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THE CONTROL OF NO_x AND SO_x EMISSIONS FROM CONVENTIONAL
PULVERIZED COAL FLAMES: THE CANADIAN PROGRAM

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MARCH 1983

For presentation at the Joint Power Generating Conference, September 30
to October 4, 1984 Toronto, Canada

ENERGY RESEARCH PROGRAM
ENERGY RESEARCH LABORATORIES
DIVISION REPORT 84-24 (OP) (TR)



ERP/ERL 84-024 (op) (TR)
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THE CONTROL OF NO_x SO_x EMISSIONS
FROM CONVENTIONAL PULVERIZED COAL FLAMES:
THE CANADIAN PROGRAM

by

G.K. Lee*, H. Whaley**' and M.P. Heap***

ABSTRACT

Research work sponsored under an International Energy Agency Agreement between Canada, Denmark and Sweden has shown that NO_x and SO_x emissions from pulverized coal flames can be reduced significantly by staged combustion concepts. During this project in which 45 coals were evaluated, it was determined that much of the fuel nitrogen in the volatile matter can be transformed to N₂ instead of NO. The conversion of fuel nitrogen to NO was from 30% to 40% in conventional flames but only from 7% to 14% in staged flames. Nitrogen retained in the char showed about 20% conversion to NO and appeared to be relatively independent of local oxygen concentration. Reductions in sulphur emissions of 50% by sorbent injection into tertiary combustion air were achievable with Ca/S ratios of 2/1.

These research results are being incorporated into two proposed field trials involving retrofits of staged combustion systems in a front-wall-fired and a tangentially-fired boiler.

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INTRODUCTION

One of the major concerns associated with the expanded use of coal for heat and electricity is the emission of acid rain precursors, NO_x and SO_x , to the atmosphere. In 1980, North American utility boilers, most of which are coal-fired, emitted over 5.9 million tonnes of NO_x and 16.6 million tonnes of SO_x with Canadian utility sources accounting for about 5% of both the NO_x and SO_x emissions.

Current developments on abatement technology indicate that these pollutants can be controlled by either flue gas scrubbing or by combustion system modification. Flue gas scrubbing has generally proven to be energy intensive, expensive and complex. For this reason, increased emphasis has been placed on combustion modification and it now appears to be the most cost effective method for control of NO_x and/or SO_x from pulverized-coal fired boilers.

This paper reviews some of the fundamental aspects of NO_x and SO_x control in flames and describes two major Canadian initiatives to suppress simultaneously the generation of NO_x and SO_x by staged-burner aerodynamics and sorbent injection.

THEORETICAL CONSIDERATIONS

 NO_x FORMATION

Flame generated NO_x is produced from two sources:

1. Thermal NO_x formed by high temperature reaction of atmospheric oxygen and nitrogen.
2. Fuel NO_x formed by the reaction of fuel nitrogen with available oxygen during combustion.

Thermal NO_x , although a major source of the total NO_x generated when low nitrogen fuels are burned, is a minor source of NO_x when high nitrogen fuels such as residual oil, coke and coal are burned in suspension firing systems.

Thermal NO_x forms fairly slowly and its formation is favoured by flame temperatures over 1400°C , low rates of heat extraction, high excess air levels with intense air/fuel mixing, and high volumetric heat release rates.

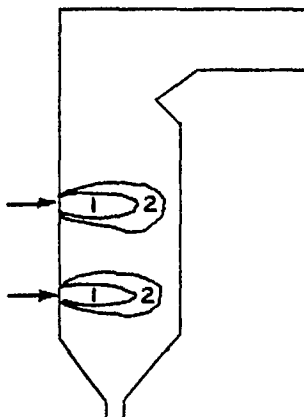
Fuel NO_x , on the other hand, appears to be only weakly dependent on temperature and forms very rapidly. Its' formation is promoted by fuel stoichiometry during pyrolysis, a large evolution of volatile nitrogen species, and low rates of heat extraction.

Burner or physical staging of combustion air is being widely studied as a cost effective means for minimizing the oxidation of fuel nitrogen. One popular technique has been to design burners, combustion chambers or combinations of both technologies with a fuel-rich primary stage followed by controlled additions of secondary air and injections of tertiary air to complete combustion slowly as shown in Fig. 1.

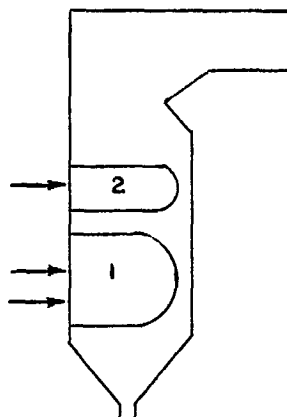
During coal combustion, upwards of 70% of the total NO_x emissions is derived from fuel nitrogen which forms N_2 , HCN, NO and NH_3 during sub-stoichiometric and devolatilization. About 20% to 35% of the fuel nitrogen can be retained in the char, but only about 20% of this char nitrogen is converted to NO_x regardless of stoichiometry or mixing intensity. Good char burnout can therefore be achieved under high excess air conditions without significantly increasing the NO_x levels entering the tertiary combustion zone.

A second method under study is to employ "fuel reburning" to chemically reduce NO formed in the initial combustion stage. In this method, also shown in Fig. 1, the fuel is burned in the lower furnace under slightly air-rich conditions. Then additional fuel is added downstream of the first zone to reduce or "reburn" most of the NO generated in the first stage under slightly sub-stoichiometric conditions to N_2 . Finally, tertiary air is injected into the fuel-rich products leaving the reburning zone to complete the combustion process under slightly air-rich conditions.

