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CONTROL OF NO_x AND SO_x EMISSIONS FROM PULVERIZED-COAL FLAMES

G. K. Lee, F. D. Friedrich and H. Whaley
Combustion and Carbonization Research Laboratory

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FROM PULVERIZED-COAL FLAMES

by

G. K. Lee*, F. D. Friedrich** and H. Whaley**

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 30% to 40% in conventional flames but only 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 stage combustion systems in a front-wall-fired and a tangentially-fired boiler.

*Manager, **Research Scientist, Combustion and Carbonization Research Laboratory, Energy Research Laboratories, CANMET, Energy, Mines and Resources Canada K1A 0G1

INTRODUCTION

One of the major concerns, associated with the expanded use of coal for heat and electricity, is the emissions of acid rain precursors, NO_x and SO_x , to 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 the NO_x and 5% of the 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 CANMET/ERL initiatives to suppress simultaneously the generation of NO_x and SO_x by staged burner aerodynamics.

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; Table 1 gives an indication of the two NO_x sources when different fuels are burned in well-mixed flames.

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 lean 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 first stage followed by controlled additions of secondary air and injections of tertiary air to progressively stage the combustion process as shown in Figure 1 (a) , 1 (b), and 2 (a) respectively.

During coal combustion, upwards of 70% of the total NO_x emissions are derived from fuel nitrogen which forms N_2 , HCN, NO and NH_3 during sub-stoichiometric devolatilization. About 1/5 to 1/3 of the fuel nitrogen may 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, Figure 2 (b):

- (1) a fuel is burned in the lower furnace under slightly air rich conditions;
- (2) additional fuel is then 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 and
- (3) finally, tertiary air is injected into the fuel rich products leaving the reburning zone to complete the combustion process under slightly air rich conditions.

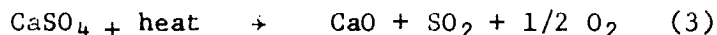
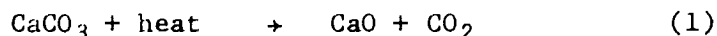
SO_x Emissions

Sulphur in fuel, regardless of the form present, 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 final combustion 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.

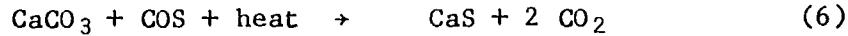
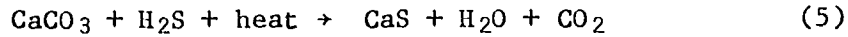
When injected into an oxidizing flame zone, the sulphur/sorbent reactions are as follows:



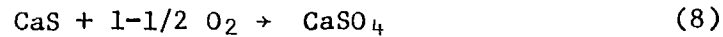
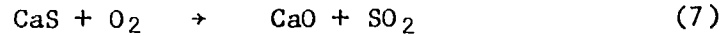
Decarbonation at temperatures above 1000°C and below 1400°C, Equation 1, produces an activated CaO. But if heated above 1350°C, the CaO is rapidly deactivated by dead burning which significantly reduces the specific surface available for reaction with sulphur. Overheating of the CaSO₄ end product is also undesirable because at temperatures above 1350°C, CaO and SO₂ are regenerated. As illustrated in Figure 3, the best SO_x capture will occur when the sorbent is a) injected into an oxidizing region which allows calcination without deactivation, b) sulphation without regeneration and c) 2 to more seconds of contact time at reaction temperatures between 750°C and 1200°C.

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.

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



The CaS may then be rendered inert or be 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, 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 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 co-operation with the Department of National Defence (DND) at Canadian Forces Base Gagetown 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 Desulphurization Panel of the Canadian Electrical Association on utility R D & D programs for reducing NO_x and SO_x emissions from flames.

IEA 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 staged combustion conditions. The analyses of the Canadian coals are given in Table 2 and illustrations of the small scale furnaces and burners is given in Figures 4 and 5 respectively.

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. Figures 6, 7 and 8;

- b) conversion of fuel nitrogen with pre-mixed flames ranged from 30% to 45% regardless of the total fuel nitrogen content, Figure 9.
- c) high-volatile coals produced lower NO emissions from coarse particles with pre-mix 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, Figure 10.

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. Figure 11.
- b) fuel nitrogen conversion ranged from 7% to 14% with the lowest NO emissions from high volatile coals, Figure 12.
- c) fine particles produced less NO than coarse particles for a high volatile coal, Figure 13.
- d) the lower rank coals, lignite and sub-bituminous, 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, Figure 14.
- e) heat extraction lowered NO emissions from low-rank coals, particularly for first stage stoichiometries above 0.7. Figure 15.

Gagetown Project

This project, involves retrofitting staged pulverized coal burners on a 17 MW 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 16 shows an illustration of the proposed installation.

Two staged burner design concepts, Figures 17 and 18, are under consideration and plans call for one of these designs to be installed and evaluated by mid-1984. The retrofitted burners are being designed to achieve a 50% reduction in both NO_x and SO_x levels at full load with:

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

Canadian Electrical Association Study

A research project is currently underway 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. The tangentially-fired system would be modified with staged burners designed to provide a flame zone with a sub-stoichiometric core and an oxidizing outer annulus. By injecting the sulphur sorbent into the outer annulus, SO_2 emissions and furnace fouling should both be reduced substantially.

Although the 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.

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 retrofit are directed at accelerating the transfer of research results to commercial application and in validating staged burner concepts for preventing NO_x and SO_x emissions under operational conditions.

REFERENCES

1. Chen, S. L., Pershing, D. W. and Heap, M. P. "International Energy Agency Project for control of nitrogen oxides emission during coal combustion:Stage 1"; Prepared for the Executive Committee of the Contracting Parties for Canada, Denmark and Sweden, December 1981; G. K. Lee and W. A. Warfe, Executive Committee Members for Canada.

Table 1 - NO_x sources for various fuels

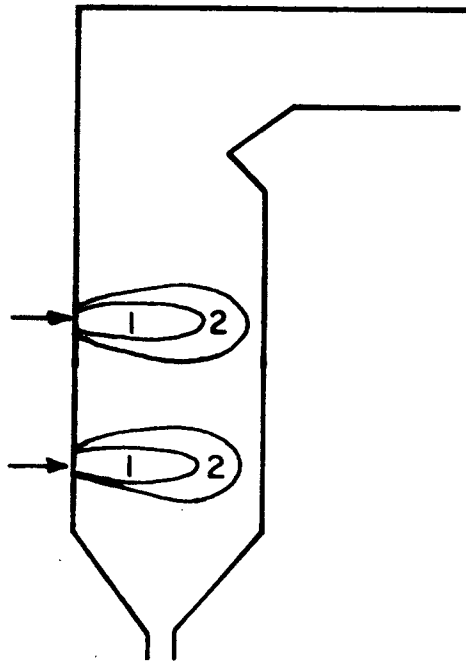
COMBUSTION - SOURCE NO_x
(CONVENTIONAL BURNERS)

Fuel Type	Fuel N % wt	NO _x Source, % Total	
		Thermal	Fuel
Gas	0	100	0
No. 2 Oil	0.2	60	40
No. 6 Oil	0.8	20	80
Hi-Vol Coal	1.8	15	85
Lo-Vol Coal	1.8	30	70

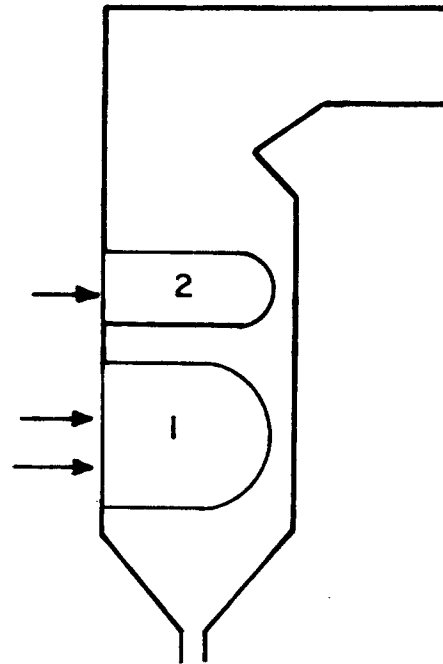
SYMBOL	◆	■	▲	▼	⬡	●	⬠	◐	◑
M*	30	27	14	7	4	1	1	2	1
A*	14	10	15	11	11	19	7	16	15
VM	28	30	33	34	35	30	34	20	21
FC*	28	33	38	48	50	50	58	62	63
S	0.9	0.4	0.6	0.3	1.2	2.4	1.9	0.3	0.2
N	0.6	1.1	1.3	0.9	1.2	1.4	1.7	1.0	1.0
C/H	18	16	15	16	15	14	15	18	18
MJ/KG*	14.3	17.6	19.3	25.2	27.7	27.7	32.8	27.7	30.2
RANK	LIG	SUB	SUB	HVB	HVB	HVB	HVB	MVB	MVB

