

## BCL SULPHUR CAPTURE OPTIONS

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### **Abstract**

BCL Limited operates a nickel and copper mining and smelting operation at Selebi-Phikwe in Botswana. The smelter comprises a flash furnace, four Pierce Smith converters, and slag cleaning electric furnaces. Concentrate feed to the smelter is a nominal 2600 tpd containing 30% sulphur, of which only 8% is captured; the difference is emitted to atmosphere. Eighty percent of the sulphur is eliminated in the flash smelting stage; hence the initial focus is on the treatment of this gas stream. Various technology options to capture sulphur have been considered, ranging from scrubbing to sulphuric acid and sulphur manufacture. The main problem facing any initiative to capture sulphur from smelters is the issue of marketability of the end product. Sulphuric acid production poses transport and logistical difficulties, owing to the distance of the BCL mine site from potential customers. Another option is the production of elemental sulphur, which is easier to store and transport. This technology is not quite proven for smelter off-gases. A third option of scrubbing and stockpiling sulphur in an inert form (usually gypsum) has the disadvantage of huge operating costs and logistical difficulty, and potential difficulty with disposal. The gypsum could be sold as an additive to cement-based products for building and construction, or could be recovered by a gypsum reduction roast / sulphur stripping process. This technology shows great promise but is currently only at the pilot-plant stage. In this paper these various options are explored in terms of technical, economic, and logistical feasibility.

### **1. Introduction**

BCL has been emitting sulphur dioxide to the atmosphere since the start of the smelter at Selebi-Phikwe in 1973. A sulphur production plant ran briefly in the 1970's but was decommissioned owing to operating and plant design problems. Since then the construction of a new facility has always been prohibited by lack of funding and the logistics of disposing of the sulphur product, especially in the case of sulphuric acid. Recently the need for sulphur capture was re-emphasised for strategic and environmental reasons. The Botswana Government Department of Environmental Affairs has also stressed the need for BCL to adopt plans to reduce sulphur emissions. A conceptual and prefeasibility study has been initiated to look at technology choices, costs, market outlook and logistics. Three basic technologies exist to recover sulphur on this scale: elemental sulphur production, sulphuric acid production, and alkali scrubbing to produce a "safe" gypsum stockpile.

It is also possible to consider a combination of these processes. Each process has its advantages and disadvantages in respect of capital cost, operating cost, technology, product logistics and marketability, and residual impact on the environment.

The case for elemental sulphur production is attractive with respect to marketability and product logistics, but has the disadvantages of high capital cost and unproven technology. The case for sulphuric acid has the advantage of ‘‘off-the-shelf’’ technology, but low future guarantee of product disposal owing to fluctuating market demands and difficult transport and storage logistics. The case for scrubbing has associated high operating costs and also the drawback of a residual environmental problem owing to leaching of the gypsum tailings dam and other related tailings dam issues.

Hybrid plant configurations incorporating dual acid-gypsum production or dual gypsum-sulphur production have better long-term environmental assurance and profitability. However, in the case of the former the capital cost is very high, and in the case of the latter the technology is still in the piloting stage.

In view of all these uncertainties the recommendation is made that a simple limerock scrubbing system be installed and gypsum stockpiled, with the possibility of future projects to include technologies for sulphur production which are currently commercially unproven.

## **2. Estimate of BCL’s Sulphur Emissions**

The flowsheet in Figure 1 presents a sulphur mass balance round the BCL smelter based on the current smelter concentrate throughput of 960,000 tpa.

In summary: 8.8% of sulphur is captured and fixed in discard slag and matte products, 9.5% leaves via the converters, and 82% leaves via the flash furnace.

The gas volumes from the flash furnace and the converters are similar: roughly 120,000 Nm<sup>3</sup>/h each allowing for false air ingress. Since the off-gas volume greatly influences the capital and operating cost of a sulphur-capture plant, and since the converter gas accounts for only 9.5% of the total sulphur emitted, it would make economic sense to focus on the flash furnace off-gas alone – this practise is also common in many first world operations, e.g. a major nickel producer in Canada also emits converter gases into the atmosphere without sulphur capture.