WALL FIRED FURNACES

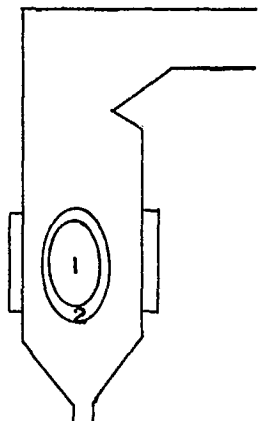


A) BURNER STAGING

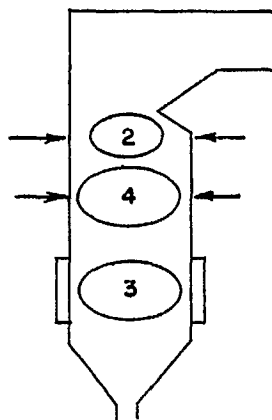


B) FURNACE STAGING

TANGENTIALLY FIRED FURNACES



C) STAGED BURNER AND FURNACE



D) STAGED FUEL REBURNING

1 = 65 % TOTAL AIR
2 = 25 % TOTAL AIR

3 = 110 % TOTAL AIR
4 = 95 % TOTAL AIR

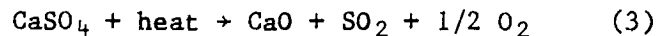
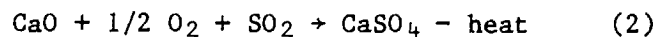
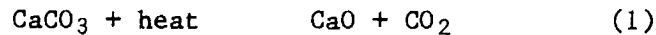
Fig. 1 - Staged combustion: burner, furnace and fuel reburning

SO_x Emissions

Sulphur in fuel, regardless of the form, generates SO_x during combustion. Some of this sulphur may react with indigenous alkaline ash constituents, but the balance is emitted as gas phase SO_x.

Most flame sulphur capture systems use calcium-based minerals to react with and convert SO_x to solid sulphates. Other less developed concepts utilize sulphur capture with a calcium sorbent under reducing conditions to produce calcium sulphide which must then be either rendered inert to prevent decomposition of the sulphide or oxidized to sulphate during the final combustion stage under excess air conditions. Therefore, the effectiveness of sulphur neutralization with simultaneous NO_x reduction will depend strongly on sorbent injection into a furnace region with the desired stoichiometry, temperature and residence time.

When injected into an oxidizing flame zone, the sulphur/sorbent reactions are as follows:



Calcination in the first reaction at temperatures above 1000°C and below 1350°C, the CaO is rapidly deactivated by dead burning which significantly reduces the specific surface area available for reaction with sulphur. Overheating of the CaSO₄ end product is also undesirable because at temperatures above 1350°C, CaO and SO₂ are regenerated, as illustrated in reaction 3. The best SO_x capture will occur when the sorbent is injected into an oxidizing region which allows calcination without deactivation. Sulphation can then occur without regeneration leading to more contact time at reaction temperatures from 750°C to 1200°C (see Fig. 2).

Sulphur capture is also enhanced by increased thermal loading of the reaction zone, increased sorbent fineness down to about 50 μm, Ca/S mol ratios over 2, and the presence of halogens.

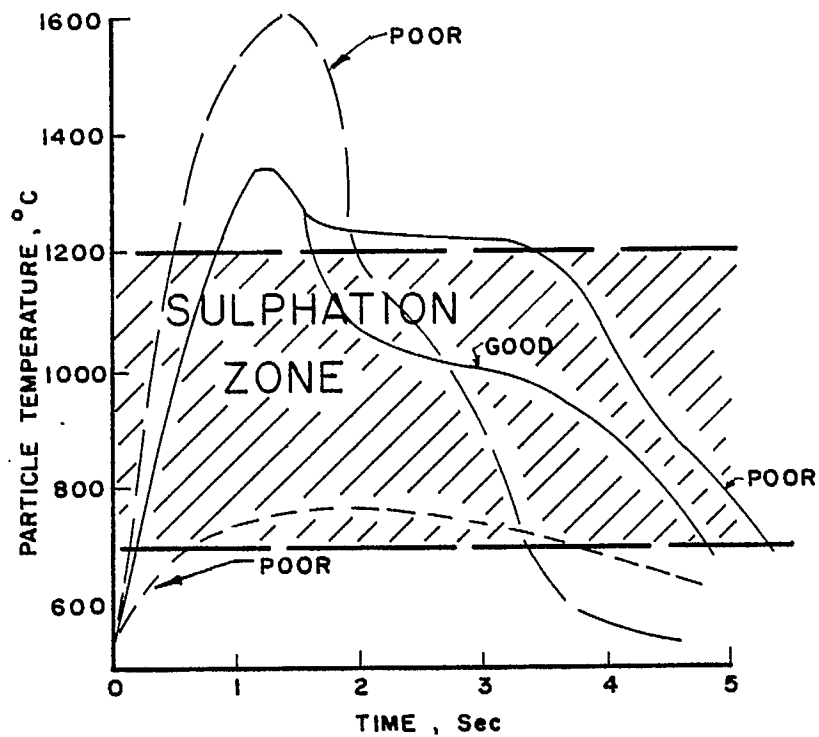
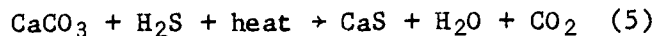
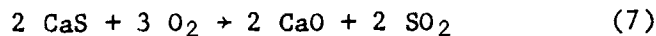


Fig. 2 - Time-temperature effect on limestone calcination

The mechanisms for sulphur reactions in a reducing atmosphere are less clear, but may follow the sequence of reactions below:



The CaS may then, if required, be sulphated or oxidized as follows:



In this process any sulphur in char that escapes the reducing zone would require neutralization in a subsequent oxidizing stage. Application of physically-staged combustion systems, which are in the embryonic stage of development, will depend on further elucidation of critical process parameters such as mixing and stoichiometric ratios of the primary and secondary combustion zones, sorbent injection points, volumetric heat release rates and char carryover from each stage.