*AS REC'D

TABLE 2 - COAL ANALYSES



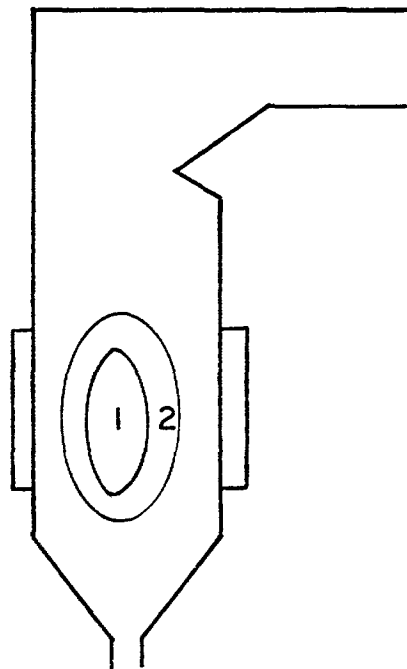
A) BURNER STAGING



B) FURNACE STAGING

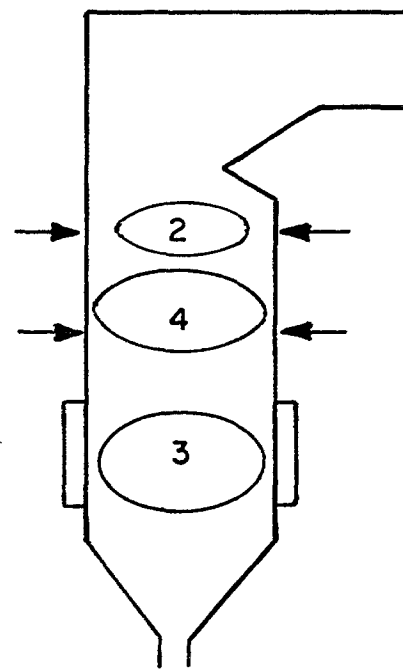
1 = 65 % TOTAL AIR
 2 = 25 % TOTAL AIR

Fig. 1 - Burner and furnace staging



A) STAGED BURNERS
AND FURNACE

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



B) STAGED FUEL
REBURNING

3 = 10 % TOTAL AIR
4 = 95 % TOTAL AIR

Fig. 2 - Burner/furnace and fuel reburning staging

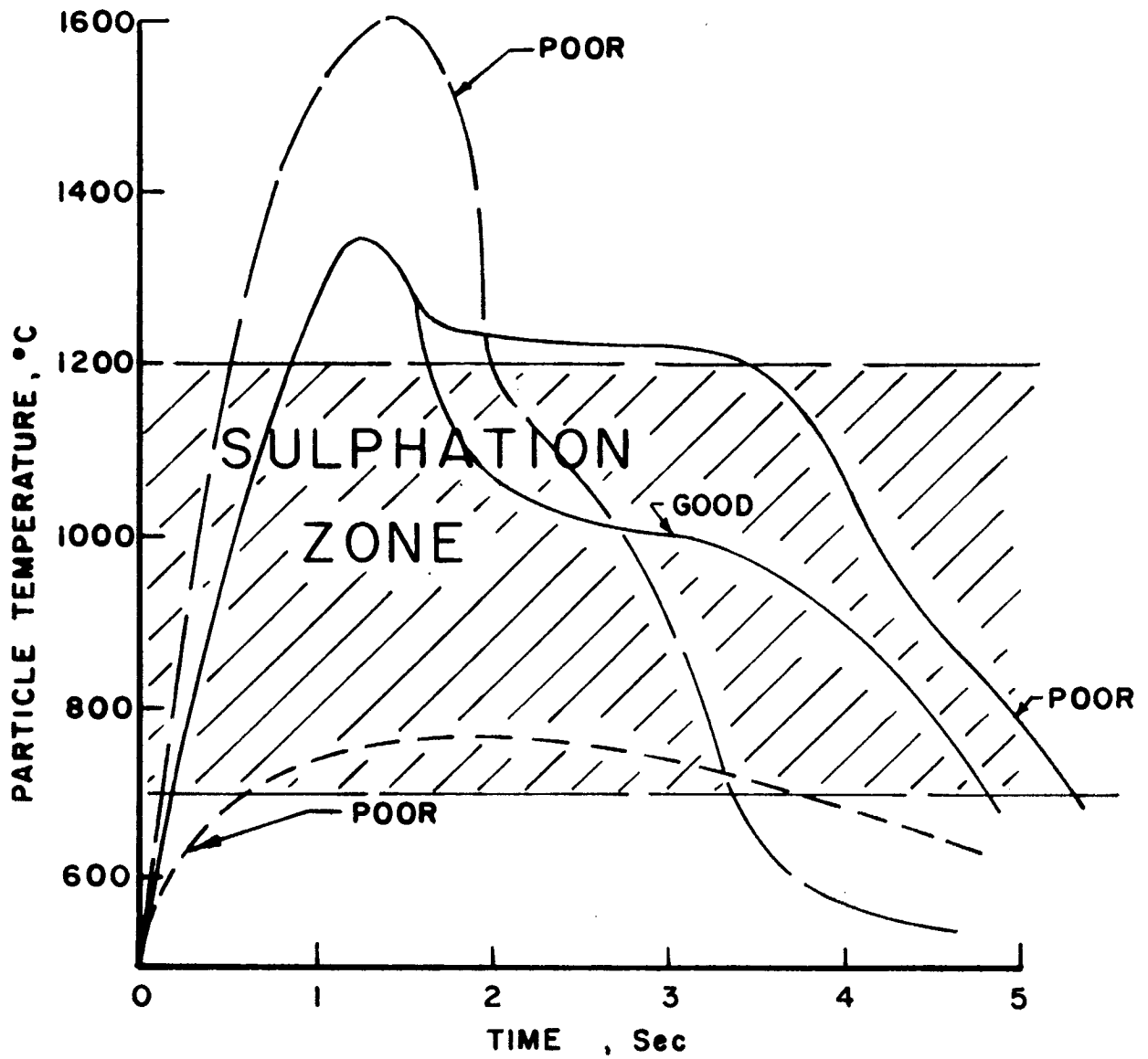


Fig. 3 - Time-Temperature Effect on Limestone Calcination

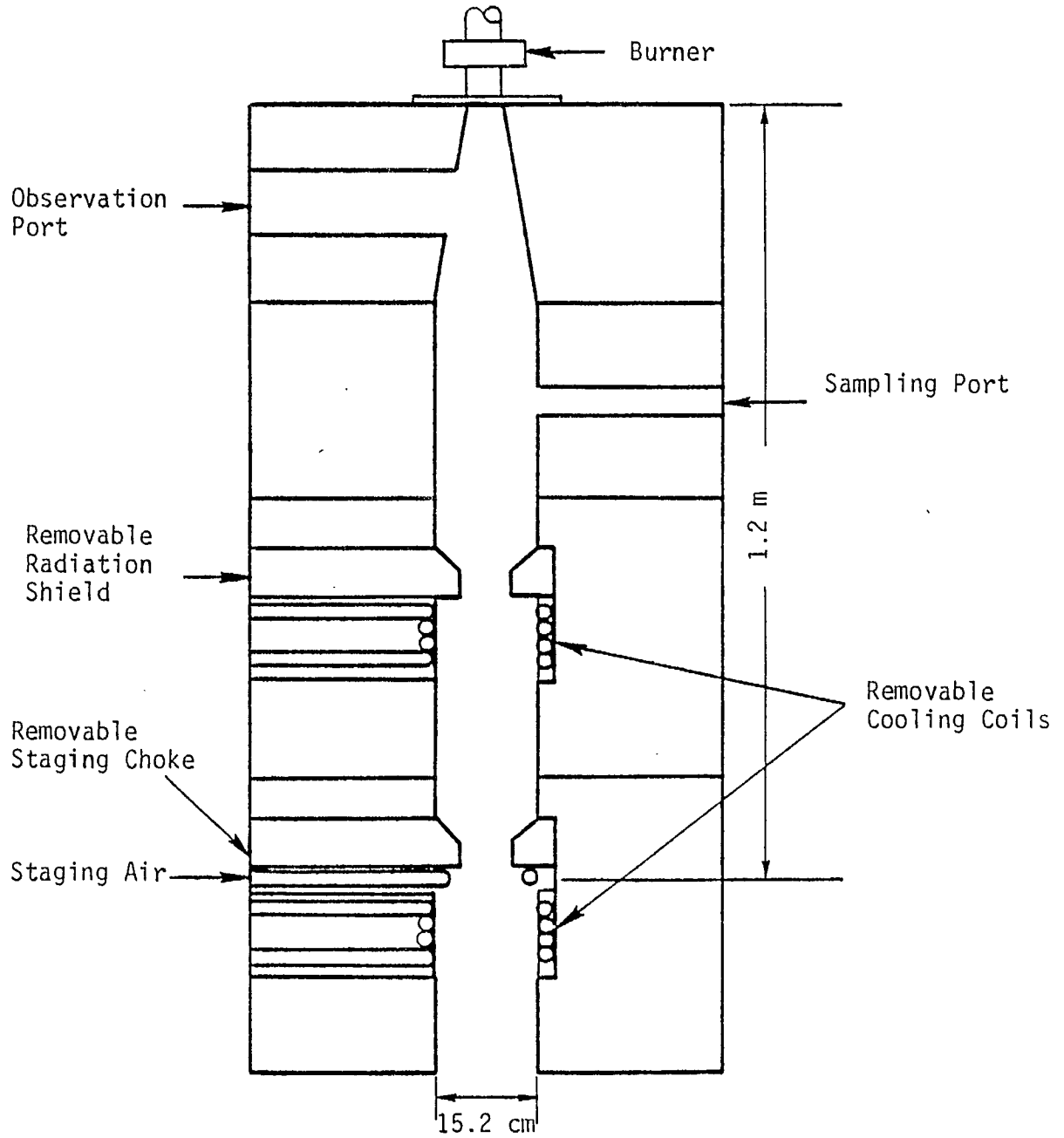


Figure 4 - Modified Furnace Sections for Staged Combustion Studies.

