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An Economic Analysis of Microbial Reduction of Sulfur Dioxide as a Means of Byproduct Recovery from Regenerable Processes for Flue Gas Desulfurization

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Index Entries: Sulfur dioxide; flue gas desulfurization; *Desulfovibrio desulfuricans;* microbial reduction.

INTRODUCTION

We have previously demonstrated that the sulfate-reducing bacterium, *Desulfovibrio desulfuricans,* may be grown in mixed culture with fermentative heterotrophs in a medium in which glucose served as the only carbon source. Beneficial cross-feeding resulted in vigorous growth of *D. desulfuricans,* which used SO_2 (1% SO_2, 5% CO_2, balance N_2) as a terminal electron acceptor with complete reduction of SO_2 to H_2S with only 1–2 s of contact time. Sulfate-reducing bacteria (SRB) cannot use simple sugars (such as glucose) as carbon and energy sources. However, the fermentative heterotrophs that developed in these cultures as a result of septic operation utilized glucose and produced fermentative end products (ethanol and lactic acid), which served as carbon and energy sources for *D. desulfuricans.* Sulfate-reducing bacteria are also strict anaerobes; mere exclusion of oxygen is not sufficient to support growth of pure cultures. Redox-poising agents are generally required to maintain a redox potential in the medium of −150 to −250 mV. However, in *D. desulfuricans* working cultures, no redox-poising agents were required (*1,2*).

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We have proposed that the concentrated SO_2 stream obtained from certain regenerable processes for flue gas desulfurization, such as the copper oxide process (*3*), may be split with two-thirds of the SO_2 reduced to H_2S by contact with a culture of sulfate-reducing bacteria (*1*). The resulting H_2S may then be combined with the remaining SO_2 and used as feed to a Claus reactor to produce elemental sulfur (*4,5*). An economic evaluation of this microbial SO_2 reduction process concept has been completed comparing microbial SO_2 reduction to conventional catalytic SO_2 hydrogenation (with H_2 generation from methane). Process flow sheets, material and energy balances, and capital investment and production cost estimates have been prepared for each process evaluated.

The chemical gas-phase reduction of SO_2 to H_2S proceeds according to Eq. (1):

$$SO_2\,(g) + 3\,H_2\,(g) \xrightarrow{\text{catalyst}} H_2S\,(g) + 2\,H_2O\,(g) \qquad (1)$$

Hydrogen (H_2) required for the chemical reduction of SO_2 is produced via conventional, gas-phase steam/methane reforming and water-gas shift techniques according to Eq. (2) and (3):

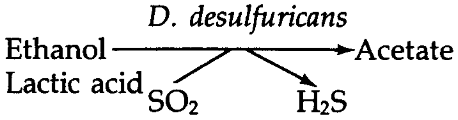
$$CH_4\,(g) + H_2O\,(g) \overset{\text{catalyst}}{\rightleftharpoons} CO\,(g) + 3\,H_2\,(g) \qquad (2)$$

$$CO\,(g) + H_2O\,(g) \overset{\text{catalyst}}{\rightleftharpoons} CO_2\,(g) + H_2\,(g) \qquad (3)$$

Therefore, for conventional, chemical SO_2 reduction, both an SO_2 reduction reactor and an external hydrogen generation system are required.

The liquid phase, microbial reduction of SO_2 to H_2S is considerably more complex, since glucose serves as an ultimate source of carbon and reducing equivalents (for SO_2 reduction) for *D. desulfuricans*. The pertinent reactions are given in Eq. (4) and (5):

$$\text{Glucose} \xrightarrow[\substack{\text{nonSRB} \\ \text{heterotrophs}}]{\text{fermentative}} \text{Ethanol} + \text{Lactic acid} \qquad (4)$$



$$\begin{matrix}\text{Ethanol} \\ \text{Lactic acid}\end{matrix} \xrightarrow[SO_2 \quad\quad H_2S]{\textit{D. desulfuricans}} \text{Acetate} \qquad (5)$$

The microbial reduction system functions essentially as both an H_2 generator and SO_2-reduction reactor. This negates the requirement for an external H_2 (or reducing equivalent) production system compared to conventional SO_2 reduction. Anticipating the possible incorporation of acetate utilizing SRB into the process culture, for the purpose of the economic