Three important factors that are strongly system dependent, but which have not yet been fully investigated are:

- a) the slagging and fouling propensity of the fuel ash and sorbent in the various combustion and downstream heat transfer zones;
- b) the amount of char emitted into the post flame gases and dust collectors; and
- c) the flame shape with respect to furnace temperature gradients and geometry.

Research data indicate that 50% sulphur neutralization is possible with Ca/S ratios of 2/1 and that Ca utilization efficiencies of up to 30% are achievable.

CURRENT CANADIAN PROJECTS

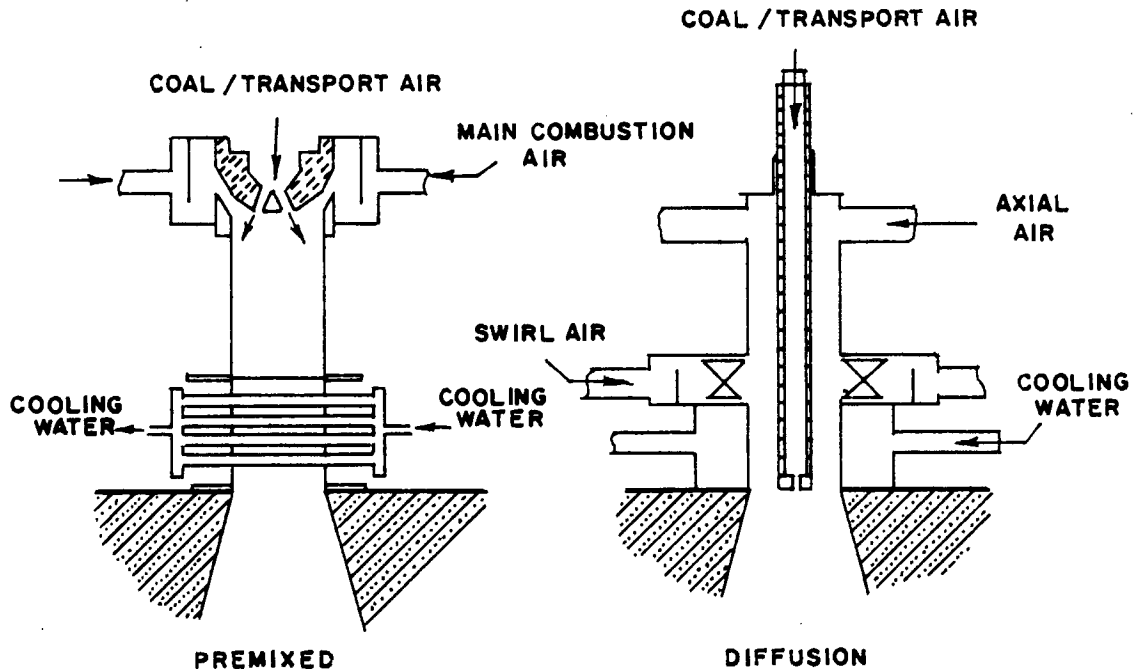
Two major Canadian initiatives in the simultaneous reduction of SO_x and NO_x from pulverized coal flames are being sponsored by the Canada Centre for Mineral and Energy Technology (CANMET). The first is an International Energy Agency (IEA) research project, co-funded by Canada, Denmark and Sweden with United States guidance and participation on the validation and optimization of advanced burner concepts. The second is a demonstration project in cooperation with the Canadian Department of National Defence (DND) at Canadian Forces Base Gaagetown, New Brunswick to accelerate the application of state-of-the art, staged-combustion burners to operating boilers. In addition to the above two projects, Energy Mines and Resources Canada collaborates with the Flue Gas Desulphurization Panel of the Canadian Electrical Association on utility R D & D programs for reducing NO_x and SO_x emissions from flames.