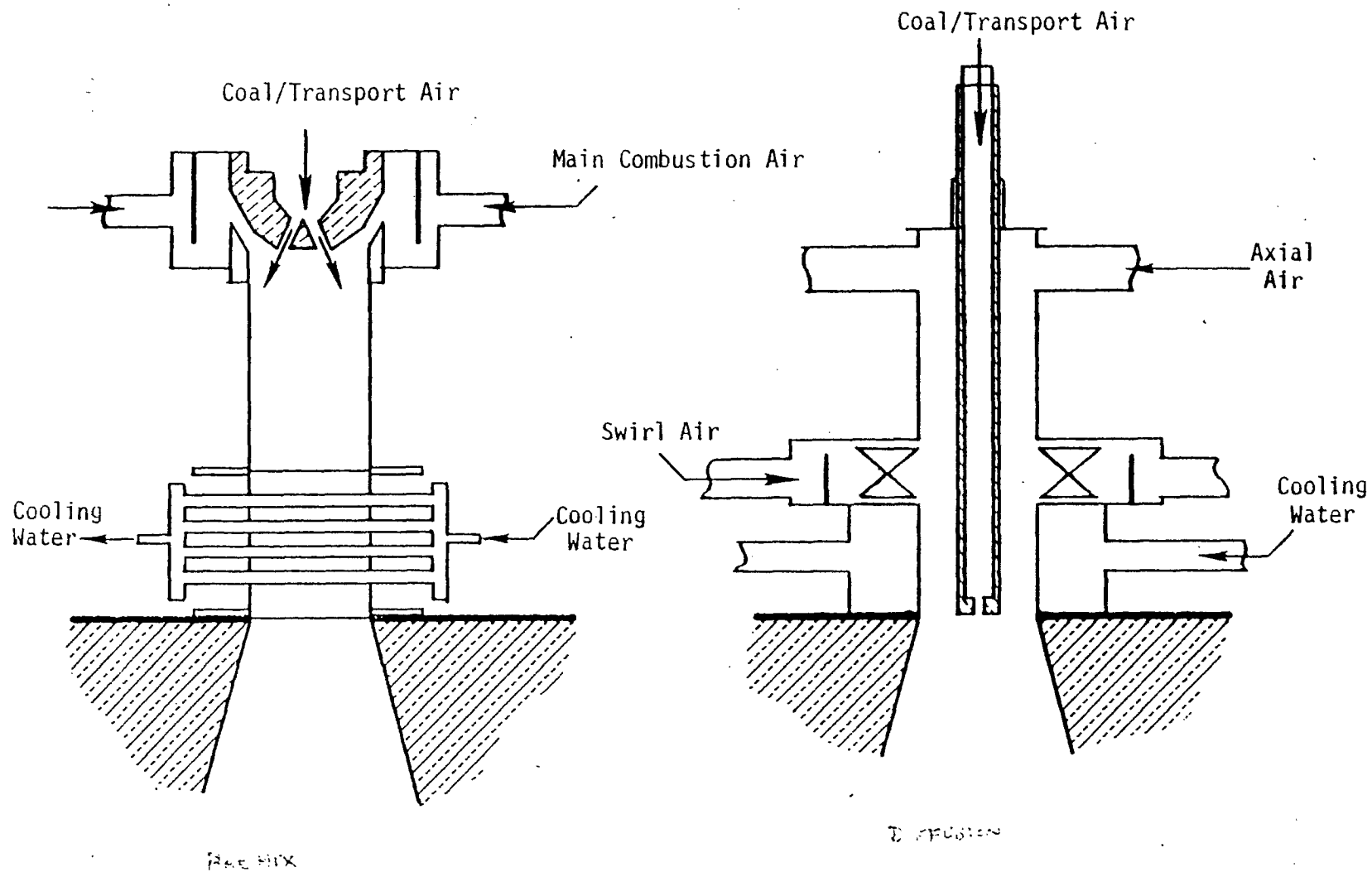


Figure 5 - Bench-Scale Burners.

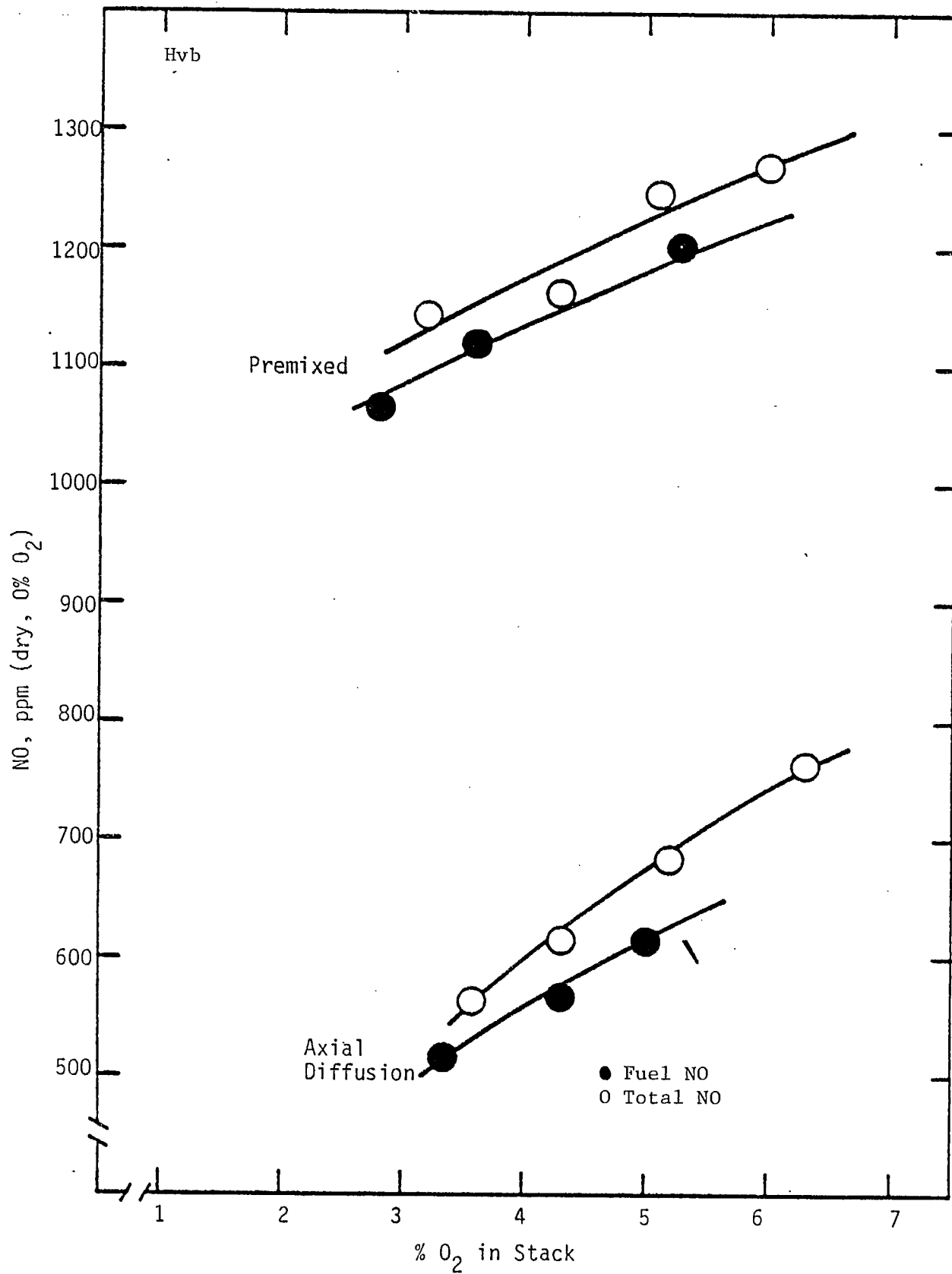


Fig. 6 - Fuel NO to total NO for premixed and axial flames, high volatile bituminous coal