Table 1
Process Design Basis For SO_2 Reduction Process

Design Basis Parameter	
Flue Gas Source	Coal Fired Power Plant
Power Plant Capacity, MW_e	1,000
Ultimate Feed Coal Analysis C/H/O/N/S, wt%	78.7/5.5/10.9/1.4/3.5
Ultimate Coal Heating Value, Btu/lb	14,130
Feed Gas Source for SO_2 Reduction	Regenerator Off-Gas from Copper Oxide Process (90% SO_2/NO_x Removal)
Reduction Feed Gas Rate, mol/hr	2,115 (stream 7 @ 82,279 lb/hr)
Reduction Feed Gas Composition $SO_2/CO_2/H_2O/CH_4$, mol %	33/22/44/1
Ultimate Product Gas for Comparison	Balanced H_2S/SO_2 Feed Gas Claus Unit
Ultimate Product Gas H_2S/SO_2 mol Ratio	2.0

evaluation, it was assumed that the SO_2/glucose stoichiometry was equal to the theoretical maximum of 4.0.

DESIGN BASIS AND DESCRIPTION

Process Design Basis

Since the process/economic comparison presented here is intended to focus on the techno-economic differences between the two SO_2-reduction methodologies, all economics were based on a common feed stock source, composition, and rate, and also on a common ultimate product gas H_2S/SO_2 mol ratio. The process design basis used for both SO_2-reduction methodologies is given in Table 1. The SO_2 regenerated flue gas was assumed to be produced by processing the raw flue gas generated by coal-fired utility boilers (1000 MW_e) through a dry, regenerative fluidized bed, copper-oxide-type flue gas desulfurization system. The product of both the microbial and conventional SO_2-reduction system was assumed to be balanced gas ($H_2S/SO_2 = 2$) suitable as a feed stock for a Claus sulfur recovery system.

Process Description—Microbial SO_2 Reduction

The overall process flow diagrams for the microbial SO_2-reduction process are shown in Figs. 1 and 2. In the initial steps (*see* Fig. 1), the SO_2-regenerated feed flue gas (stream 7 at point A) is cooled in two stages and then partially heat exchanged (HE-103) against cooler SO_2-reduction bioreactor effluent gas (stream 13). Approximately one-third (stream 8) of the feed gas is diverted to the Claus feed gas compressor (GC-102) and sent

