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Scientific Note

An Economic Analysis of Microbial Reduction of Sulfur Dioxide with Anaerobically Digested Sewage Biosolids as Electron Donor

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Index Entries: Sulfur dioxide; flue gas desulfurization; economic analysis; sulfate-reducing bacteria; anaerobic digestion.

INTRODUCTION

A concentrated stream of sulfur dioxide (SO_2) is produced by regeneration of the sorbent in certain new regenerable processes for the desulfurization of flue gas *(1)*. It has been previously proposed that this SO_2 can be converted to elemental sulfur for disposal or byproduct recovery using a microbial/Claus process *(2,3)*. In this process, two-thirds of the SO_2-containing gas stream would be contacted with a mixed culture containing sulfate-reducing bacteria (SRB) where SO_2 would act as an electron acceptor with reduction to hydrogen sulfide (H_2S). This H_2S could then be recombined with the remaining SO_2 and sent to a Claus unit to produce elemental sulfur. The Claus process is well known in the natural gas industry *(4)*.

Glucose and heat/alkali pretreated municipal sewage sludge have been shown to act as ultimate electron donors and carbon sources for SO_2-reducing cultures of *Desulfovibrio desulfuricans (5)*. Sublette and Gwozdz *(6)* performed an economic analysis of this microbial SO_2 reduction process comparing the microbial process with conventional catalytic SO_2 hydrogenation with H_2 generation from methane. The design basis was a regenerator off-gas from a copper oxide, flue gas desulfurization process applied to a 1000 MW_e coal-fired power plant burning 3.5 wt% sulfur coal. All economics were based on an ultimate product gas of H_2S and SO_2 in a 2:1 ratio appropriate for feed to a Claus reactor. The fixed capital investments for the two processes were essentially equivalent. However, the annual operating costs for the microbial process were much higher than the conventional process primarily because of the high cost of raw materials, namely DE95 corn hydrolysate, which served as the electron donor and carbon source for the SO_2-reducing culture.

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Table 1
Operating Parameters for Continuous Bench-Scale SO_2-Reducing Bioreactor with Biomass Recycle[a]

pH, T	7.0, 30°C
MLSS[b]	10,000 mg/L
% by wt SRB in biomass	5%
Culture volume	1.5 L
SRB count	1.6×10^9/mL
AD-MSS[c] feed rate	48 mL/h
Dilution rate	0.77 d^{-1}
Feed soluble COD	2500 mg/L
Settler effluent soluble COD	300 mg/L
Recycle rate	90 mL/h
SO_2 feed rate	6.8 mmol/h
SO_2 conversion	100%
Recovery of SRB in settler	98%

[a] Mixed flocculated culture of *D. desulfuricans (7)*.
[b] Mixed liquor suspended solids.
[c] Anaerobically digested municipal sewage biosolids medium *(7)*.

The analysis by Sublette and Gwozdz has led to an effort to identify less expensive electron donors. One such source of electron donors is a mixture of fermentable substrates produced by anaerobic digestion of municipal sewage biosolids with inhibition of methanogenesis by chloroform. This material has been used as an electron donor and carbon source for mixed, septic cultures of *D. desulfuricans* immobilized by coculture with floc-forming anaerobes *(7)*. Operational parameters obtained from bench-scale continuous reactors with biomass recycle are summarized in Table 1. We present here an economic analysis of the microbial reduction of SO_2 similar to that of Sublette and Gwozdz with anaerobically digested municipal sewage biosolids serving as the electron donor and carbon source for a SO_2-reducing culture of *D. desulfuricans*.

DESIGN BASIS AND DESCRIPTION

Since the process/economic comparison presented here is intended to focus on the techno-economic differences between microbial and conventional SO_2 reduction, 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 mole ratio. The process design basis used for both SO_2-reduction methodologies is given in Table 2. The regenerated SO_2 gas stream 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 *(1)*. The product of both the microbial and conventional SO_2-reduction system was assumed to be balanced gas (H_2S/SO_2 = 2 mol/mol) 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, the regenerated SO_2-feed gas (stream 7)

Table 2
Process Design Basis Parameters for SO_2 Reduction Processes

Flue gas source	Coal-fired power plant
Power plant capacity, MW_e	1000
Ultimate feed coal analysis C/H/O/N/S, wt%	78.7/5.5/10.9/1.4/3.5
Feed gas source for SO_2 reduction	Regenerator off-gas from copper oxide process (90% SO_2/NO_x removal)
Reduction feed gas rate, lb mol/h	2.115
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 to Claus unit
Ultimate product gas H_2S/SO_2 mol ratio	2.0

