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Note on

The Removal of Sulfur from Stack Gases by an Electrical Discharge

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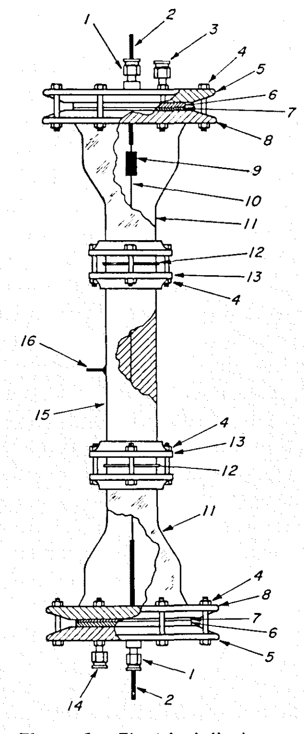
Consideration of the use of an electrical discharge for the removal of sulfur compounds from stack gases is based on the theory that the ionization energy of SO_2 and H_2S is less than the other major constituents and that they therefore may enter preferentially into reactions to form lower vapor pressure products. There is also no information on the extent to which the electrical discharge, as it normally occurs in the electrostatic precipitator, may contribute to the conversion of SO_2 to SO_3, to the formation of obnoxious nitrogen oxides, or whether the use of pulsed power may increase these reactions.

Experimental Methods and Equipment

The reaction chamber which was patterned after an electrostatic precipitator is illustrated in Figure 1. High-frequency power was obtained from a damped wave spark gap oscillator which had a carrier frequency of 10 megahertz. Since a comparison of waveforms can only be made on the basis of the same energy input, power output was estimated by using the air filled reaction chamber as a dummy load with resulting temperature increase of the chamber compared to the same temperature increase when the central 0.010 in. diameter stainless steel wire was used as a heating element by the passage of a low voltage current. The high frequency power was thus found to be 9 watts compared to the 37 watts obtained when the same



1 Central wire rod retainer
2 Central wire rod
3 Gas inlet fitting
4 Flange fasteners
5 Aluminum rings
6 Teflon disks
7 3-inch O D Teflon glass pipe gasket
8 3-inch O D glass pipe flange
9 Teflon insulator
10 Central wire
11 3-inch O D to 2-inch O D glass pipe reducer
12 2-inch O D Teflon glass pipe gasket
13 2-inch O D glass pipe flange
14 Gas outlet fitting
15 2-inch O D x 8-inch stainless steel tubing
16 Ground connection

Figure 1. Electrical discharge reaction chamber.

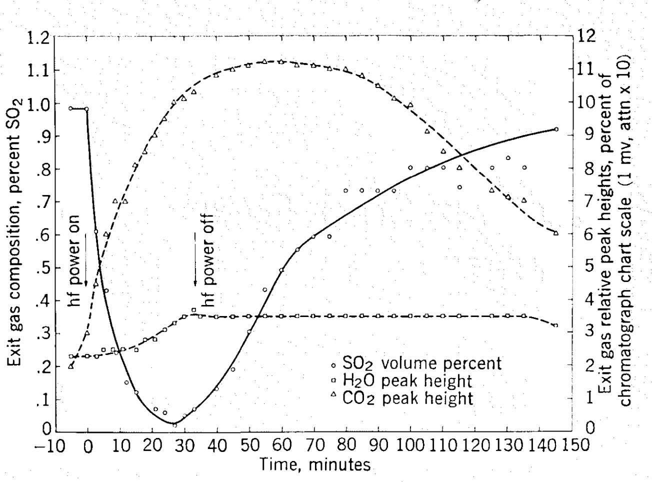


Figure 2. Typical electrical discharge reaction curve.

chamber was operated as an electrostatic precipitator with direct current. With the air filled chamber as a dummy load most of the power is converted to heat since reaction products are negligible. Any errors here would not be as great as encountered in the substitution of a solid resistance dummy load where problems of impedance matching, tuning, and radiation losses would be involved.

Standard SO_2-N_2 compressed gases containing 0.211, 0.979, and 8.40% SO_2 were used for chromatograph calibration and gas mixture preparation. Quantitative analysis of the feed and product gas was obtained by means of a Beckman GC-5 chromatograph with a 10 in. disc integrator, and a 10 ft long, ⅛ in. O.D. stainless steel column packed with 80-100 mesh Poropak QS.

Results

When the chamber is operated with direct current, some reaction products are formed but the quantity is insignificant compared to that obtained with damped wave high frequency current. This is graphically illustrated in Figure 2 where approximately 96% of the SO_2 is removed during the 33 min the pulsed power is on. For these tests, the reaction chamber was placed in an oven at 140°C, and the feed flowing at 600 ml/min was composed of a standard 0.979% SO_2 in N_2 with an added 4% of air saturated with H_2O. When H_2O was absent, only 67% of the SO_2 was removed. These results are typical of a number of tests. Reaction products were identified as elemental sulfur and sulfuric acid. Chromatographic analysis indicates that the N_2O or N_2O_3 content of the reacted gas is approximately 10 ppm, while NO, NO_2, and ozone were not detectable. However, by substituting a one inch diameter chamber, brown nitrogen oxides were obtained with air. Based on the absorption of the nitrogen oxides in water with analysis for nitric acid, it was found that the addition of CO_2 to the dry air increased the nitrogen oxide formation fivefold, and, when added to moist air, threefold.

The shock excitation of the oscillatory circuit in the damped wave generator produces a pulsing of the carrier frequency. In view of a simplicity superior to SO_2 reactions, evidence of the effectiveness of pulsing alone was obtained with a direct current discharge in air where a maximum in the small amount of nitrogen oxide formed was found to be at a pulse width of 500 μsec and a repetition rate of 1000 hertz. Further evidence was obtained with the use of the pulsed output of a radar generator. The radar generator, with a 2800 megahertz frequency pulsed at 900 hertz with a pulse width of one μsec, produced the highest concentration of nitrogen oxides, sufficiently high to color the exit gas dark brown.

Although the nitrogen oxide tests illustrate the effectiveness of pulsing, they may also be indirectly related to SO_2 removal. Some tests with simulated stack gases at 115°C, and with 4% H_2O, show that the complete removal of the SO_2 is possible. Only small amounts of nitrogen oxides are needed, and these are removed from the discharged gas as a solution in the H_2SO_4 product.

Discussion and Summary

The results show that a damped, or pulsed, high frequency current is more effective than direct current in sulfur removal. Since the data include only the initial and final compositions, nothing can be said of the mechanism of the reaction. The low concentration of nitrogen oxides in the SO_2 tests is explained on the basis that the ionization energy of N_2 is higher than SO_2, a conclusion corroborated by the increased nitrogen oxide formation with the higher electric field intensities in the 1 in. diameter tube.

Although it is too early to indicate the feasibility of adapting the discharge process to SO_2 removal, the use of nitrogen oxides as catalytic agents for SO_2 to SO_3 conversion may have more tangible prospects. Since the electrical discharge formation of nitrogen oxides is easily turned off and on, the process is of particular significance to power plants where dump energy is available. The nitrogen oxides formed by this intermittent surplus of electrical energy would be accumulated and used continuously for SO_2 to SO_3 conversion. Even though the electrical discharge synthesis of nitrogen oxides is now closely competitive to the high pressure ammonia synthesis process, further increase in efficiency is possible with a pulsed waveform and the use of CO_2 as a reaction promoter.

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