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Simultaneous Combined Microbial Removal of Sulfur Dioxide and Nitric Oxide from a Gas Stream

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ABSTRACT

A program is under way at the University of Tulsa to develop a viable process concept whereby a microbial process can impact on the problem of flue gas desulfurization and NO_x removal. We have previously reported studies of SO_2 reduction by *Desulfovibrio desulfuricans* and NO_x reduction by *Thiobacillus denitrificans*. One potential process concept is the simultaneous combined removal of SO_2 and NO_x from cooled flue gas by contact with cultures of sulfate-reducing bacteria ($SO_2 \rightarrow H_2S$) and *T. denitrificans* ($H_2S \rightarrow SO_4^{-2}$) as cultures-in-series or in coculture in a single contacting stage. Each of these contacting schemes has been investigated.

Index Entries: Sulfate-reducing bacteria; flue gas desulfurization; *Thiobacillus denitrificans;* sulfur dioxide; NO_x.

INTRODUCTION

Two process concepts have been developed for a microbial contribution to the problem of flue gas desulfurization and NO_x removal. We have previously demonstrated that the sulfate-reducing bacterium *Desulfovibrio desulfuricans* could be grown in mixed culture with fermentative heterotrophs in a medium in which glucose or molasses served as the only carbon source (*1,2*). Beneficial cross-feeding resulted in vigorous growth of *D. desulfuricans,* which used SO_2(g) as a terminal electron acceptor with complete reduction of SO_2 to H_2S in 1–2 s of contact time. Sulfate-reducing

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bacteria (SRB) cannot use simple sugars (such as glucose) as carbon and energy sources (*3*). The fermentative heterotrophs that developed in these cultures as a result of septic operation utilized glucose (and sucrose and fructose in the case of molasses) 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 −200 mV (*3*). However, in the working culture described above, no redox-poising agents were required. One possible explanation is that mixed nonSRB heterotrophs in the culture scavenged oxidants and thus kept the redox potential sufficiently negative to favor the growth of *D. desulfuricans*.

We have proposed that the concentrated SO_2 stream obtained from regeneration of the sorbent in the copper oxide process (*4*) could be split with two-thirds of the SO_2 reduced to H_2S by contact with a culture of sulfate-reducing bacteria (*1,2*). The resulting H_2S could then be combined with the remaining SO_2 and used as feed to a Claus reactor to produce elemental sulfur (*5,6*). Alternatively, a stream of H_2S and SO_2, such as obtained by regeneration of the sorbent in the NOXSO process (*7*), could be conditioned to produce the specific H_2S/SO_2 ratio (2/1) required by a Claus reactor.

We have also demonstrated that the facultative anaerobe and chemoautotroph, *Thiobacillus denitrificans*, can be cultured anaerobically in batch reactors using NO(g) as a terminal electron acceptor with reduction to elemental nitrogen (*8*). Thiosulfate served as an energy source, CO_2(g) as a carbon source, and NH_4^+ as a source of reduced nitrogen. The feed gas consisted of 0.5% NO, 5% CO_2, and balance N_2. Typically, the NO concentration in the outlet gas was 0.02%. Growth of *T. denitrificans* was indicated by depletion of thiosulfate and ammonium ion, and an increase in the biomass concentration. It is anticipated that NO_2(g) would react with water to form nitric acid and therefore make nitrate available as a terminal electron acceptor. We have proposed that the concentrated stream of NO_x as obtained from certain regenerable processes for glue gas desulfurization and NO_x removal could be converted to elemental nitrogen for disposal by contact with a culture of *T. denitrificans*.

Each of these process concepts involves byproduct recovery, or disposal from regenerable processes for flue gas desulfurization and NO_x removal. However, there is a third process concept that could incorporate a microbial contribution—simultaneous combined removal of SO_2 and NO_x from flue gas. This article describes an investigation of this process concept using cultures of *D. desulfuricans* and *T. denitrificans* in series reactors and mixed cultures in a single stage. In each configuration, SO_2 was reduced to H_2S, which was oxidized in turn to sulfate by *T. denitrificans*.

The anaerobic oxidation of H_2S by *T. denitrificans* has been described in detail elsewhere (*9–11*). *T. denitrificans* used NO as a terminal electron acceptor with reduction to N_2.