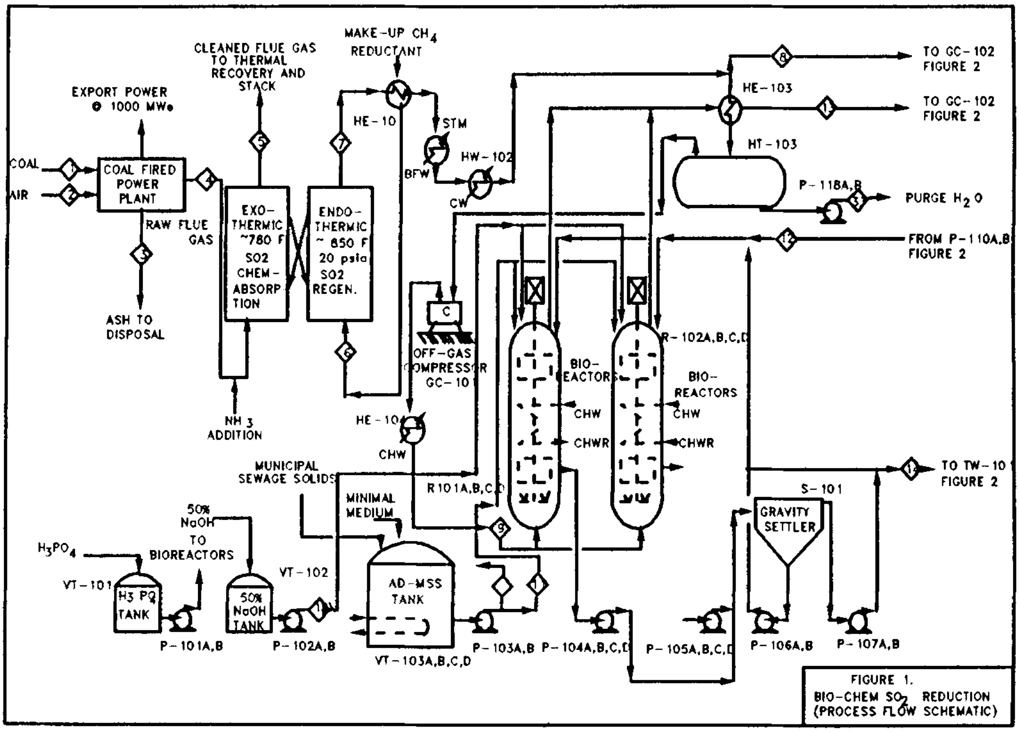

Fig. 1. Schematic diagram of process for biochemical SO_2 reduction (Part 1).

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 directly to the Claus plant. The balance of the feed gas, after water condensa-

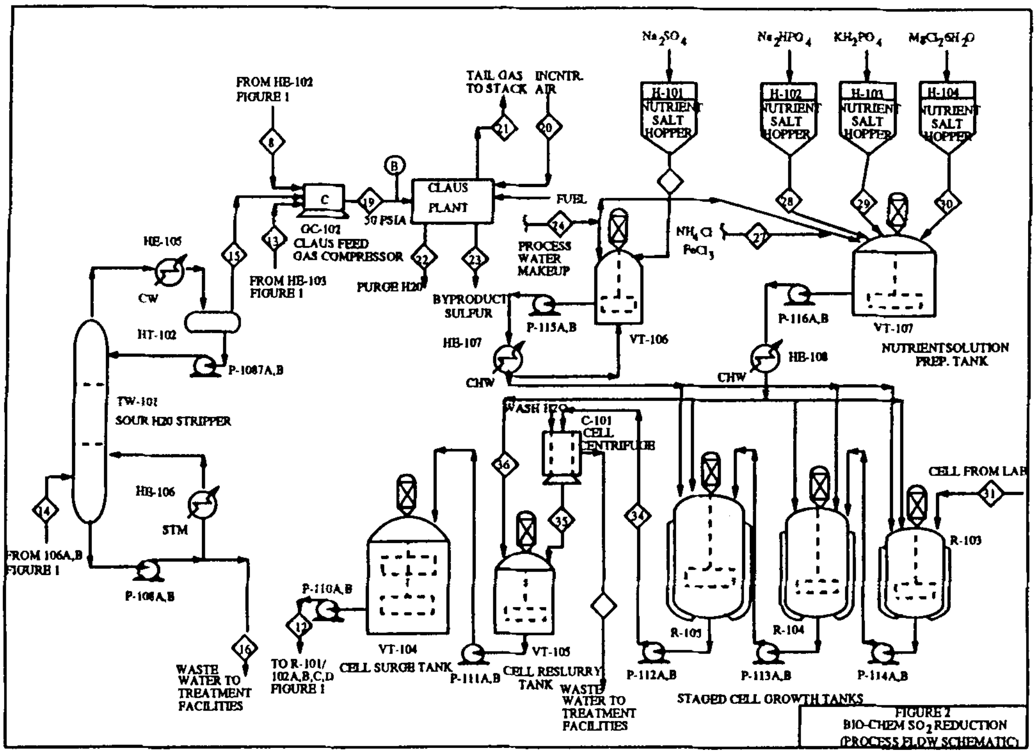

Fig. 2. Schematic diagram of process biochemical SO_2 reduction (Part 2)

