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Sulphur removal from coal or from products: is prevention better than cure? - a techno-economic review

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1 INTRODUCTION

South Africa has large reserves of various types of coal. The sub-bituminous coal reserves are mostly of low grade, grade being indicated by the percentage of ash in the coal (mineral matter left over after combustion; the higher the ash percentage, the lower the grade). These reserves are mostly used for power generation in pulverised fuel fed boilers for ESKOM (approximately 95 million ton annually), or converted into fuels and chemicals at the Sasol complexes in Sasolburg and Secunda (nearly 44 million ton during financial year 2007).

There are several by-products from the utilisation of these coals, the most obvious one being ash, but also sulphur dioxide, hydrogen sulphide and carbon dioxide is formed. Depending on the process wherein the coal is used, a greater amount of energy and effort must be used to handle the by-products or to prevent them from being formed.
In the power stations, the main product of the utilisation of the coal is the energy released in the firebox of the boilers and transferred into the water / high-pressure steam. Ash, carbon dioxide and sulphur species are by-products which do not directly influence the achievement of the main purpose of the operation of the boiler namely to convert energy in the coal to energy in the form of electricity. It should be noted though that there is a conversion efficiency penalty due to the presence of ash (mineral matter) and sulphur in the feed coal.

For the indirect coal liquefaction processes of Sasol there is more than just a thermal efficiency aspect. The main products of the overall processes are carbon molecules in various forms. The process of converting carbon-in-coal to carbon-in-value-added-hydrocarbons requires the unlocking of carbon from the coal structure. This unlocking of the carbon takes place in the gasification part of the Sasol process chain. Other components of the coal structure like hydrogen, nitrogen and sulphur are also released and exit the gasification step together with the carbon molecules. Before the carbon (in the form of carbon monoxide) can be converted with hydrogen in the Sasol Synthol process to hydrocarbons, the sulphur species and carbon dioxide need to be removed from the gas stream produced in gasification. To remove these components several process steps with expensive equipment and incurring significant operating costs are used, increasing the Total Cost of Ownership (TCO) and production costs of the final hydrocarbon end-products as well as reducing overall carbon efficiency.

The removal of the sulphur components and carbon dioxide from the gasification product stream (raw gas) results in a waste gas stream rich in carbon dioxide and containing almost all of the gaseous sulphur species produced in the gasifiers. These sulphur

species, predominantly hydrogen sulphide, should not be released into the atmosphere. Although the concentrations are low enough not to affect the health of humans or animals, they have an unacceptable smell and could, through atmospheric oxidation, potentially have a long-term effect on acid rain production and the environment in general. Process steps are therefore included in the Sasol process chain to capture the sulphur and convert this to elemental sulphur, suitable for sale. Again, these processes increase the TCO and the input-costs of final products.

With increasing throughput through the processes to support business growth, and increasing environmental pressure, it is required to further reduce the production of sulphur species and their release into the atmosphere. The reduction efforts have traditionally been focused on the capture of the sulphur species from the waste gas streams after they had been produced, and not too much attention was given to reducing the production of the sulphur species in the first place. This reduction in sulphur species production could be achieved by presenting less sulphur components to the gasification process through the removal of sulphur components from the coal used as gasifier feed material.

Sasol did not practise coal beneficiation in general (other than some rudimentary screening) until the Twistdraai Mine Coal Export plant was commissioned in the mid 1990's. The general notion appeared to be that coal beneficiation for the gasification processes was prohibitively expensive and would result in an overall reduction in complex life and economics. Earlier efforts to highlight the potential role and benefits of coal beneficiation with respect to environmental impacts and sulphur emissions, had been put down with these arguments, hence no proper evaluation of that potential exists.

This thesis focuses on the comparison between the removal and capture of sulphur species from the gasification raw gas stream, and the removal of the sulphur from the feed material (coal) to the gasifiers and specifically through the use of coal beneficiation processes. The objective is to evaluate, in the broadest sense, whether the sum total of benefits of the removal of sulphur from the coal prior to use in the gasification through coal beneficiation is more than handling the sulphur species in the gas streams after gasification. In other words: is prevention better than cure?
The scope of the thesis focuses on the Sasol Secunda complex in specific.