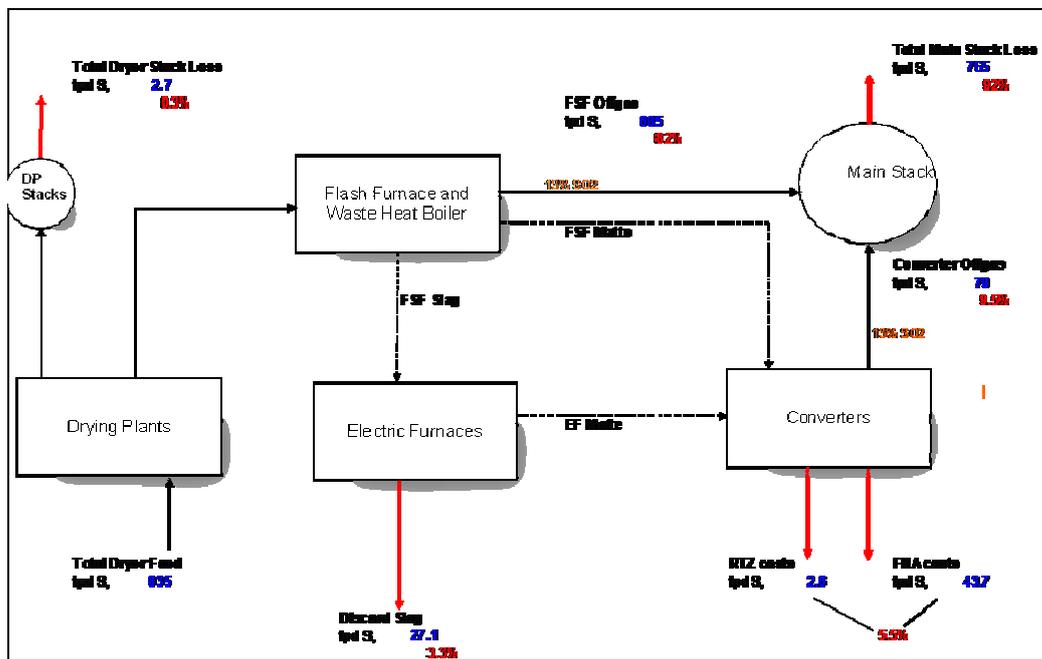


Figure 1: Mass Balance of BCL Smelter Sulphur Department (Based on 960,000 tpa concentrate)

### 3. Overview of Sulphur Markets in RSA and Southern African Region

#### Sulphuric Acid Outlook

A number of major South African companies involved in the production of sulphuric acid and its distribution were approached for their comments.

According to one major distributor and manufacturer, the current independent supply volume of sulphuric acid in South Africa is about 600,000 tonnes per annum, including a 350,000 tpa shortfall, which is filled by sulphur imports [1]. BCL would produce approximately 2000 tpd or 730,000 tpa of acid, or 75% of the total acid demand. However, there are also several regional initiatives to address the current shortfall, which would negatively impact the marketability of a future BCL sulphuric acid. At the time of writing the three new acid plants were in the pre-feasibility planning stage [2]. These plants, however, are all sulphur-burning plants. The opportunity therefore exists for a low-cost producer of primary acid to gain a large market share.

The southern African region could also be supplied, for example Namibia, DRC, Zambia. However, it is anticipated that there would be great difficulty in getting off-take agreements as consumers would want to be self sufficient in their sulphur supply, and long term agreements could not be guaranteed. Road and rail infrastructure inadequacies would add further complexity to this option.

Another disadvantage of producing sulphuric acid is the matter of transport and logistics. It is estimated that BCL's acid production would require 55 rail tankers per day; the South African railway system (Transnet) currently has only 260 tankers in use. A huge investment in new tankers would be required which Transnet may not want to commit to. The issue of the logistics of moving these tankers over long distances also poses a severe problem, e.g. space at sidings and the impact of delays and breakdowns.

