO (formed from the transformation of the organically bound Ca) with the CO₂ from the raw gas in the gasifier.

As expected, the glass phase was found to be the major part of the ash minerals in the gasification and combustion zones of the “Bernice” gasifier. This phase was composed mainly of the Ca, Mg, Na aluminosilicates with some Fe. This composition is common to the slag formed from the Fort Union lignite. There was therefore a significant amount of melting in the hotter reaction zones (i.e. gasification and combustion zones) of these gasifiers. The organically bound Ca, Mg and Na seemed to have played a significant role in the formation of this glass phase in the gasifiers.

Oxygen scavenging by the ash minerals in the combustion and gasification zones of both “Albert” and “Bernice” gasifiers was observed. In the “Bernice” gasifier, it was estimated at about 16% of the oxygen fed as agent to the gasifier. From an economic viewpoint this is significant given the high cost of producing the 99% pure oxygen for gasification.
The AFT in the feed coal to both “Albert” and “Bernice” gasifiers was found to be higher as compared to the ash samples from the ash bed. This may have implications on the design and operating philosophy since the gasifiers are normally designed to operate between the initial deformation and flow temperatures of the ASTM ash, which is not the same as the ash formed in the gasifier. This is also another new significant finding. The high concentration of the fluxing elements (i.e. Ca, Mg, Na and Fe) in the dominating glass phase determined in the gasification and combustion zones of these gasifiers was most probably the reason behind this phenomenon.

The char particles formed in both “Bernice” and “Albert” gasifiers were, as determined petrographically, mainly dominated by the dense chars which were highly reactive. An induced “coalification” process was observed in both “Bernice” and “Albert” gasifiers with the macerals/char particles being transformed from lignite to bituminous and anthracitic coal particles.

In the “Bernice” gasifier, the average temperature of solids in the combustion zone was found to be about 700 °C, peaking at 1100 °C in the combustion zone. The average temperature is in line with the predicted figures (i.e. 741 °C for the predicted temperature at which the water gas shift reaction was forced into equilibrium).

Overall, there was an excellent match in the trends of the chemical, physical, petrographic and mineralogical properties of the samples obtained at different levels of the “Albert” and “Bernice” gasifiers. This may therefore confirm plug flow during the Turn-Out sampling methodology and hence supports the hypothesis of this study.

It is hoped that the results obtained in this study will not only benefit Sasol or Sasol-Lurgi Technology Company with regard to the understanding of the reactors and improvement in modelling and design, but will also assist DGC in further optimising their lignite gasification process.
OPSOMMING

Die wêreldwye groei in steenkoolverbruik, sowel as die hoë olieprys, het gelei tot die huidige verhoogde belangstelling in die toepassing van steenkoolvergassingstegnologieë. Die Sasol-Lurgi-vastebed nie-slakkende vergassingstegnologie is een so 'n tegnologie wat die grootste markaandeel in die wêreld het en wat sy mededingende voordeel behou, in die besonder met betrekking tot vergassing van laaggraad- en laerang-steenkool. Om volghou die mededingende voordeel deur tegnologie-ontwikkeling te versek, is dit belangrik om die beginsels van die proses, sowel as die gedrag van steenkool van verschillende rang, in die reaktor te verstaan.

Die hoofdoelwit van hierdie studie was om die brandstofbed-gedrag, sowel as die transformatie van steenkooleienskappe in 'n Sasol-Lurgi-vastebed nie-slakkende (S-L FBDB)-vergasser wat Noord-Dakota (ND) ligniet vergas, te ondersoek. Daar word in hierdie studie gehypothesese dat gebruik van die FBDB-vergasser-monsternemingsmetodologie beskikbaar in die literatuur, met enkele modifikasies om die konteks van hierdie studie te akkommodeer, kan help om die brandstofbed-gedrag te verduidelik, sowel as die steenkolleienskappe se transformatiesegedrag gedurende vergassing van ligniet in die S-L FBDB-proses.

Om die hipotese te toets en die doelwitte van die studie te bereik, is twee MKIV Sasol-Lurgi-vastebed nie-slakkende vergassers (naamlik "Albert" en "Bernice") gemonster deur gebruikmaking van die "Turn-Out"-metode soos ontwikkel deur Bunt (2006) en gemonstereer in hierdie studie om ligniet te pas. Die monsters is gekarakteriseer ten opsigte van hulle chemiese, fisiese, petrografiese en mineralogiese eienskappe, wat dan geïnterpreteer is met betrekking tot hulle transformatie in die verskeie reaksioneses van die vergassers.