MATERIALS AND METHODS

Stock Cultures

D. desulfuricans (ATCC 13541) and *T. denitrificans* (ATCC 23642) were obtained from the American Type Culture Collection (Rockville, MD). Stock cultures were maintained as previously described (*1,9*).

Simultaneous Combined Removal of SO_2 and NO by *D. desulfuricans* and *T. denitrificans* Reactors-in-Series

Sulfite has been shown to be inhibitory to *T. denitrificans* (data not shown); therefore, in the series reactor system, the first stage was a *D. desulfuricans* reactor operated under SO_2-limiting conditions. In this reactor, SO_2 in the feed gas was converted to H_2S. The H_2S thus produced was then fed to a *T. denitrificans* reactor, where it was oxidized to sulfate. The *T. denitrificans* reactor was operated under sulfide-limiting conditions; otherwise sulfide would accumulate to toxic levels in the liquid phase (*9*). In order to prevent the accumulation of sulfide in an anaerobic *T. denitrificans* culture, the terminal electron acceptor had to be in stoichiometric excess. This required supplementing the culture medium of the *T. denitrificans* reactor with nitrate even when NO was included in the gas feed.

The series reactor system was developed as follows. Working cultures (2 L) of *D. desulfuricans* were developed in a Marubishi MD 300 fermentor as previously described and operated in a fed-batch mode with daily additions of 10 g/L glucose (*1*). Cultures were developed septically in a complex medium (pH 7.0, 30°C) followed by change over to a minimal medium with sulfate as the terminal electron acceptor and finally a change over to the same minimal medium with SO_2(g) (1% SO_2, 5% CO_2, balance N_2) as the terminal electron acceptor. After 24 h of operation on an SO_2 feed, the outlet gas was connected to the *T. denitrificans* reactor described below. The purpose of this incubation period was to ensure that all SO_2 in the feed gas was being reduced to H_2S and to acclimate the *D. desulfuricans* to SO_2 as a terminal electron acceptor. Ammonium chloride (0.3 g/L) was added every 48 h to prevent depletion.

T. denitrificans was grown anaerobically in thiosulfate medium in a B. Braun Biostat M fermenter as described previously (*9*). This medium contained nitrate as the terminal electron acceptor. Cultures were developed septically at pH 7.0 and 30°C. When the biomass concentration

reached approx 0.5 g/L, cells were harvested by centrifugation and resuspended in the same medium without thiosulfate. A gas mixture of 1% H_2S, 5% CO_2, and balance N_2 (35–70 mL/min) was then supplied to the fermenter under sulfide-limiting conditions as described previously (*9*). After 24 h of operation on an H_2S feed, the off-gas of the *D. desulfuricans* reactor was connected to the fermenter in place of the H_2S feed gas. The reactors were then maintained in series for as long as 48 h with periodic sampling of the reactor media and the outlet gases of both reactors to ensure healthy growth of both organisms prior to the introduction of NO.

After approx 24–48 h of operation of the reactors-in-series, a gas mixture of 0.49% NO, 5% CO_2, and balance nitrogen was fed into the *D. desulfuricans* reactor along with the SO_2 feed (and a N_2 purge). The flow rate of NO feed gas was increased stepwise from 70 mL/min to 145 mL/min (24 h at each flow rate for 72 h), whereas the flow rates of the N_2 purge and SO_2 feed gas remained constant at 357 mL/min and 31 mL/min, respectively. This gave an NO concentration in the feed of 760–1320 parts per million by volume (ppmv). Potassium nitrate (5 g/L) was added to the *T. denitrificans* reactor when needed to prevent depletion. A series reactor experiment was also conducted with an NO inlet concentration of 600 ppmv, which was held constant throughout the course of the experiment (168 h).

Simultaneous Removal of SO_2 and NO by *D. desulfuricans* and *T. denitrificans* in a Single Stage

In this series of experiments, *D. desulfuricans* and *T. denitrificans* working cultures were developed on SO_2 and H_2S feeds as described above. After *D. desulfuricans* cells were resuspended in the minimal glucose medium with SO_2(g) feed, the culture was allowed to incubate for an additional 24 h. The culture was then supplemented with components of *T. denitrificans* thiosulfate medium (except thiosulfate), which were not present in the minimal glucose medium before adding a slurry of *T. denitrificans* cells. The *T. denitrificans* cells were grown in thiosulfate medium to an OD_{460} of 0.8–0.9. Cells were then harvested by centrifugation and resuspended in thiosulfate medium without thiosulfate. A gas mixture (1% H_2S, 5% CO_2, and balance N_2) was then introduced at 35–70 mL/min for 24 h in order to acclimate the cells to utilizing sulfide as an energy source. The cells were then harvested again and resuspended in a minimum amount of 20 m*M* phosphate buffer solution (100 mL) at pH 7.0 and room temperature. Table 1 gives the overall composition of a modified medium for the growth and maintenance of *D. desulfuricans* and *T. denitrificans* in coculture. The coculture of both organisms was grown and maintained in the Marubishi MD 300 fermenter at pH 7.0 and 30°C.