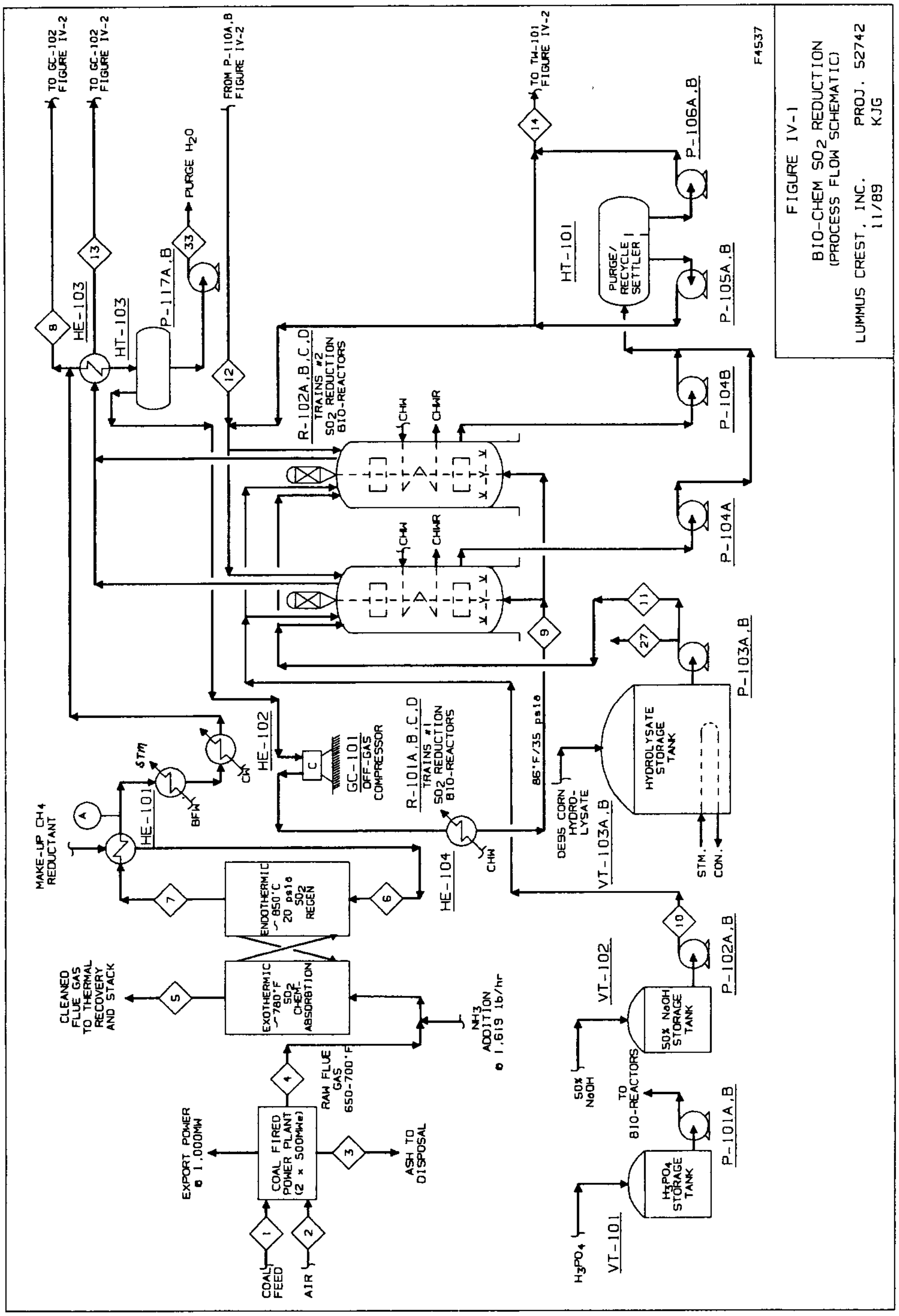


FIGURE IV-1
BIO-CHEM SO_2 REDUCTION
(PROCESS FLOW SCHEMATIC)
LUMMUS CREST, INC. PROJ. 52742
11/89 KJG
F4537
COAL FEED
AIR
COAL FIRED POWER PLANT (2 × 500MWe)
EXPORT POWER @ 1,000MW
ASH TO DISPOSAL
RAW FLUE GAS 650-700°F
EXOTHERMIC ~780°F SO_2 CHEM. ABSORBTION
ENDOTHERMIC ~850°C 20 psia SO_2 REGEN
CLEANED FLUE GAS TO THERMAL RECOVERY AND STACK
NH_3 ADDITION @ 1,619 lb/hr
MAKE-UP CH_4 REDUCTANT
HE-101
HE-102
HE-103
HE-104
STM
BFW
CW
CHW
GC-101 OFF-GAS COMPRESSOR
R-101A,B,C,D TRAINS #1 SO_2 REDUCTION BIO-REACTORS
R-102A,B,C,D TRAINS #2 SO_2 REDUCTION BIO-REACTORS
CHWR
86°F/35 psia
HT-103
P-117A,B
PURGE H_2O
TO GC-102 FIGURE IV-2
TO GC-102 FIGURE IV-2
FROM P-110A,B FIGURE IV-2
TO TH-101 FIGURE IV-2
HT-101
PURGE/ RECYCLE SETTLER
P-104A
P-104B
P-105A,B
P-106A,B
P-103A,B
HYDROLYSATE STORAGE TANK
DEB95 CORN HYDROLYSATE
VT-103A,B
STM.
CON.
VT-102
50% NaOH STORAGE TANK
50% NaOH
P-102A,B
TO BIO-REACTORS
VT-101
H_3PO_4 STORAGE TANK
H_3PO_4
P-101A,B
1
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3
4
5
6
7
8
9
10
11
12
13
14
27
33
A
C

Fig. 1.