1.1 Background: Sasol's Coal Conversion Process

Sasol is by far the world's leading company in the upgrading of low-grade coal into high value chemicals and fuels. The Sasolburg complex is in operation for more than fifty years and has been converted from coal feedstock to natural gas feedstock in the 1990's. The Secunda Sasol Synfuels complex is in operation since 1979, and converts approximately 40 million tons of sub-bituminous coal per annum into automotive fuels and high value chemicals. Two-thirds of the complex's feed is for the 80 gasifiers and the production of synthesis gas for the Sasol Advanced Synthol Fischer Tropsch hydrocarbon synthesis process. The synthesis units of Sasol Synfuels produce approximately 170 000 barrels of crude oil equivalent fuel and chemical products per day. This production will increase over the years to come, and production expansion will be based on coal as well as natural gas. Figure 1.1 shows an overview of the Sasol processes as employed in Secunda, Mpumalanga province, South Africa.

The coal reserves of Sasol in the Secunda area are of relatively low grade. Only a small portion of the coal mined is of higher grade and is exported as steam coal (approximately 3 million ton product per year), the rest is for the gasifiers and the raising of steam for the processes.

Typically, the coal yields between 20 and 35 mass% ash, 22 to 25 mass% volatile matter and contains some inherent moisture (4-6 mass%). The mineral matter and organic components in the coal contain 1-2 % sulphur by mass (air dried basis). This sulphur is chemically converted to sulphur dioxide in the combustion processes of the steam-raising boilers, and to hydrogen sulphide in the gasification reactions in the gasifiers.

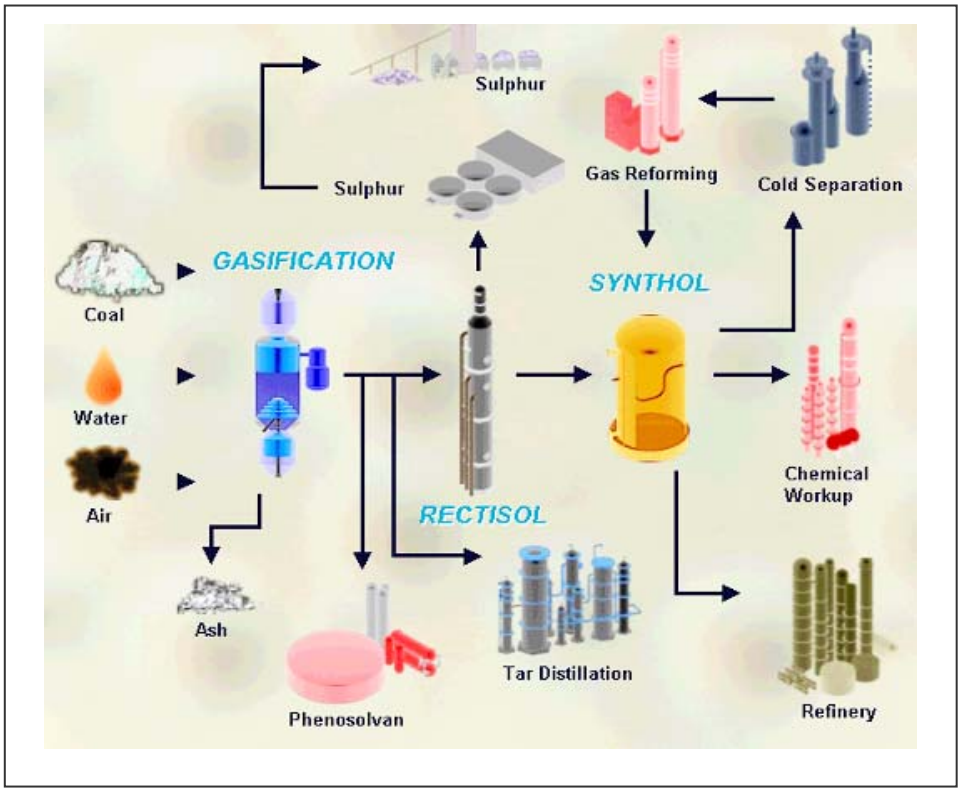


Figure 1.1 Overview of the Sasol coal conversion processes in Secunda (courtesy Sasol)