tion, 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 of the 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. Anaerobically digested municipal sewage solids (AD-MSS) feed is utilized as the ultimate source of carbon and reducing equivalents for the bioreduction process. The production of this feed has been described elsewhere *(7)*. The design parameters (Table 3) for the SO_2-reduction bioreactors are based on the assumption of attainable SRB concentrations of 30 wt% of the total biomass. The reactors are operated at a biomass concentration of 50 g/L in each reactor.

Liquid effluent from the bioreactors is pumped (P-104A, B, C, D, and P-105A, B, C, D) to the gravity settler (HT-101), where the biomass settles under gravitational force, and the aqueous layer and biomass (solids) are separated. The design of the settler is based on the underflow solid concentration of 150 g/L and the settling characteristics of the biosolids obtained from jar test experiments *(7)*. A portion of the biomass is recycled to the reactors. The amount of biomass generated above that required to maintain 50 g/L in the reactor is combined with the supernatant of the settler and sent to the sour water 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 bottom is sent to wastewater treatment facilities in order to purge the net H_2O production across the bioreduction process. The

Table 3
Design Parameters for SO_2 Reduction Bioreactors
(1000 MW_e Eq Capacity)[a]

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	1.5
Total cell (SRB + non-SRB) density, g/L[b]	50 (fixed)
Desulfo/Heterotroph cell wt. ratio	0.3/0.7 (fixed)
Total SRB count, cells/L	4.8×10^{13}
Individual SRB cell weight, g/cell	3.125×10^{-13}
Desulfuricans specific activity, lb mol SO_2/h-cell	3.72×10^{-17} (6.8 mmol SO_2/h) ($1.6 \times 10^{12} \times 1.5$ cells)
Total reactant feed gas rate, lb mol/h	818.9
Total SO_2 feed rate, lb mol/h	465.2
SO_2 conversion to H_2S, % per pass	100
Total reactor(s) SRB cell inventory, lb	51210
Total reactor(s) oper. volume, gal	409,600
Total reactor(s) des. volume, gal	512,000 (w/20% free board)
Number of reactors required	8 (in parallel)
Des. vol/bioreactor, gal	64,000
Reactor dimensions, ft dia. x ft (T/T)	14 x 55
Nutrient source	AD-MSS
AD-MSS feed consumption gal/h (total)	65600
Gravity settler, area 825 m^2, 32 m dia.	1
Underflow concentration of biosolids (g/L)	150 (fixed)

[a] Based on the assumption of 30 wt% SRB in total biomass.
[b] Best case estimate.

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 mole ratio in the Claus feed gas (stream 19) is 2/1*M*.

Supporting auxiliary equipment (*see* Fig. 2) includes facilities for making mineral salts medium required to suspend sewage solids in the AD-MSS feed tank and Na_2SO_4 (used as terminal electron acceptor during start-up) solution preparation, cell growth, and cell isolation/reslurrying. These are required as a backup for any upset in the bioreactor systems and start-up. Conventional cell growth and isolation techniques are utilized in these facilities. This auxiliary equipment is also used for immobilizing the cells by coculturing with anaerobic floc.

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.