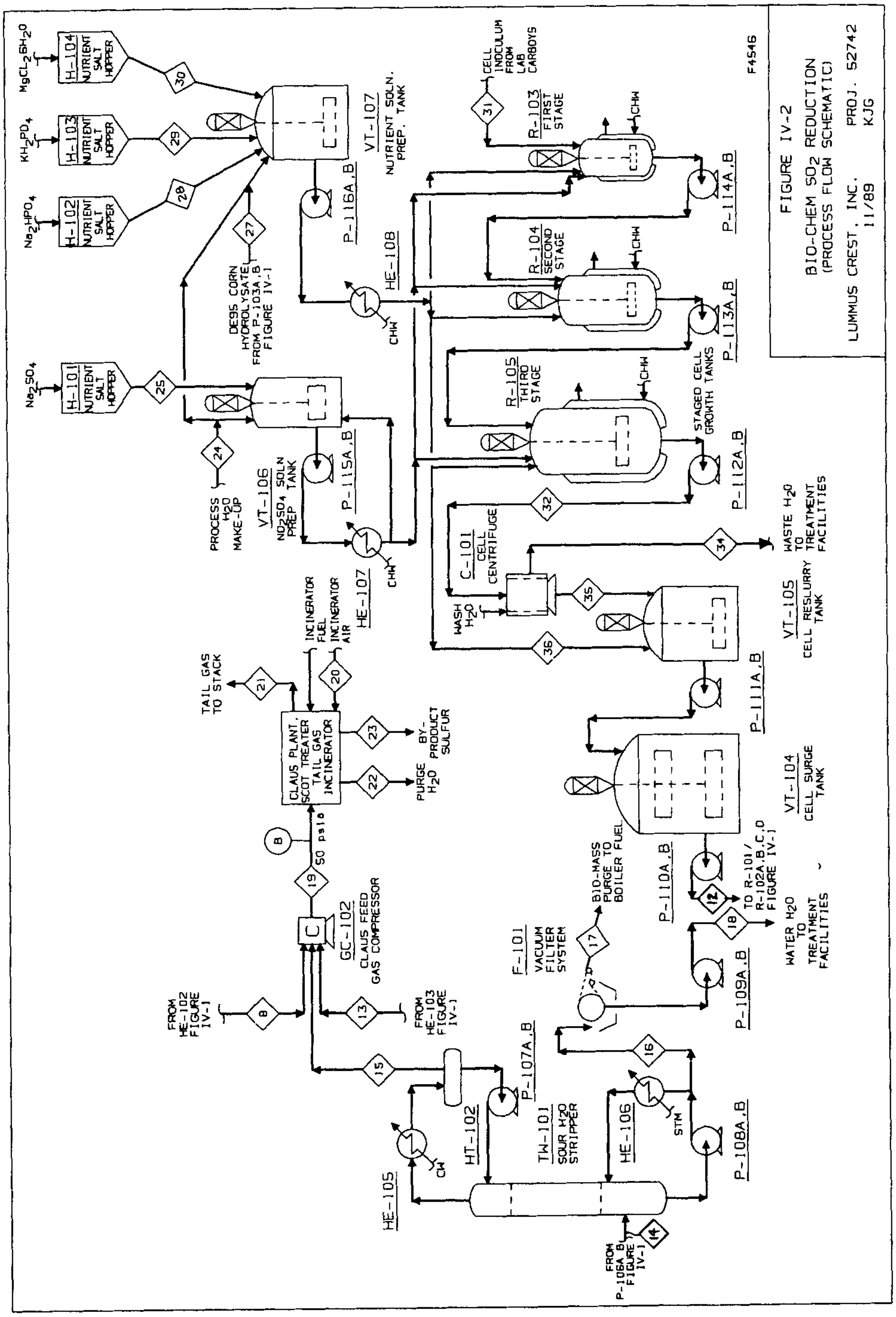
FIGURE IV-2
BIO-CHEM SO_2 REDUCTION
(PROCESS FLOW SCHEMATIC)
LUMMUS CREST, INC. PROJ. 52742
11/89 KJG
F4546
H-101 NUTRIENT SALT HOPPER
H-102 NUTRIENT SALT HOPPER
H-103 NUTRIENT SALT HOPPER
H-104 NUTRIENT SALT HOPPER
Na_2SO_4
Na_2HPO_4
KH_2PO_4
$MgCl_2 6H_2O$
VT-106 Na_2SO_4 SOLN PREP TANK
VT-107 NUTRIENT SOLN. PREP. TANK
PROCESS H_2O MAKE-UP
DEGS CORN HYDROLYSATE FROM P-103A,B FIGURE IV-1
P-115A,B
P-116A,B
HE-107
HE-108
CHW
INCINERATOR FUEL
INCINERATOR AIR
CELL INOCULUM FROM LAB CARBOYS
R-103 FIRST STAGE
R-104 SECOND STAGE
R-105 THIRD STAGE
STAGED CELL GROWTH TANKS
P-112A,B
P-113A,B
P-114A,B
C-101 CELL CENTRIFUGE
WASH H_2O
WASTE H_2O TO TREATMENT FACILITIES
VT-105 CELL RESLURRY TANK
P-111A,B
VT-104 CELL SURGE TANK
P-110A,B
TO R-101/R-102A,B,C,D FIGURE IV-1
BIO-MASS PURGE TO BOILER FUEL
F-101 VACUUM FILTER SYSTEM
WATER H_2O TO TREATMENT FACILITIES
P-109A,B
TAIL GAS TO STACK
CLAUS PLANT, SCOT TREATER TAIL GAS INCINERATOR
BY-PRODUCT SULFUR
PURGE H_2O
50 psig
B
GC-102 CLAUS FEED GAS COMPRESSOR
FROM HE-102 FIGURE IV-1
FROM HE-103 FIGURE IV-1
HE-105
HT-102
P-107A,B
TW-101 SOUR H_2O STRIPPER
HE-106
STM
CW
P-108A,B
FROM P-106A,B FIGURE IV-1
8
12
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14
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16
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Fig. 2.