The Sasol Synthol Fischer Tropsch synthesis process converts synthesis gas into hydrocarbons with the aid of a catalyst. The catalyst in the Secunda complex is iron based and has a limited tolerance for sulphur. Hence, the raw gas produced in the gasifiers is purified. Removal of hydrogen sulphide and carbon dioxide takes place in Rectisol, a low temperature methanol based absorption system. Carbon dioxide and hydrogen sulphide saturated methanol is expanded in a vacuum column to release the hydrogen sulphide and carbon dioxide from the methanol.

The off-gas from the Rectisol process has been treated with several processes over the last 30 years, with more or less success. Originally, the British Gas Stretford process was employed to handle this stream, but found to be inadequate in a high CO_2 concentration environment. Sasol continued to develop improvements and new technologies to reduce the releases of hydrogen sulphide into the environment to within accepted norms and, in co-operation with industry experts, developed a redox process based on vanadium pentoxide. This Sulfolin process converts the hydrogen sulphide into elemental sulphur. The sulphur is sold to Foskor in Phalaborwa in the Limpopo province, and is used in the production of phosphoric acid.

The complex in Secunda will continue to expand and will, in part, use coal as feedstock for this expansion, putting additional stress on the process units within the complex to

handle the process and by-product streams. The existing systems need to be evaluated for the expected extra loads, and this poses the question whether the systems should be expanded with more-of-the-same technologies, new technologies or attention be given to preventing the sulphur from reaching the gas streams in the first place.

1.2 The Coal Resource

In broad terms, coal seams were produced over 270 million years ago, through a process of deposition and decay of vegetation and subsequent coalification via peat to bituminous coal and anthracite. In the swamp-like areas of Laurasia (roughly the Northern Hemisphere), vegetation simply fell down wherever the plants and trees were standing, forming thick layers of vegetation. Over the years, the layer became thicker, more compressed and with pressure, temperature and time coalification started taking place. As the swamps and forest floors were flooded by shifting rivers and after ice ages, mineral matter was deposited on top of the seams. This led to coal seams being interspersed with mineral matter layers, where the coal layers themselves are relatively low in mineral matter.

The Gondwana-land, which includes South Africa, Australia and India, coal formation process was similar for some of the coal deposits, but many of the coal deposits were formed from vegetation that flowed into lower lying basins with rivers and streams. Together with the vegetation, minerals were deposited intermixed with the vegetation, leading to coal seams not only interspersed with mineral matter layers in between the coal layers, but also with a high mineral matter content in the coal layer itself.
Figure 1.2 shows a presentation of how the coal deposits most probably have formed, through co-deposition of organic and mineral matter into the delta areas of river systems.