INTERNATIONAL ENERGY AGENCY PROJECT

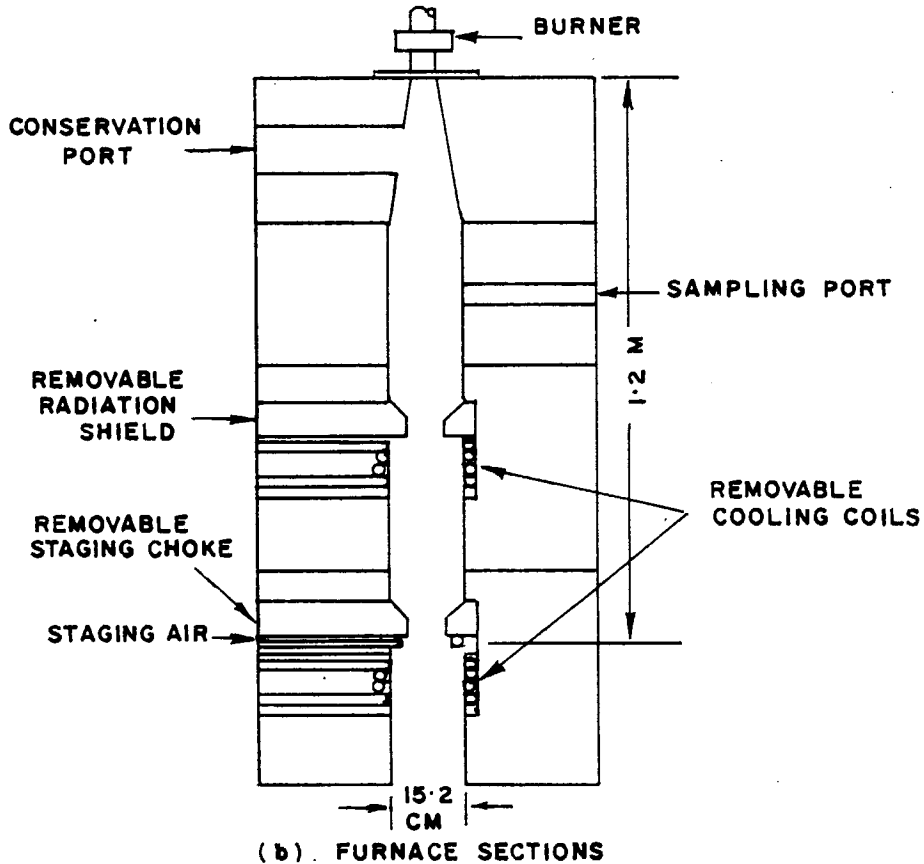
The IEA project, which is being performed by the Environmental Energy Research Corporation in the USA, was planned in three stages. (1) Stage 1, completed in March 1982, consisted of small-scale furnace trials on 45 coals including 9 from Canada, to elucidate the mechanisms of NO_x formation from fuel nitrogen under pre-mixed and stage-combustion conditions. Illustrations of the small-scale furnaces and burners are given in Fig. 3.

Highlights from this research indicate that for unstaged flames with 5% O_2 in the exhaust gases:

- a) more fuel nitrogen is converted to NO from pre-mixed than from axial diffusion flames, with the differences in NO production between the two systems decreasing from high-volatile bituminous to lignite to medium-volatile bituminous coals (Fig. 4).



(a) BENCH-SCALE BURNERS.