Table 2
Microbial Reduction Process Design Parameters For SO_2-Reduction Bioreactors[1]

(1,000 MW_e Equivalent Capacity)	
Reactor Type	Agitated stirred tank with internal cooling coil
Reactor Temperature, °C	30
Reactor Pressure In/Out, psia	35/17
Agitation Intensity, HP/1000 gal	2
Total (SRB + non-SRB) cell density, g/liter	25
SRB/non-SRB Cell Wt. Ratio	1/1
Total Cell (SRB + non-SRB) count, cells/liter	6×10^{11}
Individual Cell Weight, g/cell	0.1375×10^{-11}
Desulfuricans Specific Activity, lb mol SO_2/hr-cell	3.72×10^{-17} $\frac{1.69 \text{ mmol } SO_2}{\text{hr-}10^{11} \text{ cells}}$
Total Bioreactor Feed Gas Rate, mol/hr	818.9
Total SO_2 Feed Rate, mol/hr	465.2
SO_2 Conversion to H_2S, % per pass	100^-
Total Reactor(s) Cell (SRB + non-SRB) Inventory, lb	84,720
Total Reactor(s) Oper. Volume, gal	407,000
Total Reactor(s) Design Volume, gal	512,000 (w/20% freeboard)
Number of Reactors Required	8 (in parallel)
Design Volume per Bioreactor, gal	64,000
Reactor Dimensions, ft dia x ft (T/T)	14 x 55
Nutrient Source	DE95 Corn Hydrolysate (68% glucose)
Glucose Consumption, mol glucose/mol SO_2 Reduced	0.25

[1] The total cell density is an estimate. All other aspects of operation of the bioreactors is based on experimental work (1,2).

directly to the Claus plant. The balance of the feed gas, after water condensation, is sent to the off-gas compressor (GC-101), where the bioreactor feed gas (stream 9) is compressed and chilled to about 35 psia and 30°C, respectively, and fed to the SO_2-reduction bioreactor trains (R-101A,B,C,D and R-102A,B,C,D).

In the liquid phase, microbial SO_2-reduction bioreactors, the contained feed SO_2 is selectively reduced (hydrogenated) to H_2S in the presence of *D. desulfuricans* in coculture with mixed fermentative heterotrophs. Glucose (as DE95 corn hydrolysate at 68% glucose) is utilized as the ultimate source of carbon and reducing equivalents for the bioreduction process. Process design parameters for the SO_2-reduction bioreactors are shown in Table 2. Total SO_2 conversion to H_2S is assumed at an estimated glucose consumption of 0.25 mol glucose/mol SO_2 reduced.

Liquid effluent from the bioreactors is pumped (P-104 A/B) to the purge/recycle settler (HT-101), where the aqueous and biomass (solids) rich layers are separated. A portion of the biomass layer is recycled to the bioreduction reactors, while the balance is combined with the aqueous layer and sent (stream 14) to the sour H_2O stripper (TW-101). In the stripper, dissolved acid gases (CO_2, H_2S) are removed overhead (stream 15) and fed to the Claus feed gas compressor (GC-102). Stripper bottoms (stream 16) are then filtered in the vacuum filter system (F-101). Filter waste water (stream 18) is sent to waste H_2O treatment facilities in order to purge the net H_2O make across the bioreduction process, whereas the biomass purge (stream 17) is sent to boiler fuel. The bioreactor effluent gas (stream 13) bypassed feed gas (stream 8), and stripper off-gas (stream 15) are then compressed to about 50 psia in the Claus feed gas compressor (GC-102) and sent (stream 19 at point B) to the Claus plant for conversion of the contained H_2S and SO_2 into elemental sulfur. The H_2S/SO_2 mol ratio in the Claus feed gas (stream 19) is 2/1 molar.

Supporting auxiliary equipment (*see* Fig. 2) includes facilities for nutrient and Na_2SO_4 (used as terminal electron acceptor during start-up) solution preparation, cell growth, and cell isolation/reslurrying. These are required to make up for cell losses (stream 17) from the bioreactor system and start-up. Conventional cell growth and isolation techniques are utilized in these facilities.