The reactor was operated for 24 h following the addition of the *T. denitrificans* cell slurry to ensure that all the H_2S produced by *D. desulfuri-*

Table 1
Modified Maintenance Medium
For *D. desulfuricans* and *T. denitrificans* in Co-culture

Component	one liter
Na_2HPO_4	1.2 g
KH_2PO_4	1.8 g
$MgCl_2 \cdot 6H_2O$	2.4 g
NH_4Cl	0.5 g
$CaCl_2$	0.03 g
$MnSO_4$	0.02 g
$FeCl_3$	0.03 g
$NaHCO_3$	1.0 g
KNO_3	5.0 g
Glucose	10.0 g
Balch Vitamin Solution (*1*)	2.0 ml
Heavy Metal Solution (*9*)	15.0 ml
Mineral Water	50.0 ml

cans was utilized by *T. denitrificans* with oxidation to sulfate. At the end of this time, a gas mixture of 0.49% NO, 5% CO_2, and balance nitrogen was introduced into the reactor. The flow rate of this gas mixture was increased stepwise from 70 mL/min to 110 mL/min until it reached a toxic level. The flow rates of the N_2 purge and SO_2 feed gas were maintained constant at 357 mL/min and 31 mL/min, respectively. Glucose (10 g/L) was added daily. Potassium nitrate and ammonium chloride were also added when needed at concentrations of 7 g/L and 0.5 g/L, respectively.

Effect of Nitrate on NO Reduction by *T. denitrificans*

As will be detailed subsequently, nitrate in the medium of the *T. denitrificans* reactor appeared to suppress the utilization of NO as a terminal electron acceptor. In order to confirm this conclusion, the effect of nitrate on the utilization of NO in thiosulfate-fed cultures of *T. denitrificans* was also investigated. Working cultures of *T. denitrificans* growing on thiosulfate as an energy source and NO as a terminal electron acceptor were developed as previously described (*8*). Nitric oxide (0.49% NO, 5% CO_2, balance N_2) was supplied to the culture (1.4 L in a B. Braun Biostat M fermenter) at a molar feed rate of 1.3 mmol/h. After 48 h of operation, 5 g/L of KNO_3 was added to the culture and the outlet gas monitored for NO.

Effect of Oxygen on SO_2 Reduction by *D. desulfuricans*

Since flue gases may contain up to 3% or more O_2 (4), the effect of O_2 on microbial process cultures for SO_2/NO_x removal is an important consideration. The primary concern is the effect of O_2 on the sulfate-reducing bacteria, which are sensitive to the redox potential of the medium. *T. denitrificans* is a facultative anaerobe, and has been shown to be capable of switching rapidly between O_2 and NO_3^- as need dictates as long as NO_3^- is constantly available to induce denitrifying enzymes (unpublished data). Therefore, although O_2 will suppress NO utilization to some extent, it will not be inhibitory to *T. denitrificans*.

Working cultures of *D. desulfuricans* were prepared as described above with SO_2 (0.78 mmol/h) as the terminal electron acceptor and glucose (10 g/L every 24 h) as the carbon and energy source. When the culture stabilized on SO_2 feed with stoichiometric production of H_2S, air was introduced such that, considering all gases (SO_2 feed gas, N_2 purge, air), the O_2 concentration in the inlet gas was increased 0.9–6.5% over a period of 24 h. As will be detailed subsequently, inhibition of SO_2 reduction by *D. desulfuricans* was observed at an inlet O_2 concentration of 4.5%. A second experiment was then conducted with an inlet O_2 concentration of 1.3% to determine whether low concentrations of O_2 could be tolerated over more extended periods of time.