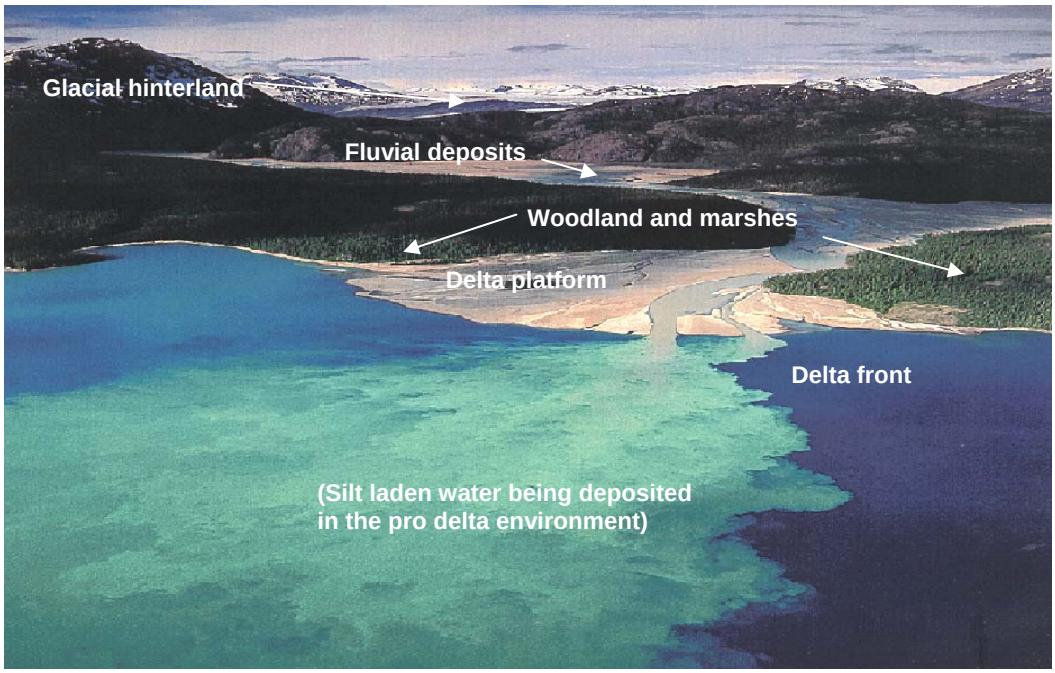


Figure 1.2 Co-deposition of organic and inorganic material (courtesy Sasol Geology Department)

The minerals in the coal seams result from various sources, and can be grouped into two categories. Intrinsic inorganic (mineral) matter is the matter that was present in the original vegetation and got eventually included in the coal as very small grains, or chemically altered and included in the organic structure of the coal. Extrinsic mineral matter is mineral matter that is introduced into the coal, like erosion products of the areas where the streams and rivers passed through, as well as from precipitation of dissolved minerals present in the water [Falcon [(1)]].

Depending on the circumstances during and after the formation of the deposit, the extrinsic mineral matter is either syngenetic (formed and deposited at the same time as the vegetation was deposited) or epigenetic (deposited or transformed after the formation of the deposit). Tectonic movement, thermal conversions due to lava intrusions, weathering, etc. all had an influence on the nature of the mineral matter.

Figure 1.3 is an artist's impression of the progression over time for the coal deposits.

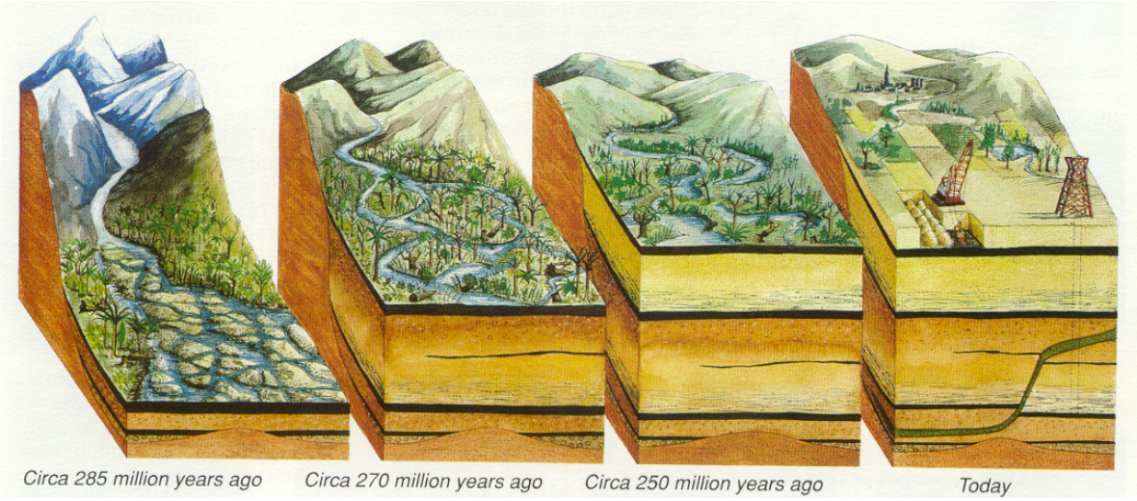


Figure 1.3 Progression of coal deposit over time [Chamber of Mines [(2)]]