Process Description—Conventional SO_2 Reduction

The overall process block flow diagram for the conventional SO_2 reduction system is shown in Fig. 3. The process design was based on the utilization of conventional-type process modules supplemented with reasonable engineering assumptions and process analogies.

In the initial step (*see* Fig. 3), the SO_2-regenerated feed flue gas (stream 7 at point A) is combined with make-up hydrogen (stream 9) and fed (stream 10) to a conventional, catalytic vapor phase, SO_2-hydrogenation reactor system. The hydrogen feed (stream 9) is produced in an integrated, conventional hydrogen plant based on steam/methane reforming technology.

The SO_2 hydrogenation conditions were selected to convert about two-thirds of the SO_2 feed to H_2S. Hydrogenation reactor effluent (stream 11 at point B), after thermal recovery and residual compression, is sent to a conventional Claus plant for conversion of the contained H_2S and SO_2 into elemental sulfur. The H_2S/SO_2 mol ratio in the Claus feed gas (stream 11) is 2/1 molar.

PROCESS ECONOMICS

Total fixed investments and annual costs of production estimates for both the microbial and conventional SO_2 reduction processes are given in Table 3. As seen in Table 3, the fixed capital investments for the two pro-

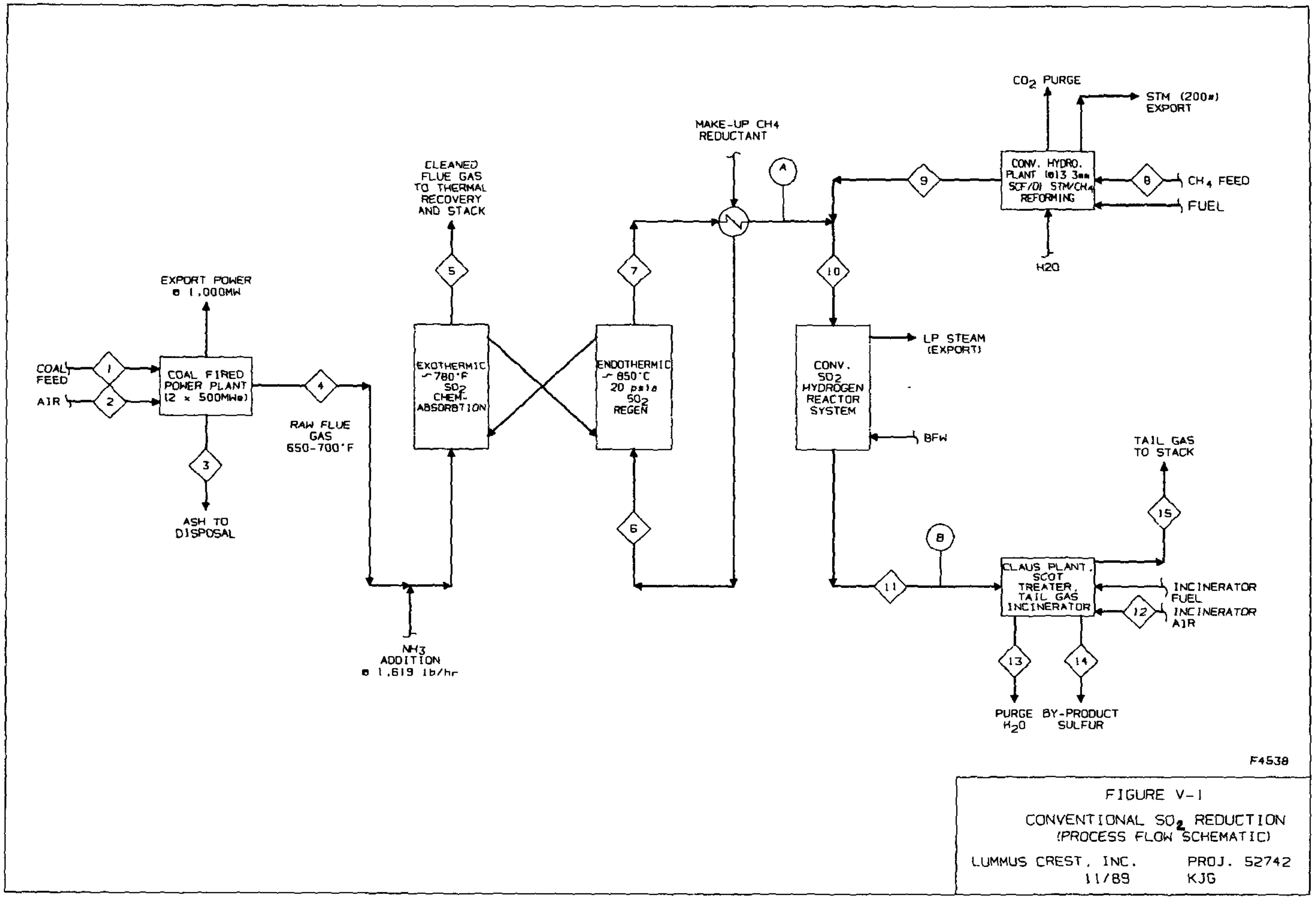
COAL FEED
AIR
1
2
COAL FIRED POWER PLANT (2 × 500MWe)
EXPORT POWER @ 1,000MW
3
ASH TO DISPOSAL
4
RAW FLUE GAS 650-700°F
NH3 ADDITION @ 1,619 lb/hr
EXOTHERMIC ~780°F SO2 CHEM-ABSORBTION
5
CLEANED FLUE GAS TO THERMAL RECOVERY AND STACK
ENDOTHERMIC ~850°C 20 psia SO2 REGEN
6
7
MAKE-UP CH4 REDUCTANT
A
10
CONV. SO2 HYDROGEN REACTOR SYSTEM
LP STEAM (EXPORT)
BFW
9
CONV. HYDRO. PLANT (@13.3mm SCF/D) STM/CH4 REFORMING
CO2 PURGE
STM (200#) EXPORT
8
CH4 FEED
FUEL
H2O
11
B
CLAUS PLANT, SCOT TREATER, TAIL GAS INCINERATOR
15
TAIL GAS TO STACK
INCINERATOR FUEL
12
INCINERATOR AIR
13
14
PURGE H2O
BY-PRODUCT SULFUR
F4538
FIGURE V-1
CONVENTIONAL SO2 REDUCTION (PROCESS FLOW SCHEMATIC)
LUMMUS CREST, INC. 11/89
PROJ. 52742 KJG