Analytical

Gas Analysis

The concentrations of H_2S and NO in the outlet gases of each reactor were determined by gas chromatography using a Hewlett Packard (HP) 5890A Gas Chromatograph with flame ionization detector (FID). The column used was a 10-ft by 1/8-in ID Teflon™ column containing 80/100 Porapak QS (Water Associates). The column temperature was 30°C, and the injector and detector temperatures were 60°C. The carrier gas was chromatographic grade helium delivered at a flow rate of 20 mL/min. A sample size of 1 mL was used. The retention times of NO and H_2S under these conditions were observed to be 1.05 and 1.55 min, respectively. Nitric oxide and H_2S were quantitated by comparing chromatograms of reactor outlet gas to chromatograms obtained from the NO and H_2S gas standards.

Culture Media Analysis

Analytical methods for quantitating biomass protein, optical density, sulfite, sulfate, elemental sulfur, nitrate, thiosulfate, and ammonia nitrogen in culture media have been previously described (*1,9*). The oxidation-reduction potential of culture media was monitored with a Chemcadet millivolt meter and a Wastewater ORP electrode (Number 5990-57), both obtained from Cole Parmer, Chicago, Illinois.

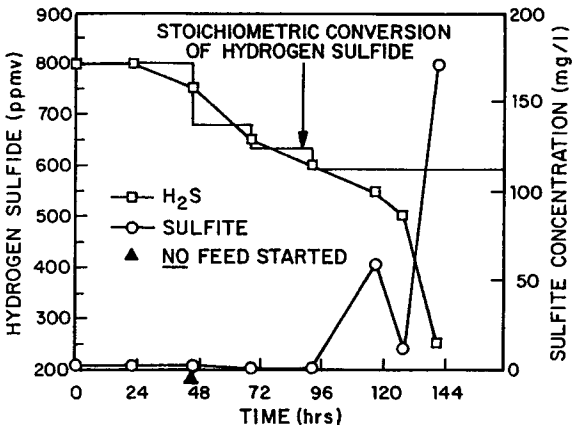

Fig. 1. Hydrogen sulfide concentration in the outlet gas and sulfite concentration in the medium of a *D. desulfuricans* reactor receiving SO_2 and NO feed (reactors-in-series).

RESULTS AND DISCUSSION

Simultaneous Combined Removal of SO_2 and NO by *D. desulfuricans* and *T. denitrificans* Reactors-in-Series

In a typical experiment, SO_2 was fed to the *D. desulfuricans* reactor at a molar flow rate of 0.77 mmol/h together with an N_2 purge. After stoichiometric production of H_2S was obtained, the off-gas of the *D. desulfuricans* reactor was fed to the *T. denitrificans* reactor, and the reactors operated in series for 48 h. During this time, stoichiometric production of H_2S in the outlet gas was observed, and no sulfite accumulated in the medium of the *D. desulfuricans* reactor. At the same time, sulfate accumulated in the medium of the *T. denitrificans* reactor, and only trace amounts of H_2S was detected in the off-gas of the *T. denitrificans* reactor.

After 48 h, NO was introduced into the feed gas of the *D. desulfuricans* reactor initially at a concentration of 760 ppmv and a molar feed rate of 0.86 mmol/h. After 24 h, the NO feed rate was increased to 1.3 mmol/hr (1060 ppmv), and finally after another 24 h, the feed rate was increased to 1.72 mmol/h (1320 ppmv) and maintained at this level throughout the course of the experiment.

As seen in Fig. 1, sulfite began to accumulate in the *D. desulfuricans* reactor at an NO concentration of 1060 ppmv about 48 h after the initiation of NO feed. As the sulfite concentration became inhibitory, less stoichiometric production of H_2S was observed in the outlet of the reactor. Dasu and Sublette (*12*) have previously operated *D. desulfuricans* and *T. denitrificans* reactors in series with SO_2 feed for as long as 100 h. Therefore, the upset conditions in the *D. desulfuricans* reactor were apparently the result

of NO inhibition. As SO_2 was removed from the feed gas of the *D. desulfuricans* reactor, the total biomass protein and the optical density of the culture increased. Removal of H_2S from the feed gas of the *T. denitrificans reactor* was accompanied by growth as indicated by the accumulation of biomass protein and sulfate, and consumption of nitrate and ammonium ion.