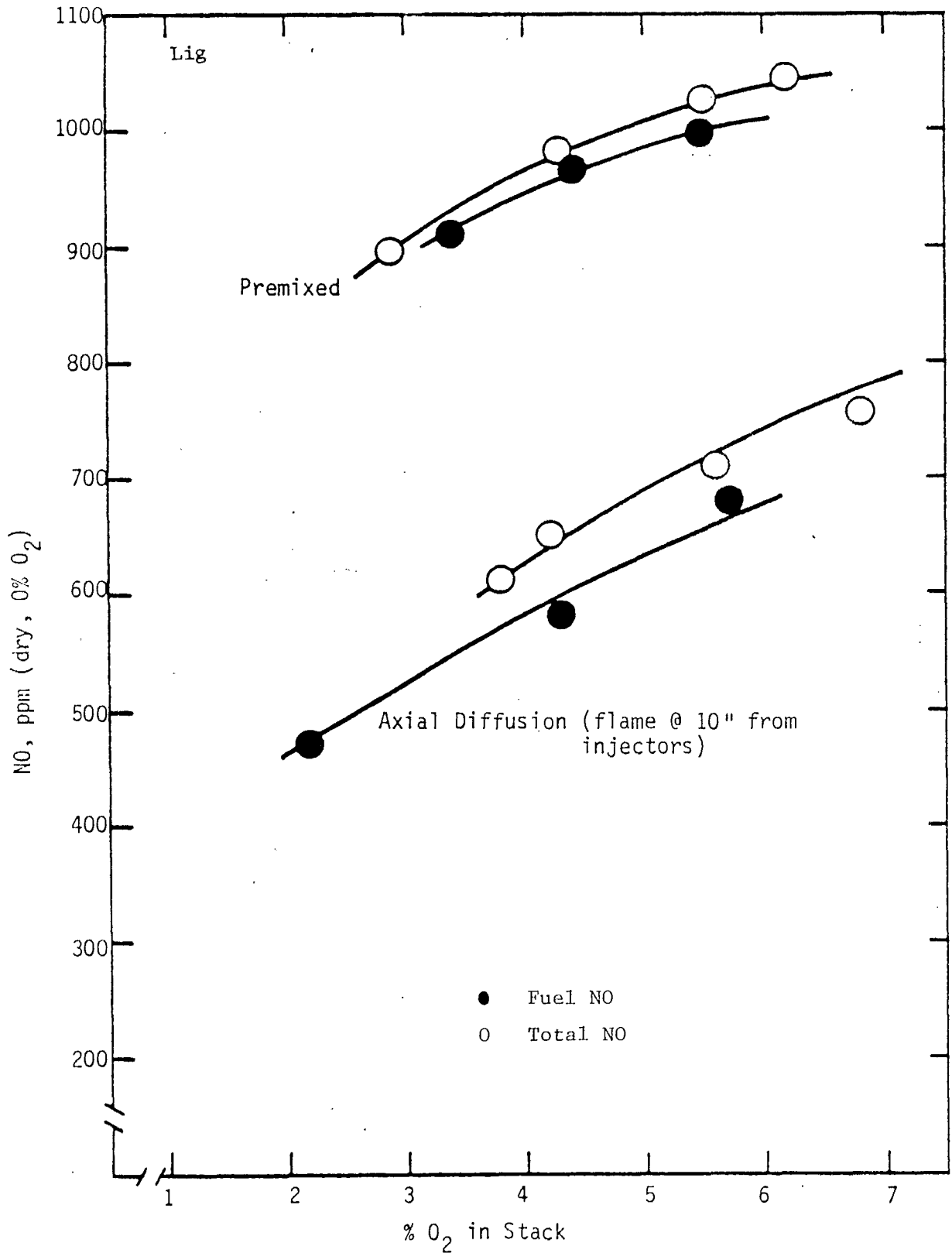


Fig. 7 - Fuel NO to total NO for premixed and axial flames, lignite

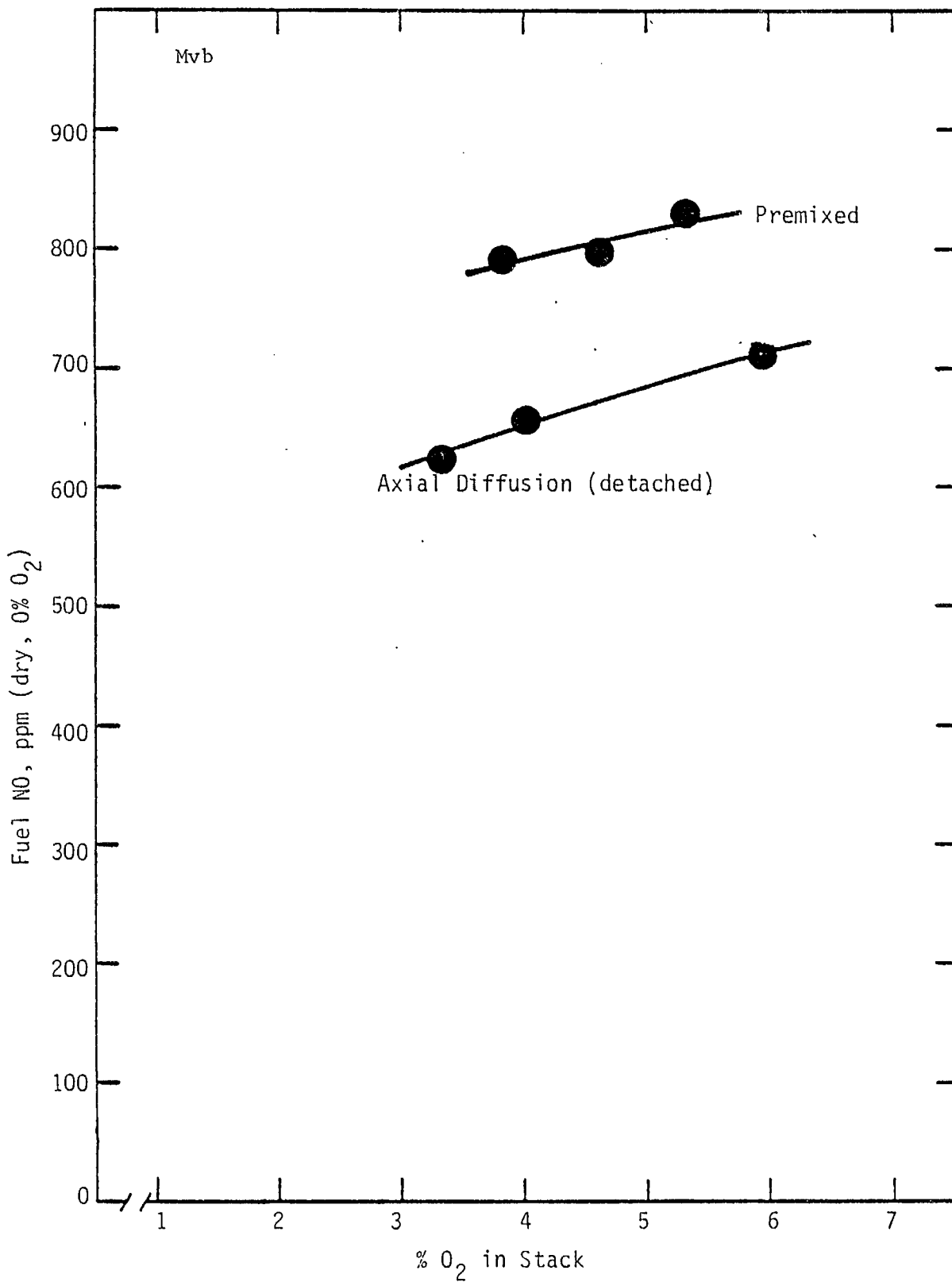


Fig. 8 - Fuel NO for premixed and axial flames, medium volatile bituminous coal