Fig. 3.

Table 3
Comparative Production Cost Summary
For Microbial and Conventional SO_2-Reduction Processes

(1,000 MW_e Equivalent Capacity)

Investment, $MM (3rd Qtr. 1989)	Microbial SO_2-Reduction	Conventional SO_2-Reduction
Inside Battery Limits (ISBL)	19.75[1]	19.25[2]
Outside Battery Limits (OSBL) @ 30% ISBL	5.92	5.78
Total Fixed (TFI)	25.67	25.03
Production Cost (see Tables 4 and 5)	$MM/yr	
Raw Materials[3]	23.60	3.55
Utilities	3.21	(0.91)
Labor, For.,Supvr. (L,F,S)	0.69	0.63
Maintenance, Material and Labor @ 4% ISBL	0.79	0.77
Direct Overhead @ 45% L,F,S	0.31	0.28
General Plant Overhead @ 65% Oper. Cost (L,F,S&M,M,L)	0.96	0.91
Insurance, property taxes @ 1.5% TFI	0.38	0.38
By-Product Credit/Debit	0	excl.[4]
Cash Cost of Production	29.94	5.61

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[1] A detailed equipment list was generated and the purchased cost of individual equipment items was estimated utilizing Lummus Crest's own internal, equipment cost data base and also selected vendor quotes. Purchased major equipment costs (3rd Qtr 1989) totalled $5,286 MM plus 10% contingency to yield grand total purchased equipment cost of $5.81 MM. ISBL was then obtained by multiplying grand total purchased equipment cost by a composite equipment installation factor of 3.4 to obtain ISBL of $19.75 MM. Incremental OSBL was estimated at 30% ISBL ($5.92 MM) for an existing power plant site. Excludes royalty/licensing fee, land, initial chemical charges and interest during construction.

[2] From a basic material balance, the capacity of a conventional type steam/CH_4 reforming hydrogen plant module was calculated to be about 14 MM SCFD. Based on internal Lummus Crest capital cost data files and also published data from various licensors (e.g. M. W. Kellogg, Haldor-Topsoe, ICI, Lurgi, etc.) the installed cost (ISBL) for the hydrogen plant module was estimated at $13.70 MM. For the conventional SO_2 hydrogenation reaction system module, the ISBL was estimated to be $3.8 MM per analogy with Shell's SCOT treater system. A contingency of 10% was added to yield a grand total ISBL of $19.25 MM (6,7).

[3] Assumes zero value for feed flue gas.