In each experiment of this type, inhibition of *D. desulfuricans* in the first stage was noted within 48–72 h after the initiation of NO feed. As noted above, the feed gas, the *D. desulfuricans* reactor outlet gas, and the *T. denitrificans* outlet gas were analyzed for NO by gas chromatography. These analyses showed that an average of 26% of the NO in the feed gas was removed by the *D. desulfuricans* reactor, which led to the speculation that NO was being used as a terminal electron acceptor by the nonSRB heterotrophs present in the reactor. Only about 20–25% of the remaining NO was removed by the *T. denitrificans* reactor. (Recall that nitrate was also available as a terminal electron acceptor in this reactor.) This was a much lower removal rate than that observed (>90% removal) when NO was the only terminal electron acceptor (*8*). Apparently, with nitrate available, NO utilization by *T. denitrificans* as terminal electron acceptor was greatly diminished. This was confirmed in *T. denitrificans* reactors operating with an NO(g) feed using thiosulfate as the energy source as described below.

As noted in the previous section, an experiment was also conducted in which the NO concentration in the inlet gas was held constant at 600 ppmv with all other conditions identical to those described above. Less stoichiometric output of H_2S and accumulaton of sulfite in the medium of the *D. desulfuricans* reactor were evident 144 h after the introduction of NO. *D. desulfuricans* reactors have been operated in our laboratory for 2–3 wk on an SO_2 feed with glucose as a carbon source without difficulty. Therefore, the upset conditions after 6 d are likely the result of the NO in the feed gas. It appears that, at lower NO concentrations, inhibitory effects simply take longer to appear.

Simultaneous Removal of SO_2 and NO by *D. desulfuricans* and *T. denitrificans* in Coculture

The growth and maintenance of *D. desulfuricans* and *T. denitrificans* in coculture were described above. The reactors were initially operated at an SO_2 feed rate of 0.77 mmol/h for approx 24 h with N_2 purge. During this time, stoichiometric production of H_2S was observed in the outlet gas of the reactor. A slurry of *T. denitrificans* cells previously grown on thiosulfate and H_2S was then added to the *D. desulfuricans* reactor together with 7 g/L of KNO_3. Within 1 h the H_2S concentration in the outlet gas was reduced to trace levels. The H_2S produced from SO_2 reduction by *D. desulfuricans* was immediately oxidized by *T. denitrificans* to sulfate, which accumulated

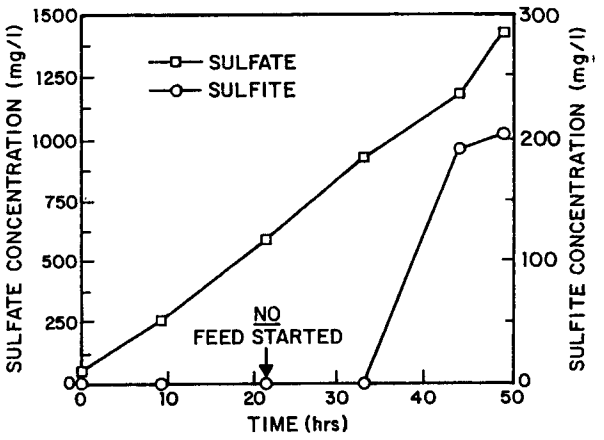

Fig. 2. Sulfate and sulfite concentrations in *D. desulfuricans* and *T. denitrificans* coculture receiving SO_2 and NO feed. (Sulfate is not inhibitory to *T. denitrificans* below a concentration of 200 m*M*.)

in the culture medium as shown in Fig. 2. No sulfide, sulfite, or elemental sulfur was detected during this time. Growth of both organisms was indicated by an increase in optical density of the culture.

NO feed was introduced at concentrations of 760–910 ppmv 24 h after the addition of *T. denitrificans* cells. At an elapsed time of 12 h after the initiation of NO feed, sulfite began to accumulate in the medium as shown in Fig. 2. Again, NO inhibition of the growth of *D. desulfuricans* was indicated.

In each experiment of this type, a tremendous amount of nitrate was required in a short period of time. For example, it was estimated that approx 100 mmol of nitrate were required for the reduction of 25 mmol of SO_2. Assuming that all the SO_2 in the feed gas was being reduced to H_2S by *D. desulfuricans*, an NO_3^-/H_2S ratio of 4 was obtained. This ratio is twice as high as that reported by Sublette (*9*) for anaerobic oxidation of H_2S by pure cultures of *T. denitrificans* in batch reactors. Apparently, the nonSRB heterotrophs present in the reactor were utilizing nitrate as a terminal electron acceptor during the fermentation process.