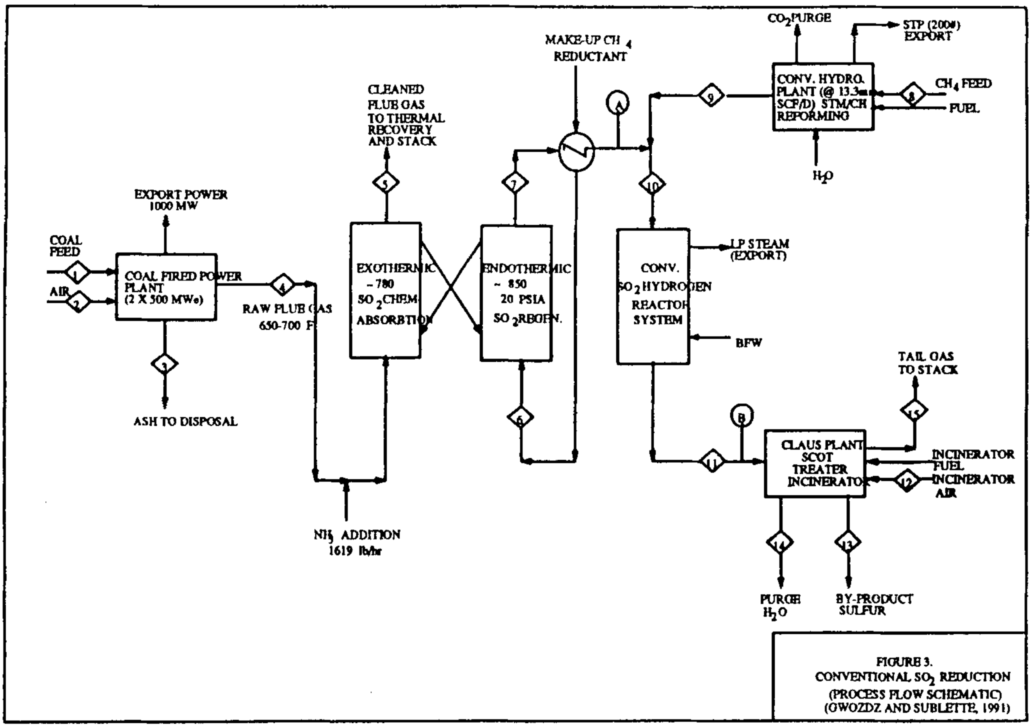

Fig. 3. Schematic diagram of convention SO_2 reduction process.

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 90) 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 fed gas (stream 11) is 2/1*M*.

PROCESS ECONOMICS

Total fixed and annual costs of production estimates for both the microbial and conventional SO_2-reducing processes are summarized in Tables 4–8. The total fixed and annual costs of production of the conventional process (and part of the microbial process) were obtained from earlier economic analyses *(6)*, since the same process was used in this analysis. However, the costs were updated for 1st quarter, 1994 by using Chemical Engineering Plant Cost Index and Marshall & Swift Equipment Cost Index (Chemical Engineering, 1994). As seen in Table 4, the fixed capital investment for the microbial process operating with 30 wt% SRB in the total biomass is about \$9.8 million higher than the conventional process. However, the production costs for the two processes are essentially equivalent. It is interesting to note that in the earlier analysis, where corn hydrolyzate was used as the carbon and energy

Table 4
Comparative Production Cost Summary
for Biochemical[a] and Conventional SO_2 Reduction Processes
(1000 MW_e Eq. Capacity

	Biochemical SO_2 reduction	Conventional SO_2 reduction
Investment (1st qtr. 1994) ($MM)		
Inside battery limits (ISBL)[b]	30.97	21.11
Outside battery limits (OSBL) at 25% ISBL	7.74	5.28
Total fixed investment (TFI)	38.71	26.39
Production cost ($MM/yr)[c]		
Raw materials	2.28	3.89
Utilities	1.60	(0.90)
Labor, formn, supvn. (L, F, S)	0.78	0.66
Maintenance, material, and labor (M, M, L) at 4% ISBL	1.24	0.84
Direct overhead at 45% L, F, S	0.35	0.30
General plant overhead at 65% oper. cost (L, F, S + M, M, L)	1.31	0.98
Insurance, property taxes at 1.5% TFI	0.58	0.40
Byproduct credit/debit	0	Excl[d]
Cash cost of production	8.14	7.01

[a] Based on the assumption of 30 wt% SRB in total biomass.
[b] *See* Tables 5 and 6.
[c] *See* Tables 7 and 8.
[d] Waste water treatment costs excluded.

Table 5
Microbial SO_2-Reduction Process ISBL Investment Estimate[a]
(1000 MW_e Eq. Capacity)

	$MM, 1st qtr. 1994
Purchased major equipment cost	8.28
Contingency at 10% purchased equipment	0.83
Total Purchase equipment cost	9.11
ISBL inv. at 3.4 x purchased equipment	30.97[b]

[a] Based on the assumption of 30 wt% SRB in total biomass.
[b] Includes purchased equipment, bulk materials, engineering, construction overhead, contractor's fee, and contingency. Excludes royalty/licensing fee, land, initial chemical charges, interest during construction, and so forth.