### **Elemental Sulphur Outlook**

South Africa imports around 1.5 million tonnes of sulphur per annum [1]. The DRC and Zambia also import large quantities of sulphur. The product is easy to store and can be transported by rail or ship relatively easily. There is generally insufficient sulphur in base metal concentrates or pyrite sources to meet acid demand. Thus, supplying sulphur to the Southern African market is assured in the long term.

## **4. Overview of Sulphur Capture Technologies**

BCL faces a number of options in treating the sulphur in the off-gas:

### *Sulphuric acid*

From a technological point of view, this is the most preferred option, as the technology is well established and can be purchased "off the shelf". The disadvantages of this option are in future acid demand and transport logistics: currently BCL's potential supply would exceed the South African demand. Capital costs are expected to be around P920 million.

### *Sulphur Production*

Technology does exist to produce elemental sulphur commercially, but this is focused mainly in the oil and gas industries, where the feed gas is hydrogen sulphide. In the case of the smelter the sulphur dioxide gas would have to be converted first to hydrogen sulphide. The main breakthroughs in this technology have centred on various catalysts and process controls for the efficient conversion of SO<sub>2</sub> to H<sub>2</sub>S. Two major companies can provide the technology, one based in Australia, and the other in the USA. Capital cost estimates range from P1.1 – 1.5 billion. A further drawback with this option is that large quantities of methane gas are required for the reduction step.

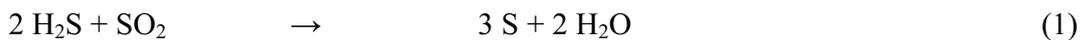
### **4.1 Alkali Scrubbing**

High energy scrubbing towers using various alkali solutions is another possibility, but these are intended primarily for low-strength gases. The most appropriate alkali to use would be calcium carbonate, as it is available close by and is relatively cheap. In this case a discard product of CaSO<sub>4</sub> (gypsum) would be produced which is water insoluble and which may be sold to the cement and gypsum-board industries. The drawback of this option is the high operating costs (2000 tpd of lime would be required). The capital cost of a DynaWave scrubber is of the order of P220 million. This for a single scrubber and excludes ancillary plant costs such as limestone milling, lime slaking, thickening, tailings dam establishment.

#### **4.2 Elemental Sulphur Production**

The production of elemental sulphur is well established in the oil and gas industries. Technology has been developed over the past century to the point where 99.9% of sulphur can be captured and converted to saleable industrial-grade elemental sulphur. In the 1970's there was a sulphur-producing plant, utilizing pulverized coal as reductant, operating at BCL and at Norilsk. However the plants were decommissioned owing to operating difficulties. During the past few decades significant advances have been made in understanding and improving the process. Norilsk is currently in the feasibility stage of installing a modern sulphur plant utilizing methane gas as reductant. A further reason for BCL to consider this technology is the fact that large reserves of coal-bed methane have been discovered in the Kalahari Basin in Botswana, and several foreign companies are poised to develop infrastructure to produce methane gas by 2011 [3].

The process used to produce elemental sulphur is the Claus process, or a modification of it. This process utilizes the H<sub>2</sub>S gas from oil refineries, converts a third of the gas to SO<sub>2</sub> by combustion with air or oxygen-enriched air, and reacts the SO<sub>2</sub> and H<sub>2</sub>S in contact with a catalyst (usually alumina based) to produce elemental sulphur and water according to the following reaction:



The reaction reaches thermodynamic equilibrium, and three or four contact stages are necessary to achieve a maximum recovery of 97%.

Tail gas cleaning systems are employed to recover the last 3% of sulphur. These are amine scrubbers and / or tail gas incinerators.