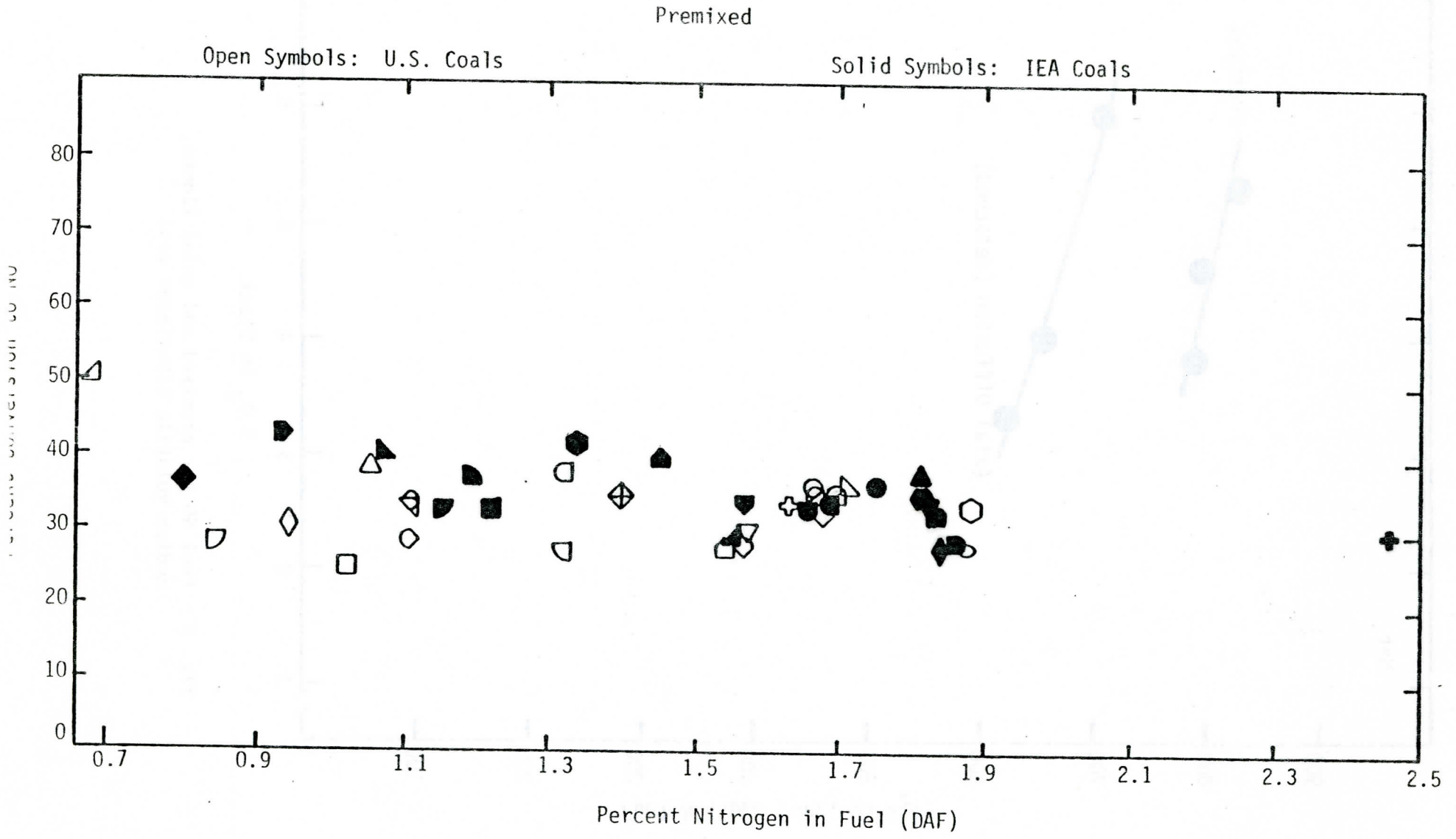


Figure 9 - Fuel Nitrogen Conversion in Premixed Flames

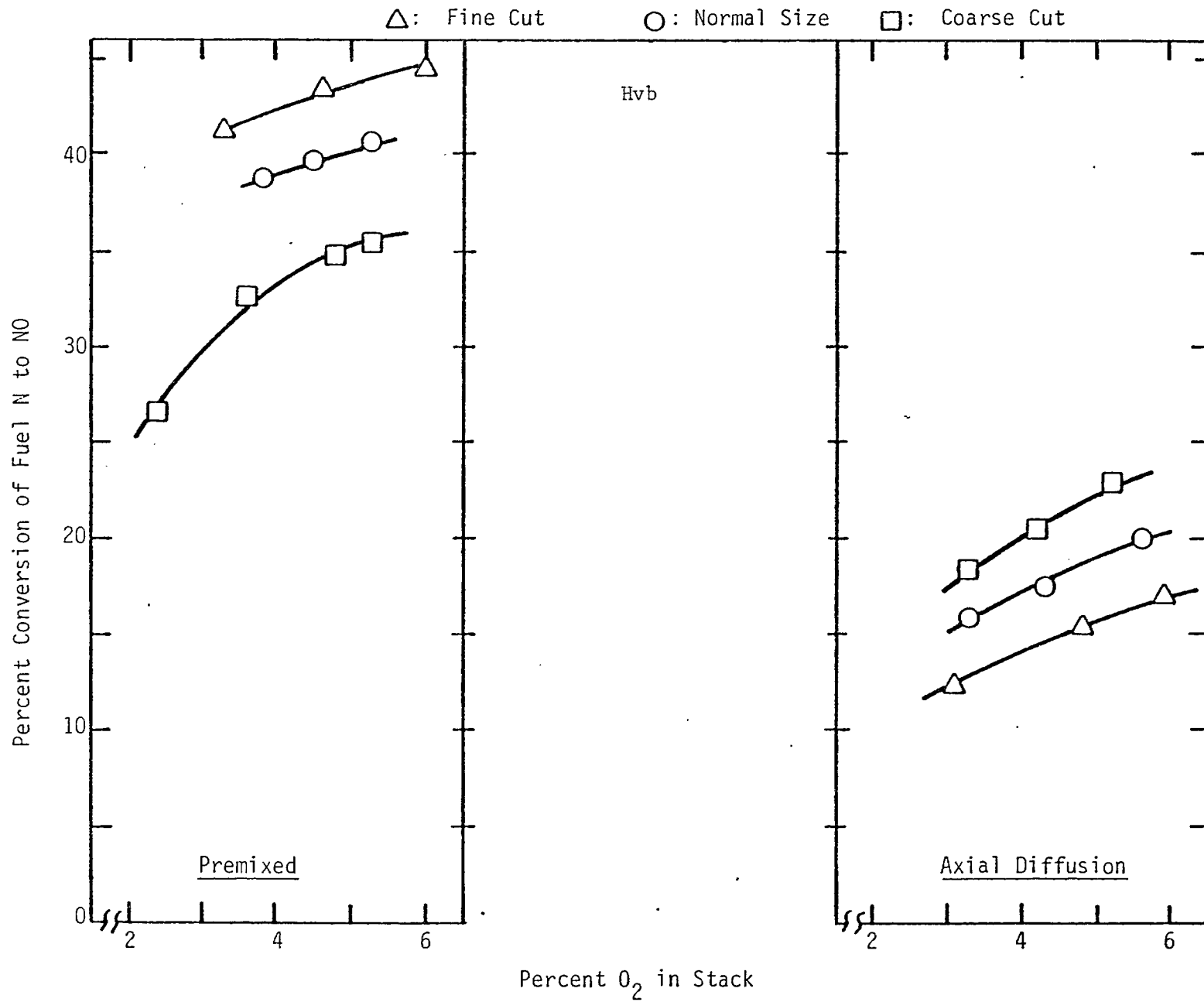


Fig. 10 - Influence of Particle Size.