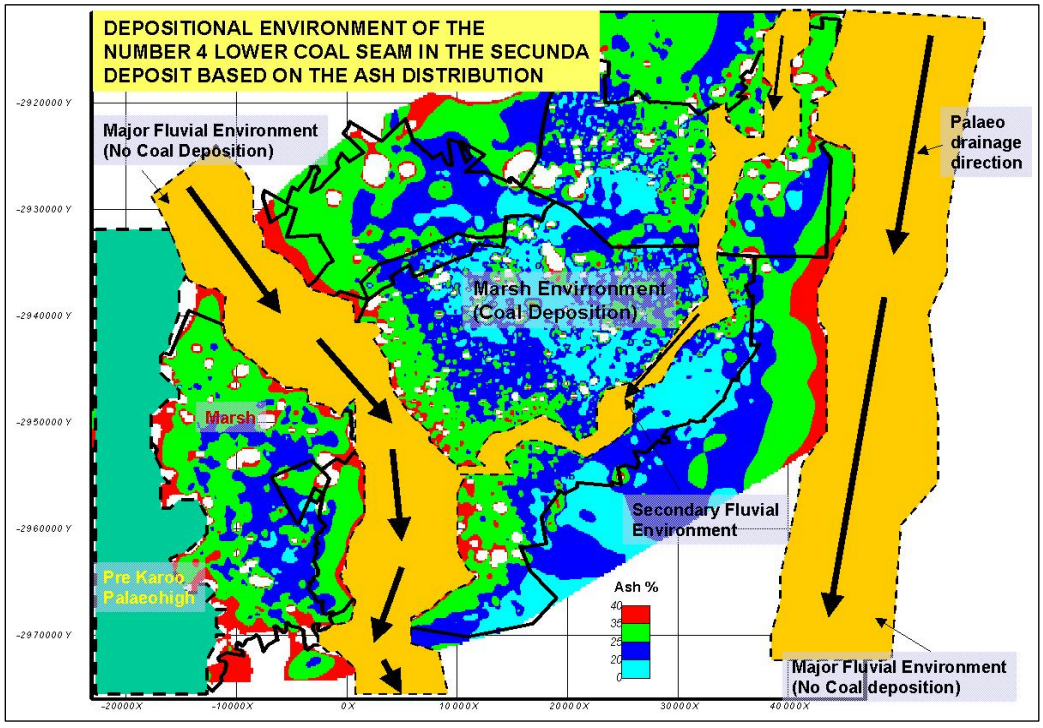


Figure 1.4 Effect of drainage channels on Secunda coal deposit (courtesy Sasol Geology Department)

Figure 1.4 displays the coal deposits as they are present in the Secunda area. This is a typical example of a deposit where the organic material was transported into the area by means of the rivers and streams, with some marshy areas in the centre of the deposit where substantially less or no water flow occurred. The paleo-environmental reconstruction shown in figure 1.4 is the result of the interpretation of data obtained from geological surveys as well as data from actual mining. The delineation of the coal deposit by the drainage channels can clearly be seen, as well as the increased higher mineral matter content closer to the interface between the coal deposits and the drainage channels. Dykes and sills were formed through lava intrusions.