The difference between the off-gas from oil refineries and that from base metal smelters is that the former is in the form of H<sub>2</sub>S (for which the Claus process has been developed) and the latter is in the form of SO<sub>2</sub>. In order to utilize the Claus process technology, two thirds of the smelter SO<sub>2</sub> needs to be converted to H<sub>2</sub>S. Current research and development is focused on different ways of producing the H<sub>2</sub>S for further reaction in the Claus process. Various technologies have been developed using proprietary catalysts, but no smelter-specific industrial-scale process has yet been commissioned. The most significant example of a possible commercial installation is the Norilsk project [4].

Two companies are currently at the forefront of this technology, an Australian company and a USA based company. The Australian company offers a thermal reduction process and a novel CS<sub>2</sub> route to produce H<sub>2</sub>S. The American company offers various versions of the thermal reduction process, including a direct reduction process using a cutting-edge proprietary catalyst.

Processes for producing sulphur have failed for several reasons:

- Catalyst poisoning by SO<sub>3</sub> and oxygen
- Catalyst bed clogging by soot

- Poor temperature control in the highly exothermic reduction process
- Generation of unwanted side reactions
- Low recoveries.

These limitations have since been eliminated by a better understanding of the chemistry involved, and better catalysts and improved process control.

#### **4.2.1 Sulphur Production via a CS<sub>2</sub> Intermediate (RSR Process)**

Named after its inventor, Mahin Rameshni, this process utilizes existing industrial technology for the production of CS<sub>2</sub> to produce H<sub>2</sub>S via a CS<sub>2</sub> intermediate [4].

Pure elemental sulphur is catalytically reacted with methane to produce H<sub>2</sub>S and CS<sub>2</sub> according to the following endothermic reactions:



The process flowsheet is shown in figure 2 below.

##### *Process Description*

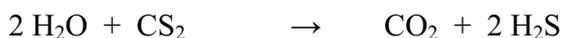
Methane and liquid sulphur are separately heated to 650 °C in an externally fired tubular heater. The methane and sulphur vapour are then mixed in a venturi nozzle and the combined stream flows through the remainder of the tubular heater, where the following reaction takes place:



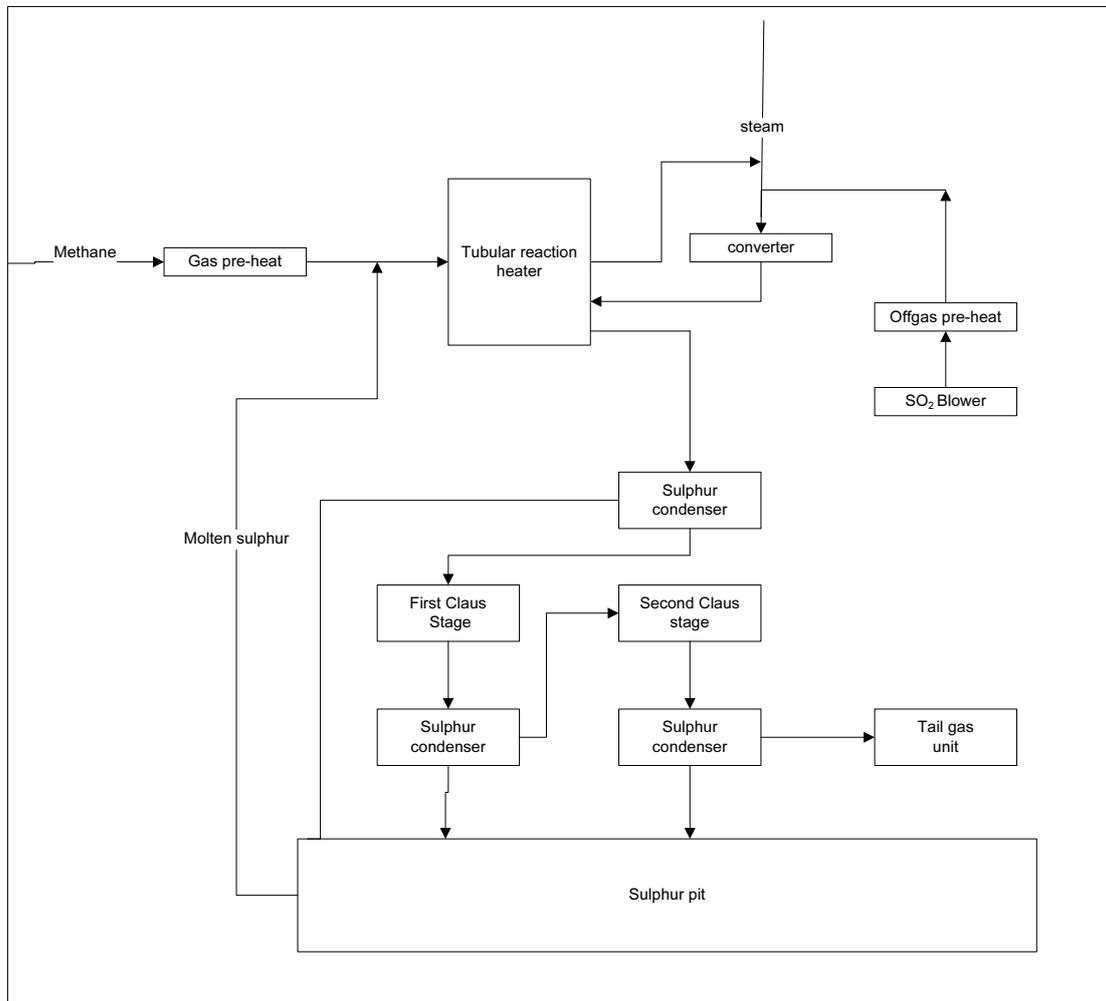
The gases then enter a catalytic converter where the H<sub>2</sub>S reacts with SO<sub>2</sub> according to the Claus reaction:



Steam is also injected into the reactor to hydrolyse CS<sub>2</sub>:



The gases leaving the reactor contain no CS<sub>2</sub>.



**Figure 2: RSR Process**

The gas leaving the reactor is cooled to 150 °C to condense sulphur before routing to two Claus stages. Residual H<sub>2</sub>S in the tail gas from the last Claus stage is oxidized at 800 °C by combustion with methane in excess air in a forced draught incinerator followed by cooling in a waste-heat boiler.

#### **4.2.2 Thermal Process for Sulphur Production**

The process combusts methane with oxygen under sub-stoichiometric conditions to produce H<sub>2</sub> and CO via a series of reactions. SO<sub>2</sub> is partially hydrogenated to H<sub>2</sub>S in the furnace and also subsequently in the hydrogenation reactor [4]. H<sub>2</sub>S is also partially reduced to sulphur in the furnace, and then finally reacted in one or more Claus stages.

*Process Description*

Methane, oxygen and SO<sub>2</sub> are fed to a reaction furnace where the gas is combusted sub-stoichiometrically at 1540 °C to produce CO and H<sub>2</sub>. At these temperatures SO<sub>2</sub> will be partially hydrogenated to H<sub>2</sub>S, and some H<sub>2</sub>S will be reduced to sulphur by the Claus reaction. The reactions are as follows:



The rates of SO<sub>2</sub> and O<sub>2</sub> addition are adjusted so that the H<sub>2</sub>S-SO<sub>2</sub> ratio at the exit of the reaction furnace is 2:1. Several intermediate products such as COS and CS<sub>2</sub> are formed but are hydrolysed to H<sub>2</sub>S in the presence of H<sub>2</sub>O formed by the SO<sub>2</sub> hydrogenation reaction.

CO does not react directly but is hydrolysed to H<sub>2</sub> by the water –gas shift reaction.

From the reaction furnace the gases are cooled in a waste-heat boiler (WHB), which generates high-pressure steam, and are routed to a condenser where sulphur vapour is condensed and sent to a sulphur pit.