(b) FURNACE SECTIONS

Fig. 3 - Modified furnace and burner sections for staged combustion studies

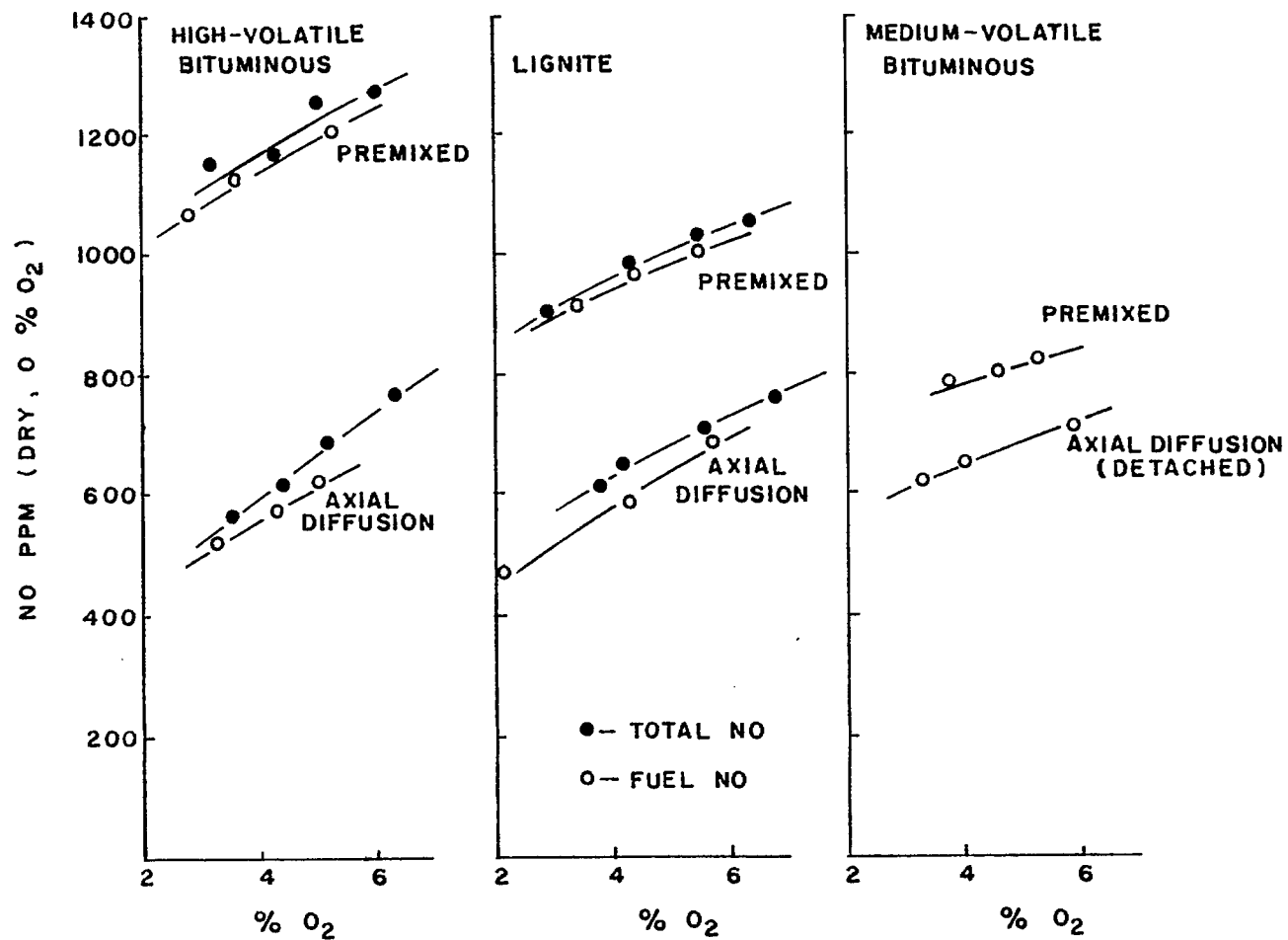


Fig. 4 - Fuel NO/Total NO for premixed and axial diffusion flames

- b) conversion of fuel nitrogen with pre-mixed flames ranged from 30% to 45% regardless of the total fuel nitrogen content (Fig. 5).
- c) high-volatile coals produced lower NO emissions from coarse particles with pre-mixed flames than with axial flames. Evidently, fuel nitrogen, which is evolved early in the flames from fine particles, is available for conversion to NO if oxygen is present (Fig. 6).

For staged flames, i.e., a sub-stoichiometric first stage and an air-rich second stage with 5% oxygen in the exit gases:

- a) all high-volatile coals, bituminous and lignitic, produced minimum NO emissions at a stoichiometric ratio of about 0.6 in the first stage; the medium-volatile coals yielded progressive increases in NO as the first stage stoichiometry increased.
- b) fuel nitrogen conversion ranged from 7% to 14% with the lowest NO emissions from high-volatile coals (Fig. 7).
- c) fine particles produced less NO than coarse particles for a high-volatile coal.
- d) the lower ranks coals, lignite and subbituminous, produced large amounts of NH_3 and HCN at stoichiometric ratios below 0.6. These species appear to favour transformation to N_2 rather than NO during second stage burn-out.
- e) heat extraction lowered NO emissions from low-rank coals, particularly for first stage stoichiometries above 0.7.