[4] Waste water treatment costs excluded.

Table 4
Microbial SO_2-Reduction Process Summary
of Raw Materials, Utilities, and Labor Costs

(@ 8760 hr/yr; 1000 MW_e Equivalent Capacity)

1.	RAW MATERIALS	Price $/Unit	Annual Cost, $MM/yr (1989)
	DE95 Corn Hydrolysate @ 31,923 lb/hr	0.082	22.93
	50% NaOH @ 828 lb/hr	0.060	0.44
	H_3PO_4 (estimated)	--	0.11
	Na_2SO_4 @ 39 lb/hr	0.045	0.02
	Na_2HPO_4 @ 37 lb/hr	0.030	0.01
	KH_2PO_4 @ 55 lb/hr	0.030	0.01
	$MgCl_2\cdot 6H_2O$ @ 90 lb/hr	0.100	0.08
	Total Raw Materials Cost		23.60
2.	UTILITIES	Price $/Unit	Annual Cost $MM/yr (1989)
	Power @ 8465 KW[1]	0.040	2.97
	Cooling Water @ 72 Mgal/hr	0.073	0.05
	Steam (200 psig) @ 4.9 Mlb/hr (coal fired)	4.260	0.18
	Process Water @ 1 Mgal/hr	0.770	0.01
	Total Utilities Cost		3.21
3.	LABOR	Price $/man-yr	Annual Cost $MM/yr (1989)
	Operators @ 18 men	28,700	0.52
	Foremen @ 4 men	32,500	0.13
	Supervision @ 1 man	39,300	0.04
	Total Labor Cost		0.69

[1] Includes 5260 tons refrigeration @ 1.12 KW/ton (CHW @ 50°F supply).

cesses are essentially equivalent; however, the production costs for the microbial process are much higher than that of the conventional SO_2-reduction process, primarily because of the high cost of raw materials in the microbial process (*see* Table 4). With zero value assigned to the SO_2-regenerated feed flue gas for both processes considered, the large raw materials cost advantage for the conventional process is almost totally attributable to the utilization of natural gas (10,300 lb/h at $0.035/lb) as a hydrogen feed stock source as compared to DE95 glucose hydrolysate (31,900 lb/h at $0.082/lb) as hydrogen or reducing equivalent feed stock source for the microbial reduction process. Utilities costs are lower for the conventional route (*see* Table 5), since the hydrogenation reaction and steam/methane reforming systems operate at considerably higher temperatures, allowing for the recovery (as steam) of the reduction reaction exotherm plus additional waste heat recovery.

Table 5
Conventional SO_2-Reduction Process Summary
of Raw Materials, Utilities, and Labor Costs

(@ 8760 hr/yr; 1000 MW_e Equivalent Capacity)

1.	RAW MATERIALS	Price $/Unit	Annual Cost $MM/yr (1989)
	Natural Gas (CH_4) @ 10,282 lb/hr	0.035	3.15
	H_2 Plant (reforming + shift) Catalysts	--	0.10
	SO_2 Hydrogenation Catalyst	--	0.25*
	Miscellaneous Chemicals	--	0.05
	Total Raw Materials Cost		3.55
	*estimated		
2.	UTILITIES	Price $/Unit	Annual Cost $MM/yr (1989)
	Power @ 680 KW	0.040	0.24
	Cooling Water @ 148 Mgal/hr	0.073	0.09
	Boiler Feed Water @ 11 Mgal/hr	1.310	0.13
	Fuel @ 17 MMBtu/hr	1.600	0.24
	Steam (200 psig) @ (57.9) Mlb/hr-export	3.18 (exgas)	(1.61)
	Total Utilities Cost		(0.91)
3.	LABOR	Price $/man-yr	Annual Cost $MM/yr (1989)
	Operators @ 16 men	28,700	0.46
	Foreman @ 4 men	32,500	0.13
	Supervision @ 1 man	39,300	0.04
	Total Labor Cost		0.63

CONCLUSIONS

In summary, it appears that the microbial reduction process is clearly not competitive with conventional SO_2-reduction techniques when glucose (DE95 corn hydrolysate) is utilized as the feedstock hydrogen source. Other and indeed considerably cheaper feedstock (carbon and energy sources) will be required to make the SO_2-bioreduction process competitive with conventional SO_2-reduction techniques. Cheaper feedstocks have been identified (sewage sludge and CO_2/H_2) that have the potential for significantly lowering raw materials costs. These carbon and energy sources are currently under investigation.

ACKNOWLEDGMENT

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