The gas is then heated by contacting with WHB bypass gas before entering the hydrogenation reactor containing a Co-Moly catalyst, where SO<sub>2</sub> is hydrogenated as follows:



Some Claus reaction also occurs:



In order to minimise the back-hydrogenation of S to H<sub>2</sub>S, the H<sub>2</sub> concentration must be minimised by carefully maximising the SO<sub>2</sub> hydrogenation above.

The reaction to elemental sulphur is completed in one or more Claus stages.

As in the RSR process, the tail gas enters a methane fired incinerator and waste heat boiler.

Both the above processes are technically viable. Both can achieve similar recoveries and consume similar quantities of methane. The advantage of the former is that the reduction stage is already proven and is a commercially established technology.

#### **4.2.3 Alternative Thermal Process (American Based Company)**

Two different technologies are offered, one with and the other without SO<sub>2</sub> pre-concentration steps. The first is basically a thermal reduction process similar to the process outlined above, although there are flowsheet differences, e.g. there is an oxygen elimination step prior to the hydrogenation step, as well as a purification step prior to the Claus stage which are not included in the thermal process described above [5]. This technology is based on a direct-reduction stage and has no large scale commercial application as yet.

The reduction and hydrogenation chemistry is exactly the same as for the earlier process, although catalysts will have some proprietary differences.

The oxygen elimination step is necessary to prevent poisoning of the hydrogenation catalyst. The chemistry is as follows:



The purification step is not specified in the literature but is probably associated with the elimination of soot, to which the Claus Co-Moly catalyst is susceptible. Using a clean, low-carbon reducing gas such as methane will greatly reduce the likelihood of soot formation.

#### **4.2.4 SO<sub>2</sub> Pre-Concentration**

This process can be used on its own to capture sulphur, or as a conditioning stage for other sulphur capture plants. It involves selectively absorbing SO<sub>2</sub> from the off-gas using an amine absorbent. The plant accounts for a major portion of the overall sulphur-recovery plant equipment, operating and capital cost. However, the advantages of pre-concentration are numerous, and include the following:

- Reduced capital and operating costs (fuel)
- Improved Claus conversion efficiency
- Elimination of sulphur and particulate contamination of the Claus catalyst
- Reduced catalyst deactivation from contamination from sulphation due to oxygen
- Improved operating control owing to constant SO<sub>2</sub> strength
- Uninterrupted Claus operation during temporary reaction furnace or other outages.

#### **4.3 Sulphur production via gypsum intermediate**

A novel process at pilot plant stage of development was described by J. Maree and colleagues [6]. In this process smelter off-gas is scrubbed with limerock slurry to produce gypsum, which is filtered, dried, and roasted under reducing conditions to produce calcium sulphide. The calcium sulphide is then reacted under acidic aqueous conditions to produce calcium carbonate and hydrogen sulphide gas. The hydrogen sulphide gas is processed to elemental sulphur via a modified Claus process (PIPco process), and the

calcium carbonate is recirculated to the inlet gas scrubber. At the time of writing this process was at pilot-plant stage.

#### **4.4 Sulphuric Acid Manufacture**

The process and chemistry are well established, and the technology is virtually generic, with the feed gases being conditioned so as to conform to the acid-plant requirements. This conditioning consists of the following stages [7]:

- Adjustment of the gas SO<sub>2</sub> strength to the required range (8-12%) by sulphur burning or air dilution
- Removal of particulates (0.001 to 0.01 g/Nm<sup>3</sup>)
- Removal of H<sub>2</sub>O by condensation
- Gas drying with a strong portion of sulphuric acid (less than 50mg/Nm<sup>3</sup> moisture)

After conditioning the SO<sub>2</sub> is reacted catalytically with oxygen to produce SO<sub>3</sub>:



The catalyst is molten V, K, Na, Cs, S, O solution on a porous silica substrate. Temperature control is critical as the reaction equilibrium becomes unfavourable with increasing temperature.

Following the conversion of SO<sub>2</sub> to SO<sub>3</sub>, the SO<sub>3</sub> is absorbed by H<sub>2</sub>O contained in strong sulphuric acid (98.5%).

