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E&E Newsletter (October 1994)

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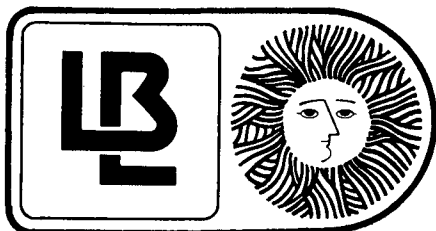
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Publication Date

1994-10-01

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Lawrence Berkeley Laboratory

Energy and Environment Division**NEWSLETTER**

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DESULFURIZATION AND DENITRIFICATION OF POWER PLANT FLUE GASES

by Ted S.-G. Chang and David Littlejohn

Power plants release oxides of sulfur and nitrogen into the air as a result of the burning of fossil fuels. At high concentration these gases directly affect human health, and when further oxidized and hydrolyzed, they are converted into sulfuric and nitric acids and fall to Earth as acid rain to corrode metals and etch buildings and monuments made of calcareous rock. Acid rain will also acidify surface and ground water to a point where toxic trace metals reach concentrations that make the water unfit for human consumption or unsuitable for aquatic animals, and inhibit crop and forest productivity. Therefore, oxides of sulfur and nitrogen in power plant flue gas should be removed before they are released to the atmosphere.

While the development of flue gas desulfurization processes has been progressing for many decades, a satisfactory process is not yet available. The lime/limestone wet flue gas desulfurization scrubber, the most widely used process in the utility industry at present, has not proved capable of reliable and efficient sulfur dioxide capture. It is also very expensive, accounting for about 25% of the capital and operating cost of a power plant. Techniques for the control of nitrogen oxides emissions have not been developed as extensively as those for control of sulfur dioxide emissions. However, these techniques are also needed because nitrogen oxides also contribute to the formation of acid rain. The acidic portion of rainfall in the eastern U.S. has been analyzed to be about two thirds sulfuric acid and one third nitric acid. In the western U.S., the acids in rain are generally about half sulfuric and half nitric, although in some areas as much as 80% of the rain's acidity can be composed of nitric acid.

The scrubber chemistry group is studying the basic chemical interactions of sulfur dioxide, nitrogen oxides, and other additives and is developing processes in which both sulfur dioxide and nitrogen oxides can be removed simultaneously in a single scrubber. The processes being developed are "wet processes" because water is used as the medium to remove these compounds from the flue gases. Both sulfur dioxide and

nitrogen dioxide are moderately soluble in water and aqueous solutions, and can be efficiently removed without too much difficulty. However, nitric oxide, which is the major constituent of nitrogen oxides in flue gases, is only slightly soluble in water. One way to circumvent this problem is to add ozone or another oxidant to the flue gases to convert the nitric oxide into nitrogen dioxide, which will dissolve into the scrubbing liquor. Another method to remove nitric oxide is the addition to the scrubbing solution of a compound that will rapidly and efficiently bind with nitric oxide. Some of the compounds we are investigating are ferrous chelates which efficiently form the nitrosyl ferrous chelates.

The chemistry of the scrubbing process is not over once the nitrogen oxides and sulfur dioxide are absorbed into solution. In fact, it is only the beginning. In the process where nitric oxide is converted into nitrogen dioxide in the gas phase, conditions can be adjusted such that nitrite ion is the major nitrogen oxyacid in solution; while sulfur oxyacids such as bisulfite and sulfite ions are formed after dissolution of sulfur dioxide. Interaction of nitrite ion with bisulfite/sulfite ions takes place in a very complicated fashion. Many concurrent and consecutive chemical reactions can occur; and many chemical species such as nitrososulfonic acid, hyponitrous acid, hydroxylamine sulfonates, aminesulfonates, hydroxylamine, sulfamic acid, ammonium ion, nitrous oxide, nitrogen, and sulfate can be produced as a result of this interaction. We are studying important reactions involved in the system to obtain kinetic and thermodynamic data on them. With this information, a model calculation of the performance of a scrubber can be carried out and an optimum scrubber of this type can be constructed.

In the case where ferrous chelates are used to promote the absorption of nitric oxide in the scrubbing liquor, nitric oxide in the resulting nitrosyl ferrous chelates can be reduced by bisulfite/sulfite ion to yield nitrogen and nitrous oxide while sulfur dioxide is oxidized to sulfate ion. The ferrous chelates are regenerated and can be used again to bind with nitric oxide. We are investigating the kinetics and mechanisms of the reactions involved to determine the regeneration rates of metal chelate catalysts and to have the ability to control the products of reaction by adjusting the scrubber operating conditions. We are also studying the kinetics and thermodynamics of the coordination of nitric oxide to various metal chelates to identify efficient catalysts for optimum absorption of nitric oxide.