Die verschillende reaksioneses in die "Bernice"- en "Albert"-vergassers is suksesvol geïdentifiseer deur chemiese analises (dws voorlopige en finale analises, sowel as Fischer-teeropbrengste). Identifisering van reaksioneses in die S-L FBDB-vergassers wat met ligniet bedryf is, is 'n eerste in die
geskiedenis van die proses. In die vergelyking van Secunda-GG41 met die DGC (Dakota Gasification Company) is die reaksiesones in "Bernice"- en "Albert"-vergassers baie verskillend gevind agv, onder andere, verskillende bedryfsfilosofieë, stabiliteit en steenkool-rang. In die geval van DGC "Bernice" en "Albert" is gevind dat meer as die helfte van die reaktorvolume in beslag geneem word deur droging en ontvlugtiging van die steenkool, wat slegs ongeveer n derde van die reaktorvolume vir vergassing en verbranding oorlaat. Nietemin, agv die hoë reaktiwiteit van die ligniet, is meer as 98% van die sintel/vastekoolstof-verbruik binne n derde van die oorlywendé vergasservolume, wat n betekenisvolle nuwe bevinding is. In lyn met die Secunda GG41-vergasser, is duidelike oorvleuelings tussen die reaksiesones in die "Bernice"- en "Albert"-vergassers waargeneem. Dit bevestig dus die geleidelike oorgang van een reaksiesone na n ander, soos gerapporteer deur Higman en van der Burgt (2003).

Die vuigtige materiaal in die as van beide die "Bernice"- en "Albert"-vergassers was ongeveer 10% (droë basis). Hierdie vuigtige materiaal is heelwaarskynlik anorganies van aard, gegee die teenwoordigheid van Kalsiet (CaCO₃), Gips (CaSO₄.2H₂O) en Melanteriet (FeSO₄.7H₂O) in die monsters verkry uit die as-bed. Die gebruik van slegs die vuigtige materie, soos bepaal in voorlopige analysies, om die pirolise-sone te bepaal, sal dus in die geval van DGC-vergassers misleidend wees.

Soos verwag, is die die meeste van die H, N en S vrygestel in die pirolise-sone van beide "Albert"- en "Bernice"-vergassers. n Betekenisvolle toename in die reaktiwiteit van die sintels vanaf beide die "Bernice"- en "Albert"-vergassers is in die vergassingsones waargeneem. Dit is weens hierdie vermeerderde reaktiwiteit dat die sintel/koolstof in hierdie vergassers binne slegs n derde van die vergasservolume verbruik is. Die vermeerderde reaktiwiteit is heelwaarskynlik agv die katalise deur die organiesgebonde alkali en alkali-aard metale, in die besonder kalsium.

Aansienlike termiese fragmentering is gevind by al die steenkool wat getoets is. Daar is gevind dat die steenkool afneem in grootte tot 90% fyn partikels
van <6.3mm in die droging- en pirolise-sones van beide "Albert"- en "Bernice"-vergassers. Hierdie is ook 'n nuwe betekenisvolle bevinding in die geskiedenis van die S-L FBDB-vergassingsproses, wat tradisioneel met grewe steenkool bedryf word.

Mineraal-materiaal in die voersteenkool na die "Bernice"-vergasser is hoofsaaklik oorheers deur die organiesgebonde kalsium. Die kristallyne fases in die vergassing- en verbrandingsones is oorheers deur gehleniet en bredigiet wat gevorm kon gewees het deur die transformasie, by hoër temperature, van die organiesgebonde Ca en Mg na CaO en MgO en opvolgende interaksie met die reaktiewe silika en transformasieprodukte van die kleie.

In die "Bernice"-vergasser is 'n betekenisvolle hoeveelheid kalsiet gevind, gevorm in die begin van vergassingsone, naby die einde van pirolise, en wat effens ontbind in die warmer verbrandingsone. Daar word voorgestel dat die kalsiet gevorm word deur die reaksie van GaO (gevorm deur die transformasie van die organiesgebonde Ca) met die CO₂ in die rougas van die vergasser.

Daar is gevind dat die glasfase die belangrikste deel van die as-minerale in die vergassing- en verbrandingsones van die "Bernice"-vergasser uitmaak. Hierdie fase het hoofsaaklik bestaan uit Ca-, Mg- en Na-aluminosilikate met 'n bietjie Fe. Hierdie samestelling is algemeen in die slak wat vorm uit die Fort Union-ligniet. Daar was dus 'n betekenisvolle hoeveelheid van smelting in die warmer sones van hierdie vergassers. Die organiesgebonde Ca, Mg en Na het blykbaar 'n betekenisvolle rol gespeel in die vorming van hierdie glasfase in die vergassers.

Suurstof-opruiming deur die as-minerale in die verbranding- en vergassingsones is waargeneem in beide "Albert"- en "Bernice"-vergassers. Vir die "Bernice"-vergasser is dit beraam as ongeveer 16% van die suurstof wat as agent na die vergasser gevoer is. Uit 'n ekonomiese oogpunt is dit betekenisvol, gegee die hoë produksiekoste die 99%-suiwer suurstof vir vergassing.