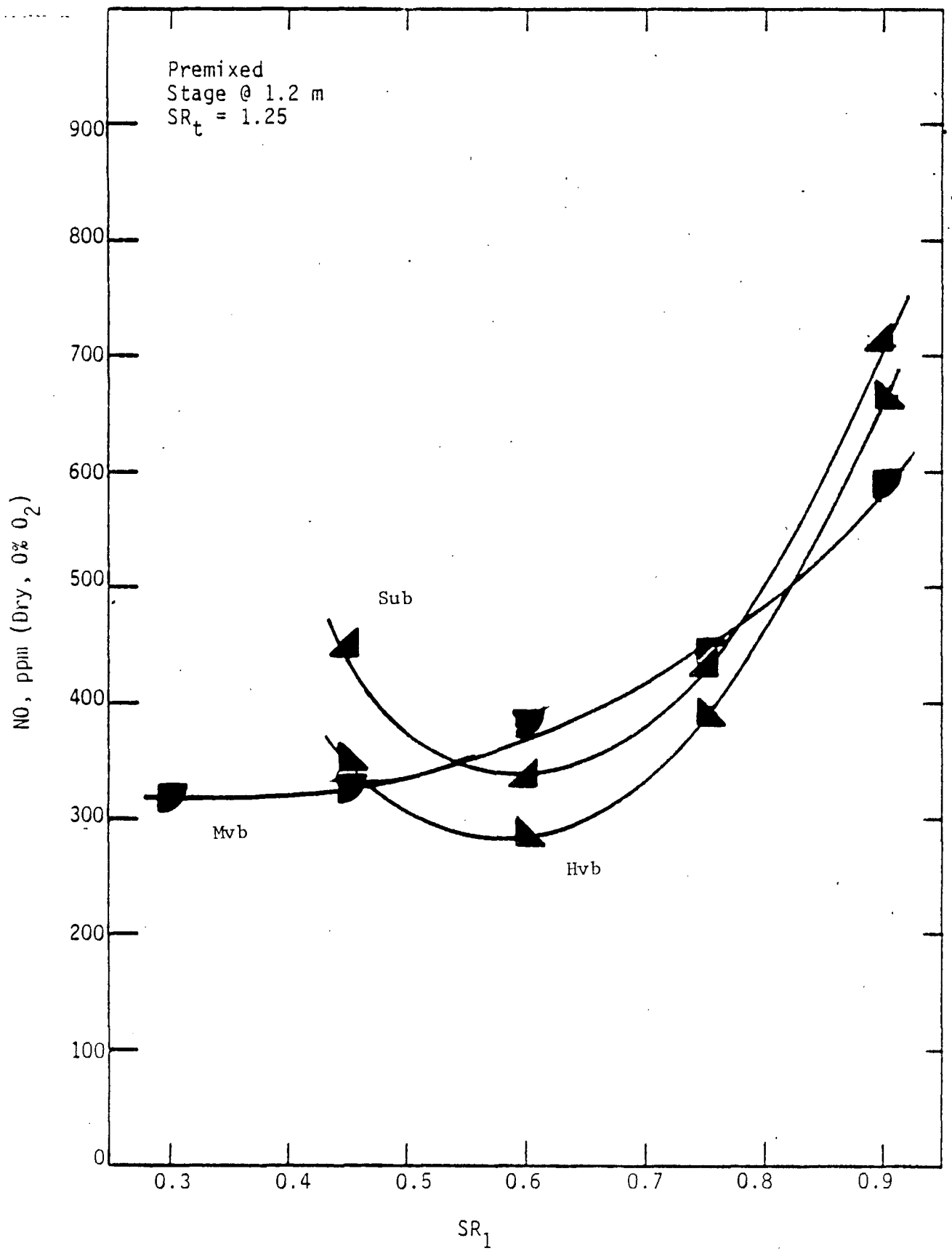


Fig.11- Influence of First Stage Stoichiometry on Exhaust NO Emissions

Premixed, Staged @ 1.2 m, $SR_t = 1.25$

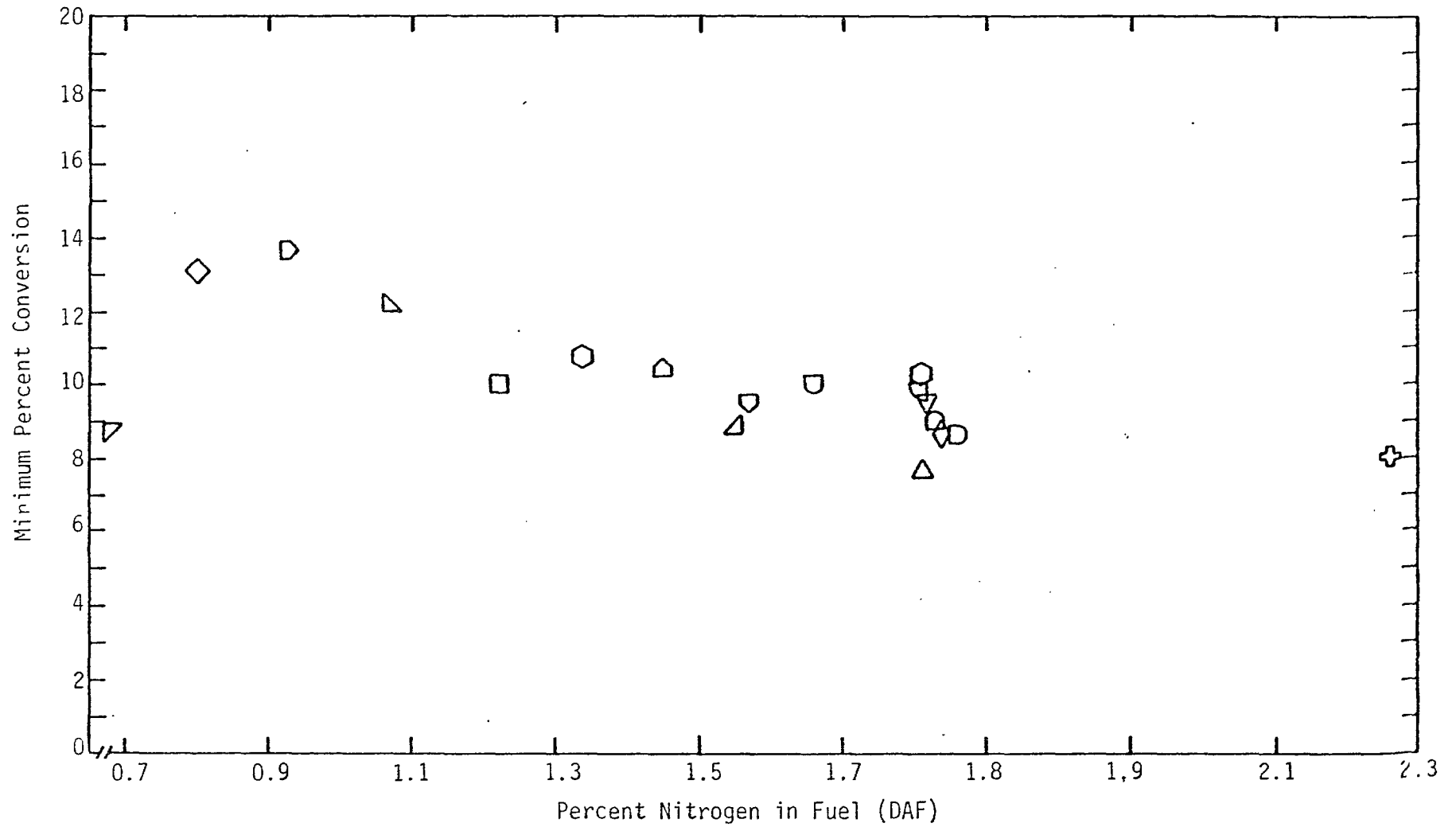


Fig.12- Minimum Conversion of Fuel Nitrogen to NO Under Staged Combustion Conditions.

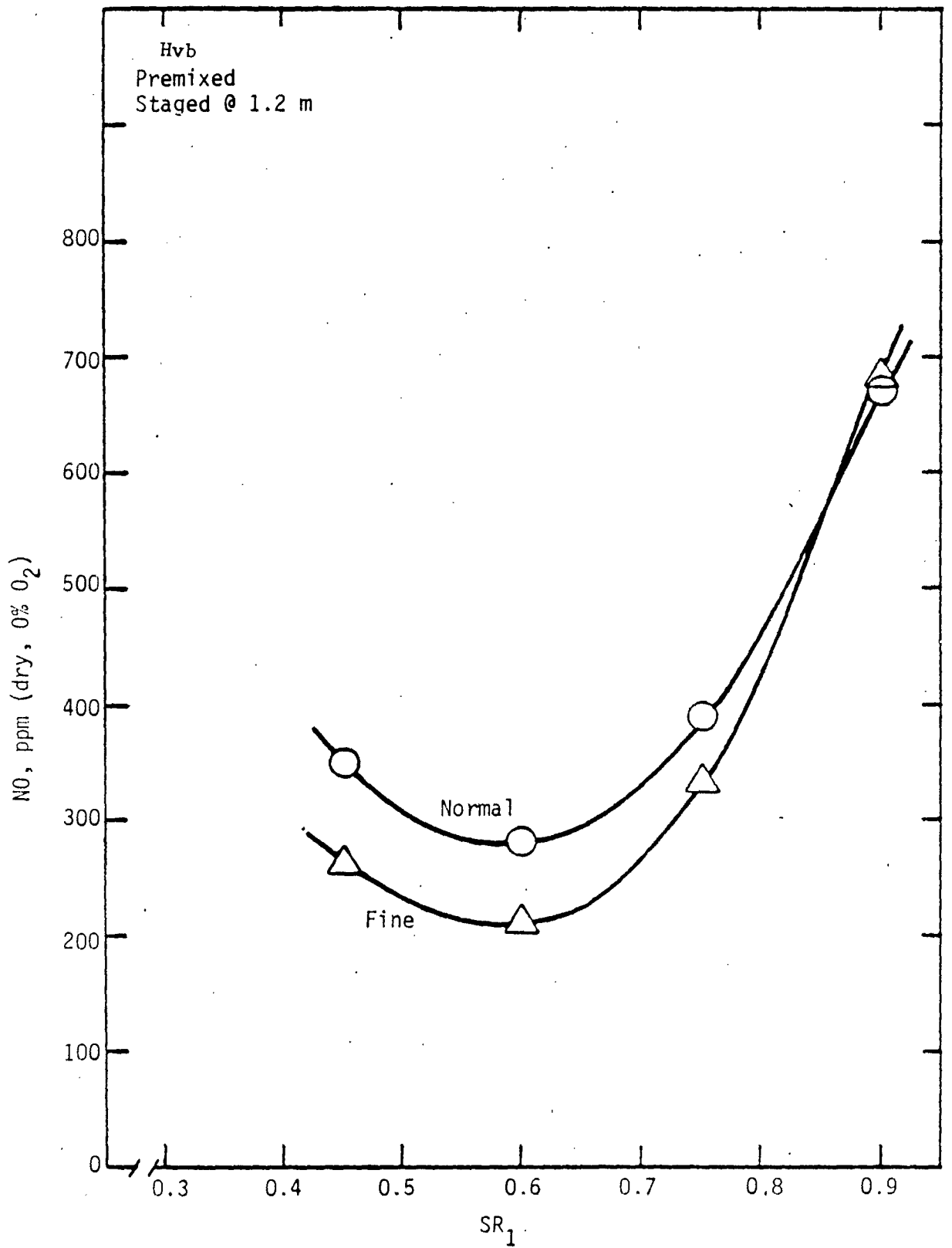


Figure 13 - Influence of Particle Size on NO Emissions.