Modern acid plants are manufactured as single-contact or double-contact plants. The term refers to the number of H<sub>2</sub>SO<sub>4</sub>-making steps in the process. A single contact plant can typically convert 99% of the SO<sub>2</sub>, whereas a double contact plant can convert 99.9% of the SO<sub>2</sub>.

Stage conversion drops off dramatically as the third stage is passed on the SO<sub>2</sub>-conversion equilibrium curve. This problem is overcome by introducing an intermediate acid-making stage before a fourth catalyst bed. The feed gas to the fourth stage has been scrubbed free of SO<sub>3</sub>, allowing the SO<sub>2</sub> oxidation step to go virtually to completion, and an overall 99.99% efficiency to be attained.

#### **4.5 Scrubbing Systems**

Various scrubbing systems to capture SO<sub>2</sub> emissions are in operation in industry. These range from once-through lime- or limestone- slurry systems to various dual alkali systems. The problems with these systems are that

- They are not as efficient as the above processes (90-97% recovery)
- They function best with feed gas of low SO<sub>2</sub> strength (less than 6% SO<sub>2</sub>)
- They have very high operating costs, mainly associated with chemicals
- There is the residual environmental impact associated with the dumped waste product

Two scrubbing systems are options for the BCL off-gas: a limestone process and a lime-sodium dual-alkali process. The limestone process is more suitable for lower strength gases, but could be more efficient in a high-intensity scrubber, or in staged scrubbers. The dual alkali process is more suitable for high-strength gas but has a more complex associated plant.

#### **4.5.1 Limestone Process**

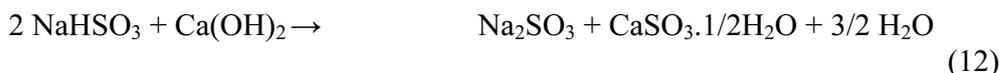
The chemistry for the lime process is:



Thus one mole of CaCO<sub>3</sub> is required for each mole of SO<sub>2</sub>, or 3.1 tonnes limerock/tonne sulphur.

At an estimated cost for mining, purchase and transport of P150/tonne limestone, the total monthly cost is of the order of P10m (USD1.4m)

#### **4.5.2 Lime-Sodium process**



Thus the waste product is gypsum, and a strong scrubbing caustic solution is recovered. This process is expected to be much more expensive to operate than the limestone process owing to the need for large quantities of hydrated lime, a refined and therefore much more expensive commodity than limestone.

From the above equations, one mole of Ca(OH)<sub>2</sub> is required for each mole of SO<sub>2</sub>, or 2.3t Ca(OH)<sub>2</sub> /t sulphur. Slaked lime is available from South Africa at an estimated cost of P1500/t including transport. Thus monthly operating costs would be of the order of P78m (USD11.1m).

### **4.5.3 Scrubbing Equipment**

Various types of scrubbers can be used for this process. The most efficient is the proprietary DynaWave wet scrubber, which incorporates particulate removal, quenching and absorption, all in one unit. The scrubber uses a unique principle of a reverse jet flow of liquid where the countercurrent gas flow is contacted in a high intensity froth zone for maximum mass transfer rates.

## **5. Alternative Plant Configurations**

### **5.1 Hybrid Alternative 1: Acid Plant and Scrubber in Parallel**

An option for consideration would be to build a smaller acid plant and a limerock scrubber run in parallel. Sulphuric acid would then be produced to satisfy market demands, and the excess sulphur would be scrubbed and dumped as gypsum. Although the capital and operating costs are high for this option, the investment can be recovered, whilst long-term environmental standards are complied with. Two scrubbers could be placed in parallel, both acting as conditioning units for the acid plant under full acid production conditions, and switched to dual acid-gypsum production when the acid market slumps.