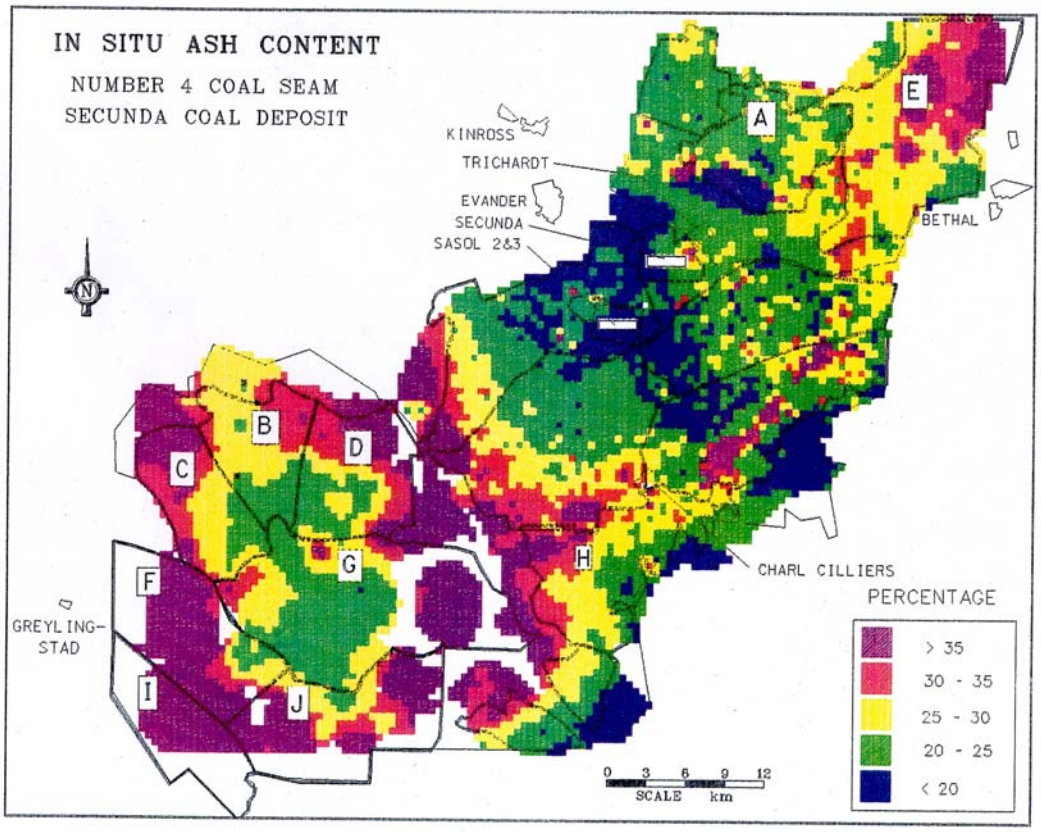


Figure 1.5 Ash yield distribution of the Secunda coal deposit (courtesy Sasol Geology Department)
(Note: A, B, C etc. refer to the individual coal blocks within the greater Sasol reserve)

The drainage channels in figure 1.4 were indicated from the contours of the distribution of the ash yields in figure 1.5. The two synfuels complexes are indicated on the figure as Sasol 2 and 3 – together they are currently known as Sasol Synfuels. They are positioned in the best parts of the coal reserves (lowest percentage of ash in the coal) and with very acceptable geological conditions for mining. However, the mining activities have progressed through these good parts and will soon be reaching areas of higher in-

situ ash yield and different geological conditions. The mining conditions will have to be adjusted to ensure a continued production of acceptable quality coal at ever increasing production rates. Selective in-seam mining may have to be considered.

The focus of this investigation is on the sulphur compounds in the coal seams.
A similar distribution of the sulphur content in the coalfield is observed. Figure 1.6 is a representation of that distribution. As is with the ash yield of the coal, the areas mined in the past were of lower sulphur content. Future mining will encounter different, mostly higher sulphur contents. The increase in sulphur content is fortunately not that pronounced as that of the ash yield.