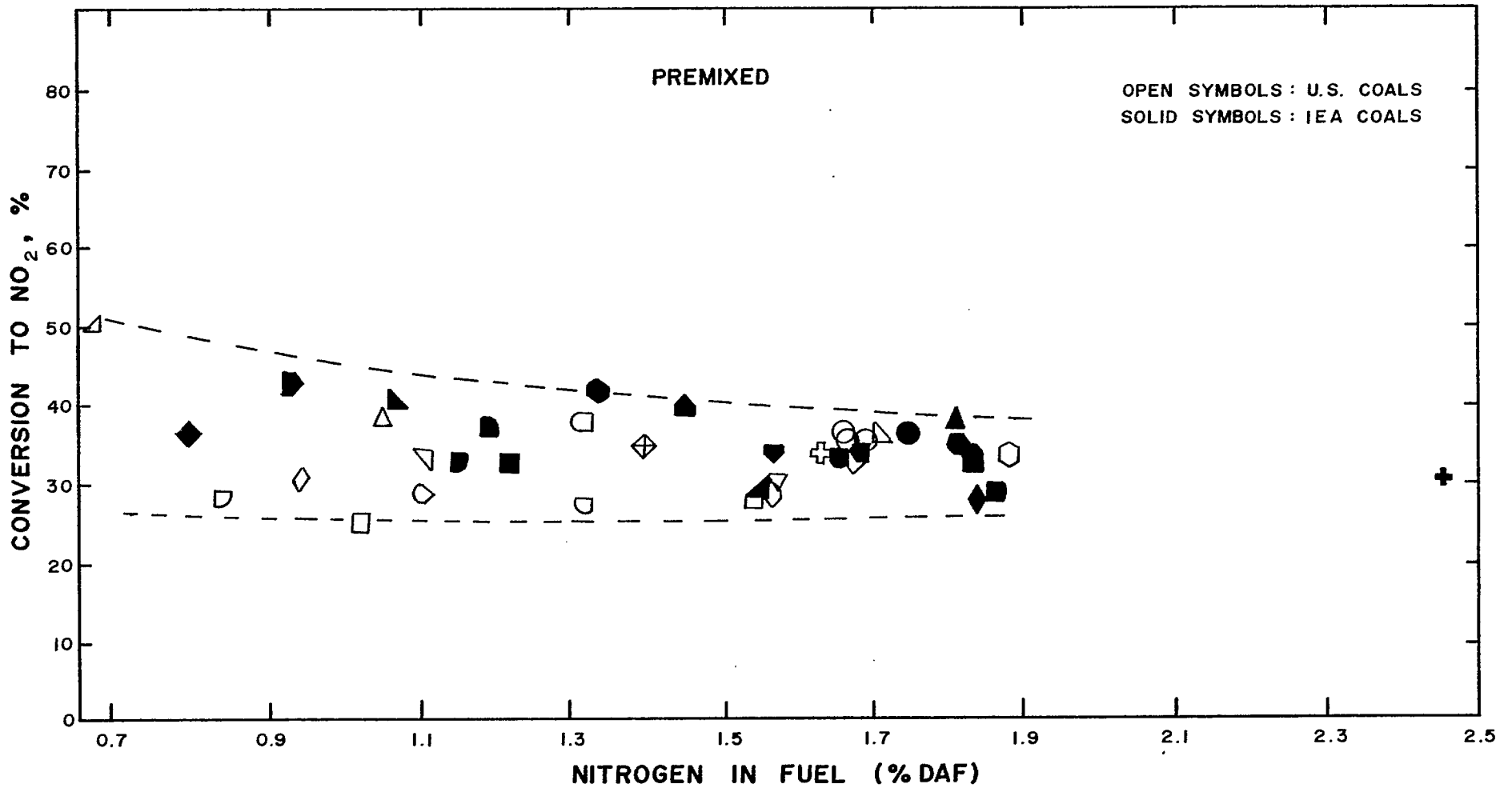


Fig. 5 - Fuel nitrogen conversion in premixed flames

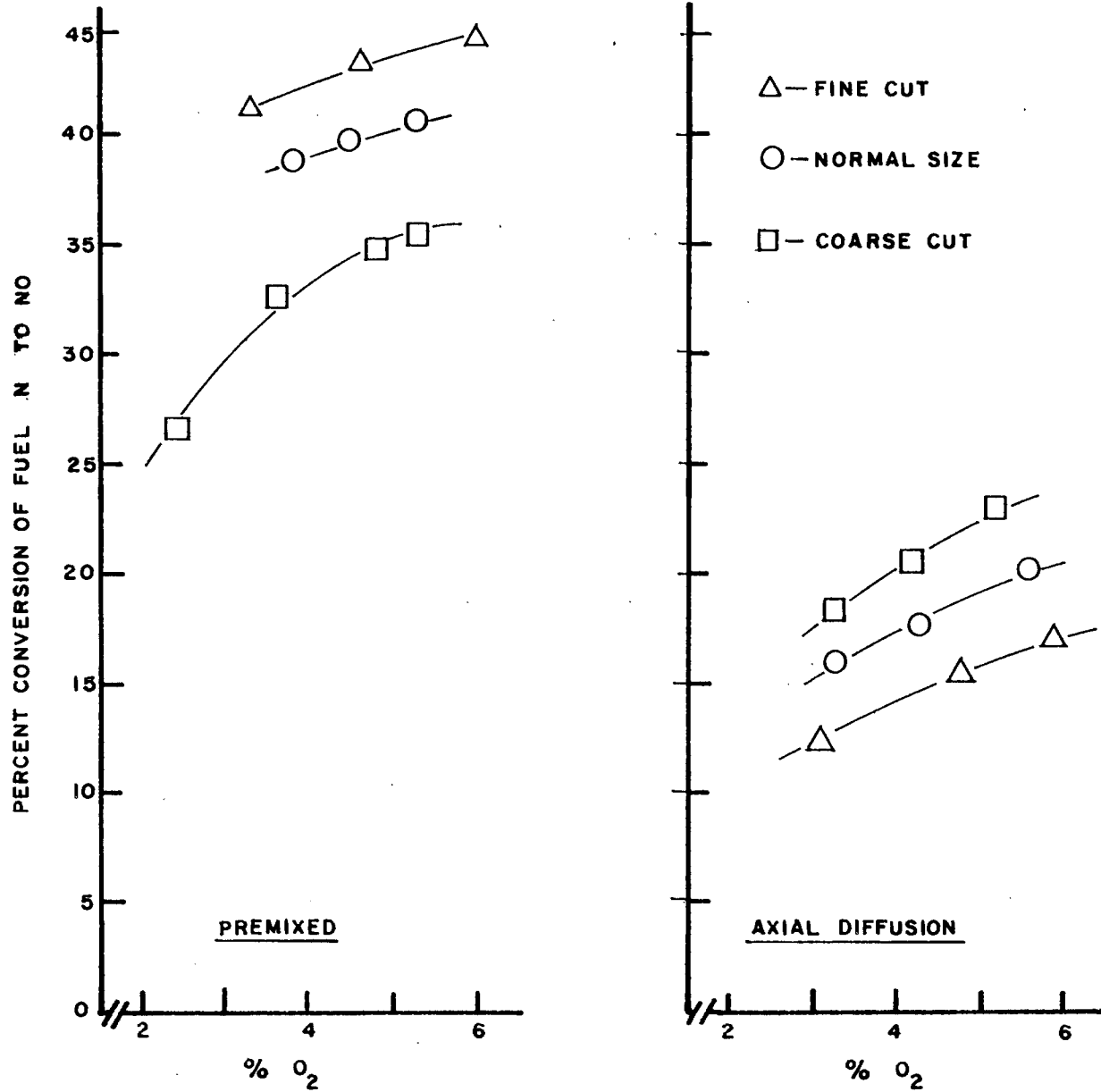


Fig. 6 - Influence of particle size, high volatile bituminous coal

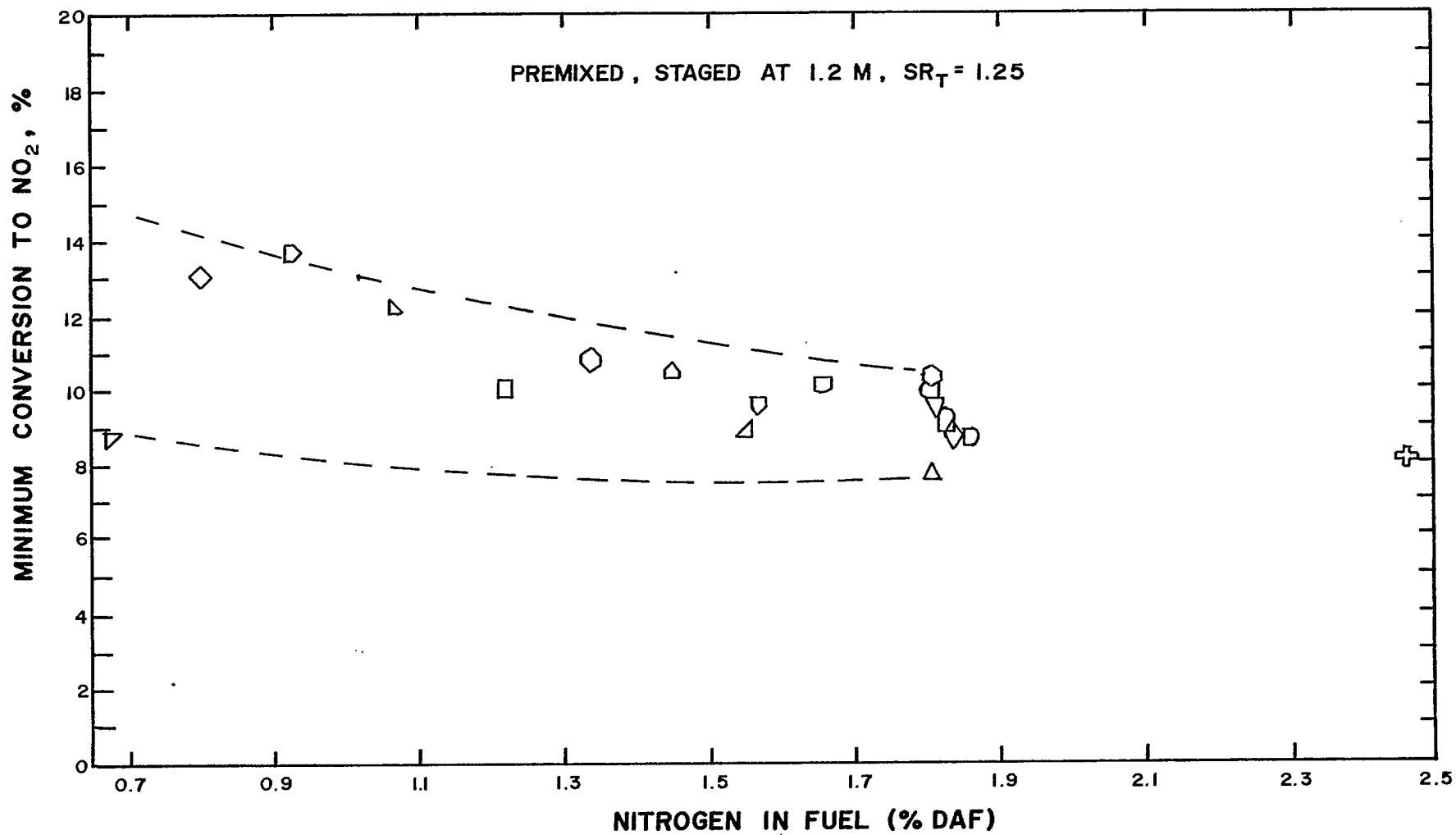


Fig. 7 - Minimum conversion of fuel nitrogen to NO under staged combustion conditions

GAGETOWN PROJECT

This project involves retrofitting staged pulverized coal burners on a 17 MW(th) front-wall-fired heating boiler at Gagetown, New Brunswick to reduce simultaneously