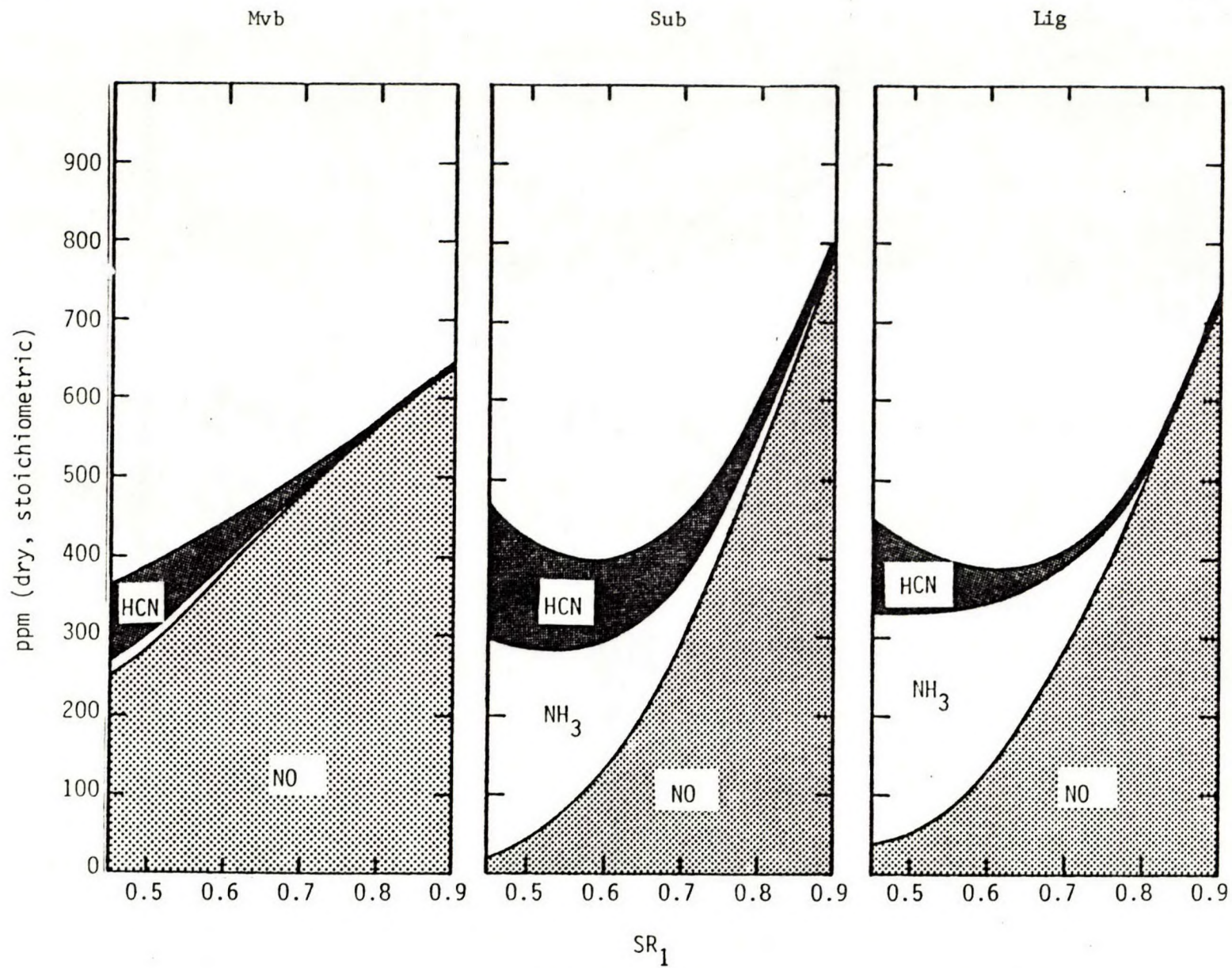


Fig. 14 - TFN Distribution.

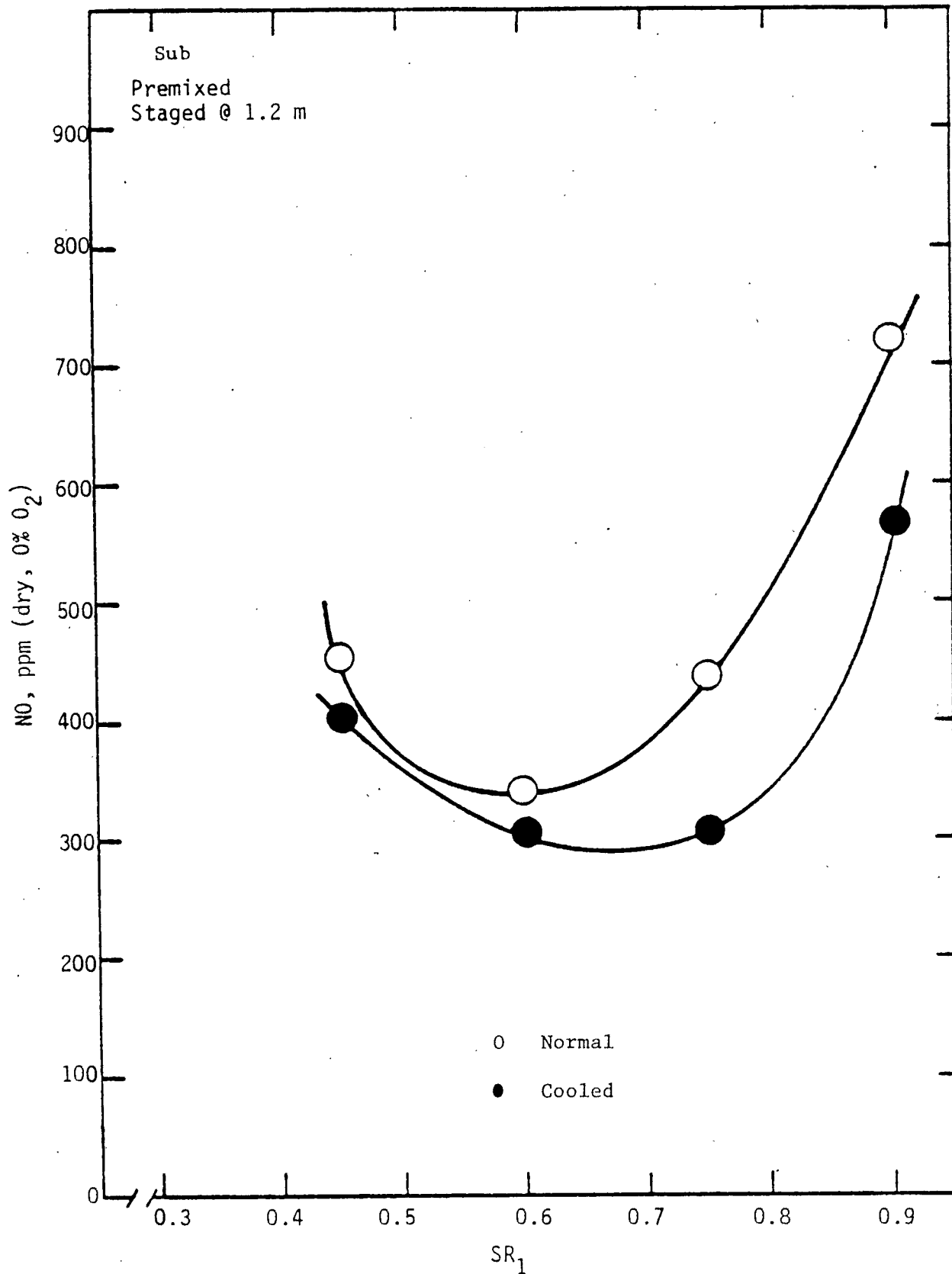


Fig. 15 - NO emissions from staged combustion, with and without cooling, low-ranked coals