Effect of Nitrate on NO Reduction by *T. denitrificans*

As described above, an anaerobic working culture of *T. denitrificans* was developed in thiosulfate medium with nitrate as the terminal electron acceptor. Following a change of medium to thiosulfate medium without nitrate, an NO feed was initiated at a molar flow rate of 1.3 mmol/h at a concentration of 4900 ppmv. The culture was maintained at these conditions for 48 h. During this time, about 90% removal of NO was observed with a concommitant decrease in the concentration of thiosulfate and production of sulfate.

After 48 h of operation, potassium nitrate (5 g/L) was added to the culture. A few minutes after the addition of nitrate, the NO concentration in the outlet gas began to rise. At an elapsed time of 28 h after the addition of nitrate, the NO removal was down to 65% and leveled off. With the addition of nitrate, the rates of thiosulfate utilization and the sulfate accumulation were seen to increase. These results indicated that nitrate was a preferable terminal electron acceptor over NO.

Effect of Oxygen on SO_2 Reduction by *D. desulfuricans*

When air was introduced to *D. desulfuricans* working cultures operating on an SO_2 feed, the redox potential increased immediately from −230 to −165 mV. As the O_2 concentration was increased to 6.5%, the redox potential increased more gradually, ultimately to about −100 mV. When the O_2 concentration reached about 4.5% (about 12 h after the first introduction of air), sulfite began to accumulate in the culture medium, indicating inhibition of SO_2 reduction by *D. desulfuricans*. In a separate experiment, a lower, constant O_2 concentration (1.3%) was used in the inlet gas. Again the redox potential was seen to increase sharply (from −240 to −200 mV) when air was introduced into the reactor, but then remained relatively constant for 65 h. During this time, little or no sulfite could be detected in the culture medium. After 65 h, the O_2 concentration was increased to 2.8%. The redox potential then increased to about −150 to −160 mV, and sulfite began to accumulate in the culture medium.

It is apparent that mixed cultures of *D. desulfuricans* and fermentative heterotrophs have a significant amount of redox-buffering capacity. However, there is probably not sufficient buffering capacity to allow these cultures to function properly in contact with flue gas.

CONCLUSIONS

The simultaneous combined SO_2/NO_x removal from flue gas based on direct contact of the cooled gas with *D. desulfuricans* and *T. denitrificans* cultures-in-series or cocultures does not appear to be technically feasible at the present time for the following reasons:

1. NO inhibition of SO_2 reduction by *D. desulfuricans*—Although the NO concentrations used in the experiments described above are somewhat higher than that found in a typical flue gas, it appears that at lower NO concentrations (or partial pressures), the inhibitory effects will simply take longer to become apparent. One interpretation of these experiments is that NO transferred into the liquid phase inhibited (or killed) *D. desulfuricans* cells one by one, until there were insufficient "active sites"

available to reduce SO_2 as fast as it was sparged into the culture. At this point, sulfite began to accumulate in the liquid phase, further inhibiting the biomass. The long-term effects of lower concentrations of NO on SO_2 reduction by *D. desulfuricans* should be investigated. However, at best NO inhibition may impart a borderline stability on microbial SO_2 reduction.

2. Nitrate suppression of NO removal—As noted previously, the cultivation of *T. denitrificans* in a microbial flue gas treatment system (either one or two stages) requires sulfide-limiting conditions. Therefore, the electron acceptor must be in excess, requiring nitrate in the *T. denitrificans* process culture. As shown in experiments described above, nitrate significantly suppresses the removal of NO from a feed gas, making simultaneous SO_2/NO_x removal impractical by these microbial means.
3. O_2 inhibition of SO_2 and NO reduction—It was demonstrated that *D. desulfuricans* working cultures are tolerant of up to 1.3% O_2 in the feed gas. Apparently, at low O_2 feed rates, facultatively anaerobic nonSRB heterotrophs in the culture scavenge O_2, keeping the redox potential sufficiently low to favor growth of *D. desulfuricans* (and SO_2 reduction). However, further increases in the O_2 partial pressure in the feed gas resulted in O_2 inhibition of SO_2 reduction. These inhibitory levels of O_2 are comparable to those concentrations found in flue gases. Therefore, in any process in which raw flue gas contacts a *D. desulfuricans* culture, marginal stability at best can be expected. Oxygen in the feed gas will also produce a suppression in NO removal similar to the effect of nitrate.

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