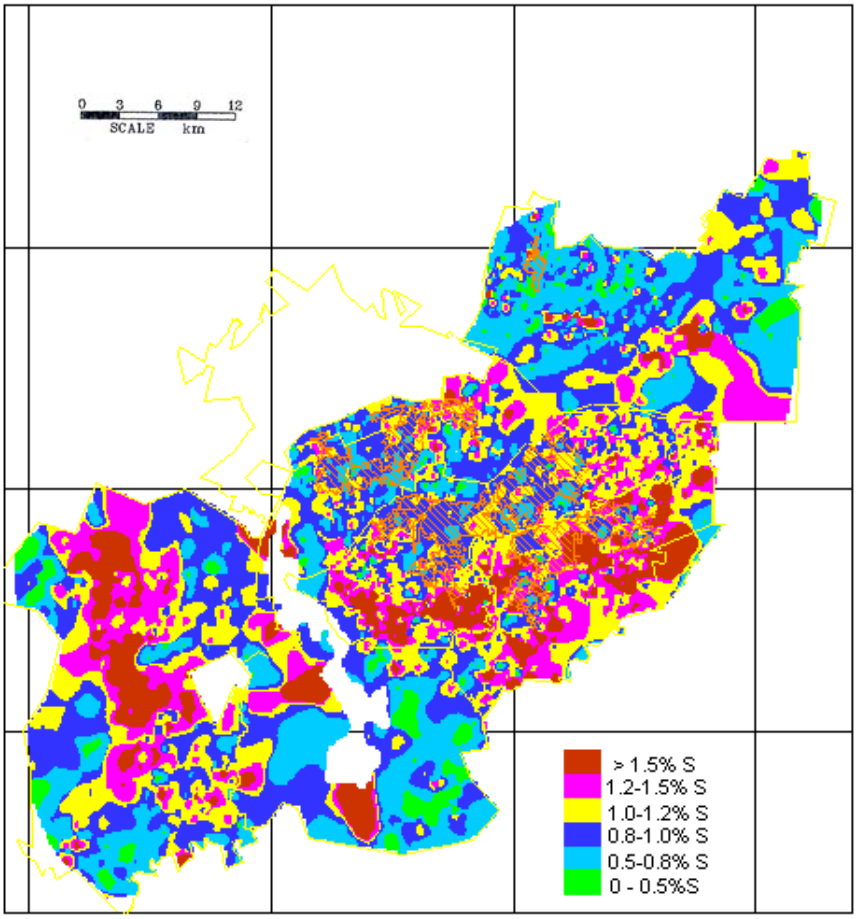


Figure 1.6 Sulphur content distribution of the Secunda coal deposit (courtesy Sasol Geology Department)

When reviewing the data for this thesis it was observed that the sulphur content seems to be lower nearer to the flow channels in the deposit, and higher further away from these channels. This may be an area for further study to investigate whether this observation relates to the formation of the deposit (i.e. the types and amounts of organic and mineral

material deposited), or due to a preferential washing-out of sulphur species nearer to the flow channels.

If coal beneficiation were to be applied to the coal produced in the future, there is potential to reduce both the sulphur and the ash yield at the same time. This potential synergy needs to be incorporated into the investigation.

The inclusion of a coal beneficiation step in the process chain of the Sasol Secunda processes could also allow the mining of coal of lesser quality or a higher degree of mining-derived contamination (floor, roof, interburden). The beneficiation process can remove a substantial portion of this material, if not all, and render an acceptable coal quality for Sasol Synfuels.
Increased coal reserve utilisation is then possible, extending the commercial life of the complex, and enhancing the utilisation of a National Resource.

1.3 Process Consequences Of Coal Quality

The coal produced by the mines is stockpiled in the Coal Supply areas of Sasol Coal. A certain amount of blending and mixing takes place to ensure that the variation in coal quality in the coal from the various shafts is reduced. As the stockpiles were originally designed as emergency supply stockpiles and not as proper blending systems, the effectiveness of the variation-reduction is not that high. The effectiveness will reduce even further when the coal supplied to the stockpiles declines in grade, quality and degree of variation when the mining operations move into new areas of possibly lower quality coal.

Although the gasifiers are in continuous operation, the feed system to the individual gasifier is a batch process. Hence, any variation in quality over time in the coal supplied to the gasifier feed system causes a step change in the operation and operating conditions of the gasifiers, and therefore in the also in the product quality of the raw gas. This variation is carried forward into the Synfuel complex and introduces instability in the conversion process that will be noticeable all the way down to the synthesis process. The instability could not only cause a reduction in complex loads, but will certainly cause the complex to operate away from the optimum efficiency point and be less economically efficient for the duration of the instability.

Variations in coal quality are indicated by the Operation Teams as, predominantly, variations in size distribution, ash yield of the coal, “stone content” (a Sasol-colloquial term for adventitious mineral matter) in the coal and ash (mineral) melting temperatures.