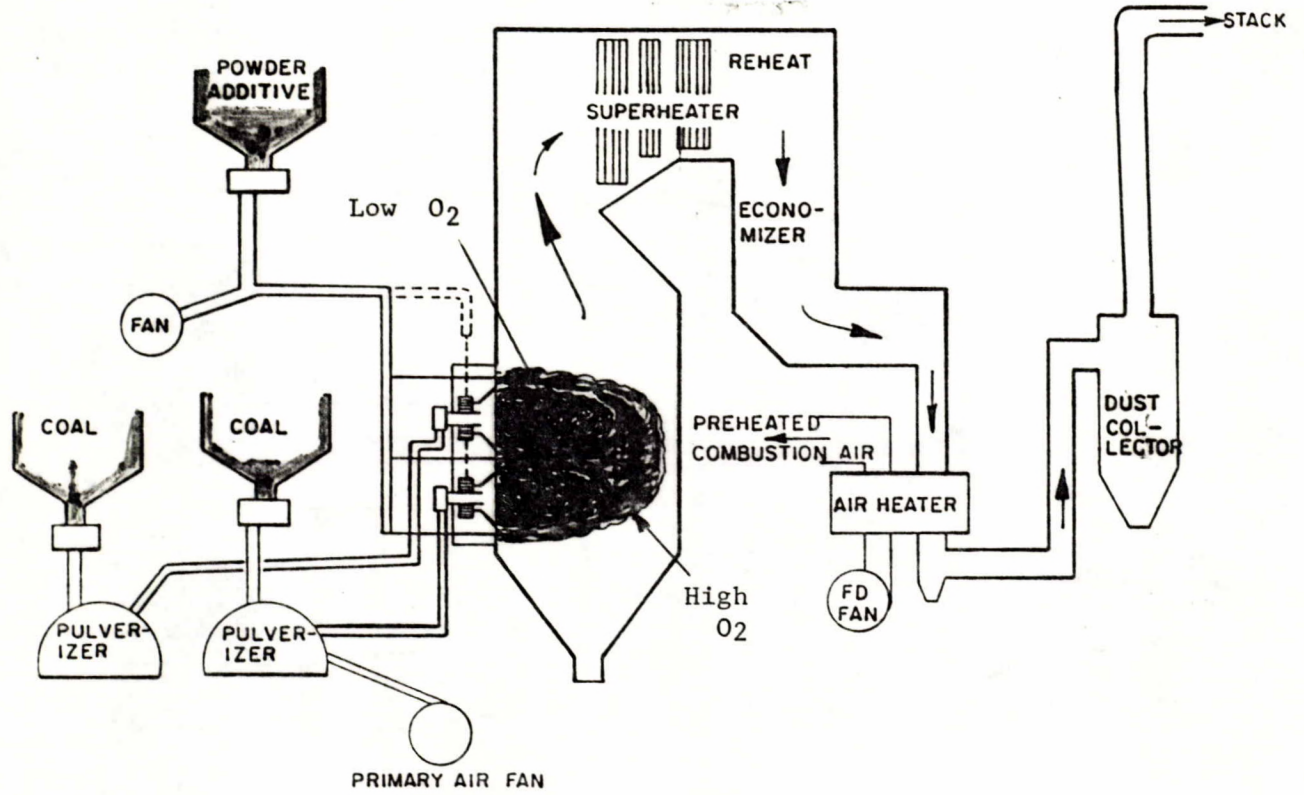


Fig. 16 - Illustrations of Gagetown boiler showing combined NO_x/SO_x control system

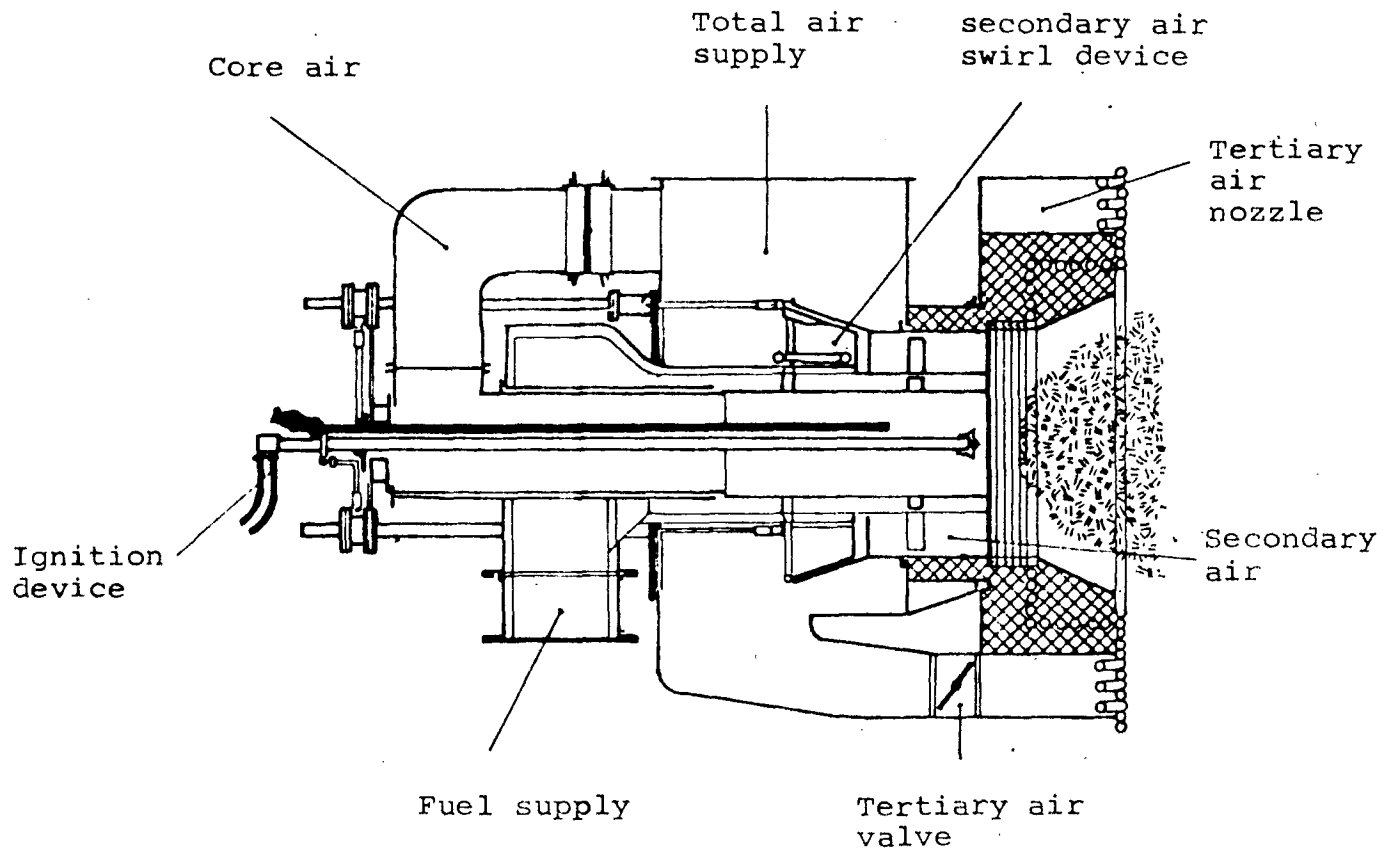


Fig. 17 Staged mixing burner (SM-burner)

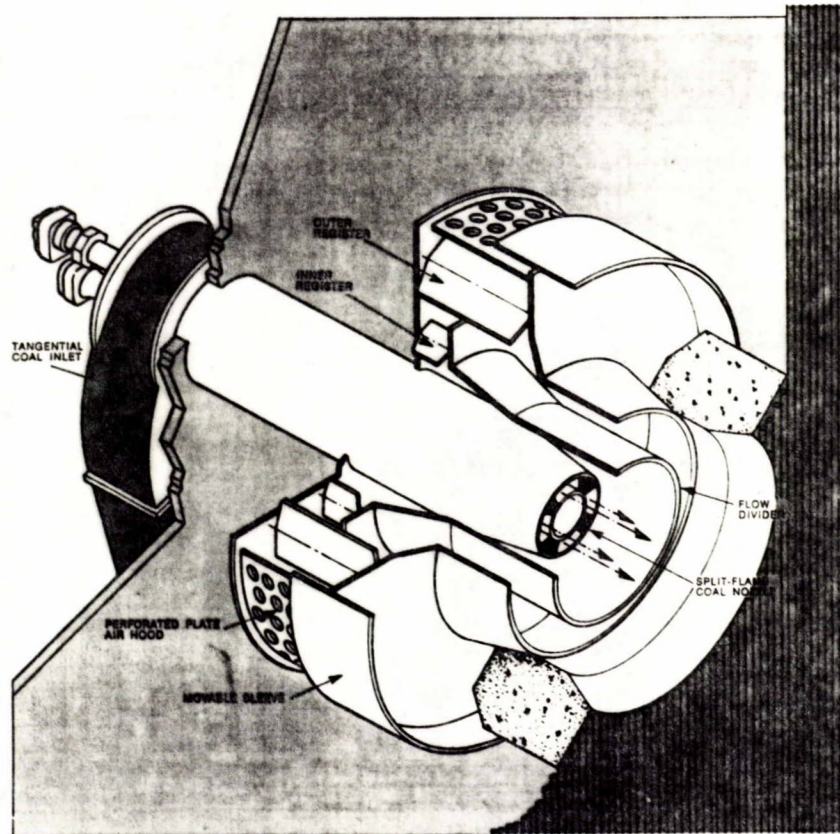


Fig. 18 - Controlled flow low NO_x coal burner
(split-flame type)