Confidential
Daar is gevind dat die AFT in die voersteenkol na beide “Albert”- en “Bernice”-vergassers hoër is in vergelyking met die as-bed. Dit mag implikasies hé vir ontwerp- en bedryfsfilosofie, aangesien die vergassers gewoonlik ontwerp word om bedryf te word tussen die aanvanklike verving- en vloeitemperatuur van die ASTM-as, wat nie dieselfde is as die as gevorm in die vergasser nie. Hierdie is ook ’n verdere nuwe betekenisvolle bevinding. Die ooreersende glasfase, soos gevind in die vergassing- en verbrandingsones van hierdie vergassers, was heel waarskynlik die rede vir hierdie verskynsel.

Die sintel-partikels gevorm in beide “Bernice”- en “Albert”-vergassers is, soos petrografies bepaal, hoofsaaklik ooreers deur die digte sintels wat verbasend reaktief is. Dit mag dus die hipotese aangaande die katalisering van die vergassingsreaksies deur kalsium verder versterk. ’n Geïnduseerde “versteenkolisifisering”-proses is waargeneem in beide “Bernice”- en ”Albert”-vergassers met die maseraal/sintelpartikels wat getransformeer word van ligniet na bitumineuse en antrasitiese steenkoolpartikels.

Daar is gevind dat die gemiddelde maksimum temperatuur van die soliedes in die “Bernice”-vergasser ongeveer 700 °C is, met ’n piek van 1100 °C in die verbrandingsone. Hierdie temperature is in lyn met die voorspelde waardes (dws ewewig water-gas skuifreaksietemperatuur van 741 °C).

Globaal was daar uitstekende ooreenkomste in die neigings van die chemiese, fisiese, petrografiese en mineralogiese eienskappe van die monsters, verkry op verskillende vlakke van “Albert”- en “Bernice”-vergassers. Dit mag dus propvloei gedurende die "Turn-Out"-monsterneming-metodologie bevestig en dus die hipotese van hierdie studie ondersteun.

Daar word gehoop dat die bevindinge van hierdie studie nie net Sasol of Sasol-Lurgi Technology Company sal begunstig met betrekking tot die verstaan van die reaktore en verbetering van modellering en ontwerp nie, maar DGC ook sal help in die verdere optimisering van hulle ligniet-vernassingsproses.
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<th>Description</th>
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<tbody>
<tr>
<td>AFT</td>
<td>Ash Fusion Temperature</td>
</tr>
<tr>
<td>ASTM</td>
<td>American Society for Testing and Materials</td>
</tr>
<tr>
<td>BEPC</td>
<td>Basin Electric Power Cooperative</td>
</tr>
<tr>
<td>BFW</td>
<td>Boiler Feed Water</td>
</tr>
<tr>
<td>BGL</td>
<td>British Gas Lurgi</td>
</tr>
<tr>
<td>CCSEM</td>
<td>Computer Controlled Scanning Electron Microscope</td>
</tr>
<tr>
<td>CLL</td>
<td>Chemie Linz-Lurgi</td>
</tr>
<tr>
<td>CTL</td>
<td>Coal to Liquids</td>
</tr>
<tr>
<td>DAF</td>
<td>Dry, Ash Free</td>
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<td>DB</td>
<td>Dry Basis</td>
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<tr>
<td>DGC</td>
<td>Dakota Gasification Company</td>
</tr>
<tr>
<td>DT</td>
<td>Initial Deformation Temperature</td>
</tr>
<tr>
<td>DRI</td>
<td>Direct Iron Reduction</td>
</tr>
<tr>
<td>EI</td>
<td>Ergun Index</td>
</tr>
<tr>
<td>EXAFS</td>
<td>X-ray Absorption Fine Structure Spectroscopy</td>
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<tr>
<td>FT</td>
<td>Fischer – Tropsch</td>
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<tr>
<td>ICCP</td>
<td>International Committee for Coal and Organic Petrology</td>
</tr>
<tr>
<td>ISO</td>
<td>International Organization for Standardization</td>
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<tr>
<td>LTA</td>
<td>Low Temperature Ash</td>
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<td>MEE</td>
<td>Multiple Effect Evaporators</td>
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<td>MTI</td>
<td>Microbeam Technologies Inc.</td>
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<tr>
<td>ND</td>
<td>North Dakota</td>
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<tr>
<td>PSD</td>
<td>Particle Size Distribution</td>
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<td>RoM</td>
<td>Run of Mine</td>
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<tr>
<td>SABS</td>
<td>South African Bureau of Standards</td>
</tr>
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<td>SANS</td>
<td>South African National Standard</td>
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<td>S-L FBDB</td>
<td>Sasol-Lurgi Fixed Bed Dry Bottom</td>
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<td>SNG</td>
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<td>SRU</td>
<td>Sulphur Recovery Unit</td>
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