emissions of SO_x and NO_x . The boiler is now fired with two conventional pulverized coal burners with one pulverizer per burner. Figure 8 shows an illustration of the proposed installation.

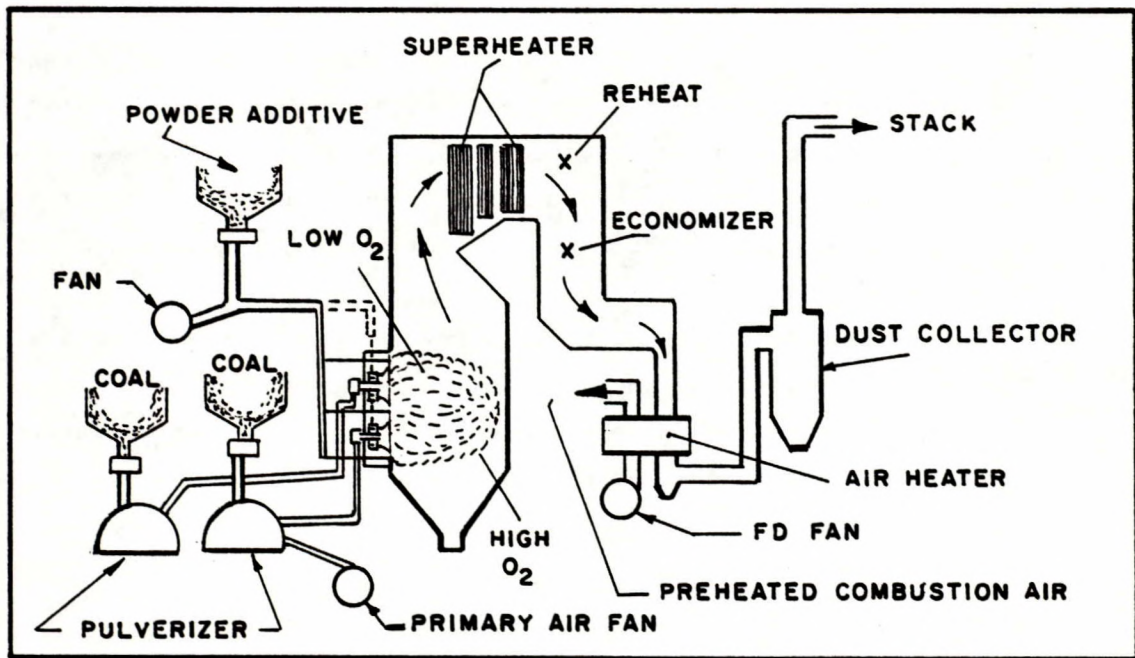
The staged burner design concept shown in Fig. 8 was installed in the spring of 1984 with commissioning and evaluation scheduled for the 1984/85 heating season.(2) The retrofitted burners are designed to achieve a 50% reduction in both NO_x and SO_x levels at full load with:

- a) 3% high-volatile bituminous coal;
- b) Ca/S ratios of 2.5 or less;
- c) combustible in ash of 7% or less;
- d) no flame impingement on walls;
- e) boiler efficiency of 82% or more and
- f) no furnace slagging with routine soot-blowing.

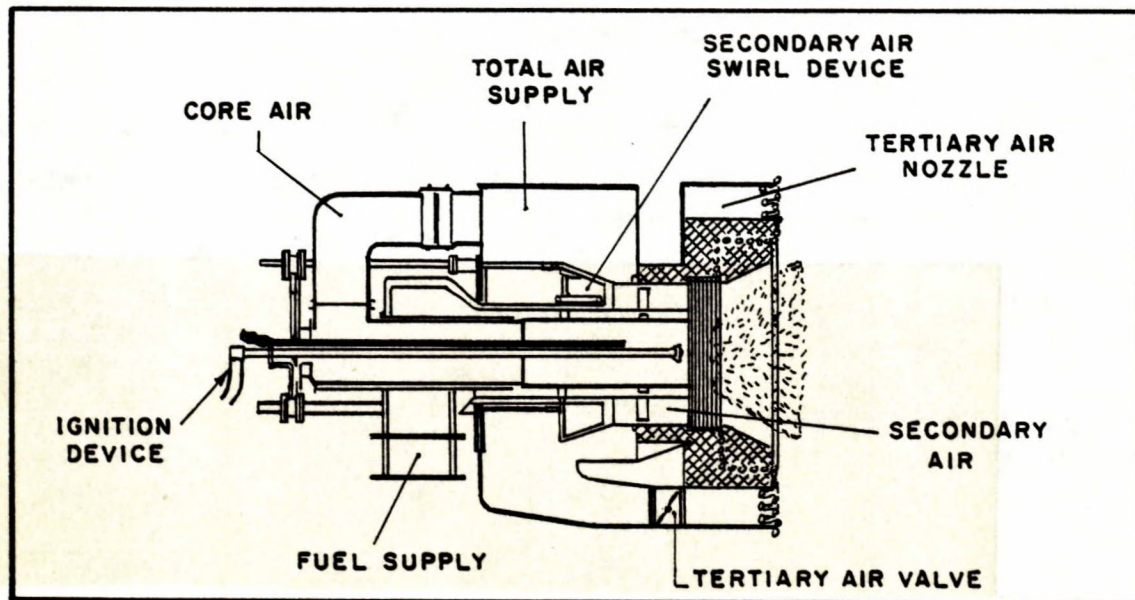
CANADIAN ELECTRICAL ASSOCIATION STUDY

A research project to study the feasibility of retrofitting a 350 MW, lignite-fired utility boiler in Western Canada with a low NO_x/SO_x burner system was recently completed. The tangentially-fired system was modified with staged burners designed to provide a flame zone with a sub-stoichiometric core and an oxidizing outer annulus. The sulphur sorbent was injected through the lower coal burners to maximize residence time and to provide the optimum temperature for calcination.(3)

At full load with 3% O_2 in the flue gas, NO_x levels were reduced from 425 ppm to 200 ppm with about a 40% reduction in sulphur emissions at a Ca/S ratio of 2.5.(3) Although lignite typically contains less than 1% sulphur on a moisture free basis, Western Canadian utilities now generate over 5500 MW of electricity from low-sulphur, low-rank coals and are major regional sources of NO_x and SO_x emissions.



a) Schematic of Gagetown boiler system



b) Staged mixing burner (SM burner)

Fig. 8 - Illustration of boiler and burner systems for Gagetown NB

The research programs, although still in the early stages of development, indicate that staged-combustion concepts can be used to reduce simultaneously both SO_x and NO_x emissions in boiler furnaces. Staged combustion may however be only marginally effective in reducing NO_x emissions from coals in which most of the fuel nitrogen is present in the fixed carbon.

The demonstration projects involving burner retrofits are directed at accelerating the transfer of research results to commercial application and in validating staged burner concepts for reducing NO_x and SO_x emissions under operational conditions.

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2. L & C Steinmuller GmbH, "Design study for pulverized coal burner for the combined reduction of nitrogen and sulphur oxides", Gummersbach, West Germany, March 1983.
3. Winship, R.D., Morrison, S.A., Clements, B.R., and Lee, B.Y. " SO_2 and NO_x reduction by combustion modification and furnace dry sorbent injection." Canadian Electrical Association 1984 Spring Meeting Toronto, March 1984.