Increased attention to the sulphur emissions from the complex has highlighted the need to analyse for and manage (within the capabilities of the system) the total sulphur content of the coal as well. Variations in the sulphur content of the coal result in changing hydrogen sulphide content in the raw gas from the gasifiers. This causes an increase in the amount of sulphur to be removed in the Rectisol process and hence an increased loading of the Sulfolin sulphur recovery unit. Since this recovery unit has a limited process capacity and efficiency, any variation in the sulphur loading will result in a variation in the final hydrogen sulphide emissions into the atmosphere. Compliance with environmental regulations is taken very seriously, to the extent that total complex production is reduced in case of non-compliance rather than exceeding the agreed-to limits. On the scale that the Secunda complex operates, even a slight reduction in the production rates has a significant impact on production income.

1.4 Benefits Of Coal Beneficiation

As mentioned in paragraph 1.3, an increased sulphur content of the coal will lead to higher hydrogen sulphide loads to the sulphur recovery unit. The potential consequences of increased sulphur content of the coal are exceeding the agreed emission limits due to insufficient sulphur recovery capacity and possible complex load reduction to stay within the allowed limits. Using the 2007 financial year performance of Sasol Synfuels [Sasol [(3)]], R 16 251 million was contributed to Sasol profits, equivalent to R 44.5 million per day for the Synfuel complex. Even a small reduction in production for a part of the day would result in a significant amount of lost income.

Depending on the method selected for the reduction of the sulphur load in the complex, there will be more or less benefits. If the sulphur recovery units were to be expanded, or an alternative process is implemented to complement the existing processes, additional saleable product could be made. This could be elemental sulphur if similar processes as the current are installed, or sulphuric acid or ammonium sulphate if different processes are selected.

Beneficiation of the coal to the gasification units would have multiple benefits. Not only will beneficiation reduce the sulphur content of the coal, but also the ash yield. The reduced sulphur content will reduce the sulphur load in the raw gas and to the recovery units, therefore debottlenecking the sulphur recovery unit (albeit at the cost of lower income from the sale of elemental sulphur).

The reduced ash yield of the coal has multiple benefits. Ash (or mineral matter) is relatively inert in the gasification process but is heated-up together with the useable part of the coal. This heat is lost when the hot ash (approximately 300 °C) is discharged at the bottom of the gasifiers into a water sluiceway system. The heat thus lost was originally released from the coal through the combustion of carbon with oxygen (from the supplied pure oxygen as well as from the dissociation of steam) and therefore represents a cost-negative use of utilities and feed material.

The ash / mineral matter is abrasive, and causes erosion on the mechanical parts of the gasifier: coal feeders, ash removal grates and ash hopper systems. Reducing the mineral matter content of the coal through a beneficiation process would lower the maintenance costs as well as the associated cost of production loss when equipment is down for maintenance in bottleneck periods.

As mentioned, the gasifier is fed through a batch coal feeding system. Hence, there is a volumetric limitation to the amount of coal that can be fed per time unit to the gasifier. For operation at higher gas production loads, sufficient carbon must be fed into the gasifier. If the mineral content of the coal increases, the number of coal locks required to supply the necessary amount of carbon will increase, to a point where it becomes impossible to match the carbon needed with the carbon supplied. Reducing the mineral content of the coal in a beneficiation step will allow the gasifier to produce at higher loads without exceeding the volumetric feed limitation, and also operate at normal loads with a lower number of coal lock filling cycles per time unit. Higher loads lead to more income, less coal lock cycles incur less maintenance costs.

The most significant benefit of coal beneficiation could however be the extension of the complex life through increased extraction and reserve utilisation. Coal beneficiation would enable the mining process to remove more coal from the reserve's areas of higher ash yield and/or less advantageous mining conditions since the higher mineral matter content material would be removed from the feed stream in the beneficiation process. Admitted, some carbon will be lost in the beneficiation process due to inherent beneficiation process inefficiencies (and this is included in the carbon balance calculations in chapter 8 and compensated for in the financial evaluations in chapter 9) , but the net gain in useable carbon from a larger part of the reserves should be substantial and more than adequately compensate for this as well as the capital and operating costs of the beneficiation process.

This thesis investigates the technical and economic feasibility of the coal beneficiation options to reduce the sulphur load to the complex (specifically to the gas production section of the complex) in comparison to the implementation of additional or complimentary sulphur recovery processes on the gas production off-gas systems, with due recognition of potential benefits and disadvantages on the greater Secunda Sasol complex.