The scrubber chemistry group has recently found that some products of the reaction of sulfur dioxide and nitrogen oxides in solution, as well as nitric oxide and nitric oxide containing metal chelates, can react with hydrogen sulfide to produce elemental sulfur, which is a commercially useful material. Research is in progress to understand the reaction scheme for this more desirable regenerable process.

As coal and petroleum with high sulfur content become more attractive energy sources, the need for processes such as those we are developing becomes more urgent. We believe that an efficient and cost-effective scrubber for simultaneous control of flue gas sulfur dioxide and nitrogen oxides can be developed only if the basic chemistry of

sulfur dioxide and nitrogen oxides is well understood. It is toward this goal that we are working.

TRIPS, CONFERENCES AND PRESENTATIONS

November

- Art Rosenfeld spent a month in Beijing, China attending and speaking at two sessions of the first U.S./China Conference on Energy, Resources and the Environment. Art also attended and lectured at seminars on energy efficiency in buildings as a guest of the China Association of Science and Technology, sponsored by the U.S. National Academy of Sciences.
- Dick Fish was an invited speaker at two functions: he spoke at a seminar sponsored by the Bartlesville Energy Technology Center, held in Bartlesville, Oklahoma, on "Speciation of Inorganic Arsenic and Organoarsenic Compounds in Fossil Fuel Precursors and Products and the Speciation of Vanadyl Porphyrin and Non-Porphyrin Compounds in Heavy Crude Petroleums" and gave a talk at the New York Academy of Sciences conference on catalytic transitions-metal hydrides entitled "Selective Catalytic Reductions of Polynuclear Heteroaromatic Nitrogen Compounds Using Both Homogeneous and Heterogenized Forms of Chlorotris (Triphenylphosphine) Rhodium(I)."

December

- Art Rosenfeld was in Knoxville to speak at a seminar on energy policies and programs to 35 federal officials from DOE and DOD.

DIVISION NEWS

- Howard Coleman, Deputy Assistant Secretary for CRE, and Norman Hughes, Assistant to the Assistant Secretary, visited the Lab on November 17. Their primary interest was to become familiar with our work for Maxine Savitz, which includes building energy research, electrochemical energy storage, and some of the combustion work.

- At the end of November the Thermal Energy Storage (TES) branch of CRE invited the Lab to make a proposal to become a lead center for part of the TES program. As a lead center, we would have an in-house research program and management responsibilities for work at other institutions. A number of LBL researchers with an interest or expertise in various aspects of thermal storage developed short descriptions of advanced research topics. The topics were assembled in a research plan, which was then coupled to a management plan patterned after that of the lead center for Electrochemical Energy Storage (Cairns and McLarnon). Elton presented the combined research/management plan at DOE headquarters to an audience which included James Swisher, Director of Energy Storage, and other DOE officials. The response was generally favorable, and there is a possibility that some of the research topics will be funded this fiscal year. However, a decision to alter the present management structure to include LBL is to be held in abeyance pending a clearer picture of the FY84 budget for thermal storage.
- Alvin Trivelpiece, Director of the Office of Energy Research, DOE, will visit the Lab on Friday and Saturday, December 10 and 11, to review the projects funded by the Office of Health and Environmental Research. E&E will have a half-hour to discuss atmospheric aerosol and indoor air quality research.

ANNOUNCEMENT

The Energy and Environment Division Christmas Party will be held on December 17 at 12:00 p.m. in the third floor conference room and surrounding area. All employees are invited. Please bring a refreshment to share.



RECENT REFEREED JOURNAL ARTICLES

"Real-Time Measurement of the Absorption Coefficient of Aerosol Particles," A.D.A. Hansen, H. Rosen, and T. Novakov, Applied Optics, 2: 17 (September 1982).

"Large-Area, High-Sensitivity Heat-Flow Sensor," J.H. Klems and D. DiBartolomeo, Rev. Sci. Instrument., 53:10 (October 1982).

"Synthesis, Structural Elucidation, and Stereochemistry of Five-Coordinate Organoarsenic Catecholates," Richard H. Fish and Raja S. Tanous, Organometallics, 1:1238 (1982).

"Thermodynamics and Kinetics of the Coordination of NO to Fe^{II} NTA in Aqueous Solutions," Nenhua Lin, David Littlejohn, and Shih-Ger Chang, I&EC Process Design & Development, 21:725 (1982).

 Wishing You
Good Health and Good Cheer
this Holiday Season 

PUB-432 11-82/450 This work was supported by the U.S. Department of Energy under Contract Number DE-AC03-76SF00098.
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