### **5.2 Hybrid Alternative 2: Limerock Scrubber and Future Sulphur Process via Gypsum Intermediate**

A South African company is currently piloting a process that recovers elemental sulphur from smelter and power-plant off-gases [6]. The process incorporates a gypsum reduction roast to produce CaS, a sulphur stripping step with CO<sub>2</sub> to produce CaCO<sub>3</sub> and H<sub>2</sub>S, and a modified Claus process called the PIPco process to produce elemental sulphur from H<sub>2</sub>S and SO<sub>2</sub>. If this process becomes commercially viable, it would be the preferred option, as it has lower capital and operating costs than the other options, and the costs associated with limerock would be favourably offset by the sale of elemental sulphur.

A limerock scrubber could be installed and gypsum stockpiled. Future consideration can then be given to the gypsum- intermediate sulphur-recovery process once the technology matures.

### **5.3 Hybrid Alternative 3: Limerock Scrubber and Future RSR Sulphur Process**

The commercialization of the Australian / American sulphur recovery technology is imminent with the Norilsk project coming on line. It would be wise to wait and see how it performs. In the meantime a scrubber could be installed to partially address the environmental problem in the short to medium term. Once a future process is decided upon the scrubber could still be used as a conditioning unit upstream of the sulphur capture plant (whichever technology is chosen), and the capital would not have been wasted.

#### 5.4 Other systems

There are other systems for capturing SO<sub>2</sub> from smelter off-gases, but they are not suitable for the BCL case, and are mentioned briefly for interest.:

- Amine absorption and regeneration, eg CanSolv
- Organic scrubbing
- WSA (wet gas sulphuric acid)

#### 6. Cost Comparison

Table 1 lists estimates of the capital costs for the various sulphur capture options.

Table 1: Summary of Cost and Performance Data

Case No		Capital Millions USD	Operating Cost, MUSD Per Month	Product	Product Disposal	Recovery	Profit scale	Technology Risk
1	<b>RSR Sulphur Plant</b>	157	4.6	Sulphur	Sales	90%	High	Med
2	<b>Thermal Sulphur Plant</b>	171	4.0	Sulphur	Sales	90%	High	High
3	<b>Alternate Thermal Sulphur Plant</b>	198	4.0	Sulphur	Sales	97%	High	High
4	<b>Double Contact Acid Plant</b>	131	0.14	H <sub>2</sub> SO <sub>4</sub>	Sales	99.9%	Low-Med	Low
5	<b>Limerock scrubbing (dual scrubbers in series / parallel)</b>	57	3.0	CaSO <sub>4</sub>	Stockpile	90%	Negative	Low
6	<b>Dual Alkali Scrubbing</b>	31	11.1	CaSO <sub>4</sub>	Stockpile	99%	Negative	Low
7	<b>1000 tpd AP and scrubber</b>	146	1.7	H <sub>2</sub> SO <sub>4</sub> /CaSO <sub>4</sub>	Stockpile/Sales	90 – 99.9	Low	Low
8	<b>Dual scrubber and “gypsum” sulphur recovery plant</b>	114	1.7	Sulphur	Stockpile/Sales	90%	High	High
9	<b>Scrubber and RSR process</b>	203	6.0	Sulphur	Stockpile/Sales	90%	Medi	Med

#### 7. Conclusions

- Technologically the most acceptable solution is a sulphuric acid plant. However, market conditions and logistics in the southern African region preclude this as a solution in its own right.
- Elemental sulphur production offers the most stable source of revenue and a guarantee of product disposal. However, although imminent, the technology is not yet commercially proven.

- Limerock or dual alkali scrubbing systems offer a short-to-medium-term solution, but have very high associated operating costs, as well as residual environmental issues.
- Hybrid plants incorporating scrubbing systems and a smaller acid plant are a means of achieving environmental conformance whilst having the opportunity to recover capital costs.
- Hybrid plants incorporating scrubbing systems and some future sulphur-capture plant, such as the RSR or the gypsum-intermediate process, are a means of ensuring capital payback and future revenue gains, provided the technology becomes commercially viable.

### **Acknowledgements**

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