Table 6
Conventional SO_2 Reduction Process ISBL Investment Estimate
(1000 MW_e Eq. Capacity)

	Installed cost $MM, 1st qtr. 1994
Hydrogen plant at 14 mm SCF/D (via conventional steam/CH_4 reforming)	15.30[a]
Conventional SO_2 hydrogenation reaction system	3.89[b]
Contingency at 10%	1.92
ISBL investment	21.11[c]

[a] Investment-based data for conventional reforming technology.
[b] Investment estimate based on analogy with similar, commercially available, conventional catalytic hydrogenation technologies.
[c] Includes purchased equipment, bulk materials, construction overhead, contractor's fee, and contingency. Excludes royalty/licensing fees, land, initial catalyst and chemical charges, interest during construction, and so forth.

Table 7
Microbial SO_2-Reduction Process Summary
of Raw Materials, Utilities, and Labor Costs[a]
(at 8760 h/yr; 1000 MW_e Eq. Capacity)

	Price $/unit	Annual cost, $MM/yr, 1994
Raw materials		
Municipal sewage solids at 125,000 lb/h	0	0[b]
50% NaOH at 828 lb/h	0.065	0.48
H_3PO_4 (estimated)		0.12
Na_2HPO_4 at 750 lb/h	0.033	0.21
KH_2PO_4 at 1125 lb/h	0.033	0.32
$MgCl_2 \cdot 6H_2O$ at 440 lb/h	0.11	0.42
NH_4Cl at 125 lb/h	0.11	0.12
$FeCl_3 \cdot 6H_2O$ at 25 lb/h	0.60	0.13
$CHCl_3$ at 315 lb/h	0.11	0.30
Total raw material cost		2.1
Utilities		
Power at 2125 kW + 4775 kW/0.5 hr/d	0.044	1.30
Cooling water at 120 Mgal/h	0.08	0.10
Steam (200 psig) at 5 Mlb/h (Coal fired)	4.260	0.18
Process water at 1.5 Mgal/h	0.770	0.02
Total utilities cost		1.60
Labor		
Operators at 20 people	30,000	0.60
Foremen at 4 people	35,000	0.14
Supervision at 1 person	40,000	0.04
Total labor cost		0.78

[a] Based on the assumption of 30 wt% SRB in total biomass.
[b] Transportation cost of sewage solids from treatment plant is compensated for negative cost of the sewage solids.

Table 8
Conventional SO_2-Reduction Process Summary of Raw Materials, Utilities, and Labor Costs (at 8760 h/yr; 1000 MW_e Eq. Capacity)

	Price $/unit	Annual cost, $MM/yr, 1994
Raw materials		
Natural gas (CH_4) at 10,282 lb/h	0.038	3.42
H_2 plant (reforming + shift) catalysts		0.11
SO_2 hydrogenation catalyst		0.27[a]
Miscellaneous chemicals		0.06
Total raw material cost		3.86
Utilities		
Power at 680 kW	0.044	0.26
Cooling water at 148 Mgal/h	0.08	0.10
Boiler feed water at 11 Mgal/h	1.43	0.14
Fuel at 17 MMBTU/h	1.75	0.26
Steam (200 psig) at (57.9) Mlb/h, export	3.38	(1.66)
Total utilities cost		(0.9)
Labor		
Operators at 20 people	30,000	0.60
Foremen at 4 people	35,000	0.14
Supervision at 1 person	40,000	0.04
Total labor cost		0.78

[a] Based on the assumption of 30 wt% SRB in total biomass.
[b] Estimated.

source for microbial reduction of SO_2, just the opposite was true *(6)*. In the case of corn hydrolyzate as the carbon and energy source, the cost of raw materials for the bioprocess resulted in very high production costs compared to the conventional process. With AD-MSS as the source of the carbon and energy for SO_2-reducing cultures, the raw material costs are seen to be quite low. However, although less costly, the AD-MSS medium is very dilute (2500 mg/L COD) compared to DE95 corn hydrolyzate (68% glucose), resulting in increased capital costs for producing, storing, and utilizing the medium.

CONCLUSIONS

In summary, it appears that the microbial reduction process is not competitive with conventional SO_2-reduction techniques when AD-MSS medium is used as the feedstock hydrogen source. This is owing not to the intrinsic cost of the medium, but to the low concentration of fermentable substrates in this medium when prepared as previously described *(7)*. The greatest incremental decrease in costs of the microbial SO_2-reduction process would result from a decrease in the hydraulic loading to the bioreactors. Potentially this could be achieved by increasing the concentration of fermentable substrates in the AD-MSS medium through improvements in the anaerobic digestion process by which the medium is produced. Process economics could also be improved by increasing the SRB concentration in the process culture to >30 wt%. This would result in reduced bioreactor volume required and lower capital costs.

ACKNOWLEDGMENT

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