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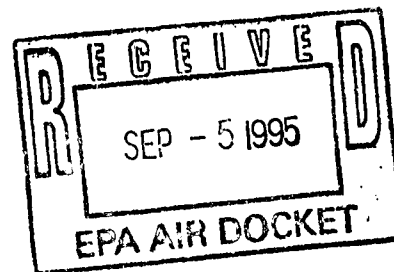
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NO_x Control in a Brown Coal-Fired Utility Boiler



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NOx Control in a Brown Coal-Fired Utility Boiler

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ABSTRACT

Rheinisch-Westfälisches Elektrizitätswerk A.G. (RWE) initiated a number of tests during 1987 of the NOxOUT^(R) Process for nitrogen oxides (NOx) reduction on a 150 MWe brown coal-fired boiler. The NOxOUT Process uses urea and enhancer chemicals for the reduction of NOx to molecular nitrogen, water vapor and carbon dioxide. The test objectives of up to 50% NOx reduction and ammonia (NH₃) slip of <5 ppm were met over a range of operating conditions.

In 1988, a commercial NOxOUT system was subsequently installed on a 75 MWe brown coal-fired boiler to comply with a controlled NOx level of 200 mg/Nm³ (approximately 100 ppm). NOx was first reduced to 300 mg/Nm³ (approximately 150 ppm) by the use of combustion modifications. Additional NOx reduction down to 180 - 195 mg/Nm³ (approximately 90 - 98 ppm) has been achieved using the NOxOUT Process. NH₃ slip has been controlled to a level of <2 ppm through the combination of enhancer chemicals plus selective injection.

INTRODUCTION

The Federal Republic of Germany is recognized as a world leader for the control of sulfur and nitrogen oxides emitted from stationary combustion sources. Acid rain arising from these pollutants is not only affecting human health but is also considered to be one of the major contributors to the damage of lakes and streams with their populations of aquatic life, forests, and to historical buildings (1, 2). Current legislation and local agreements between industry and the state authorities require that nitrogen oxides be reduced to a maximum of 200 mg/Nm³ (approximately 100 ppm) for all boilers in excess of 300 MWth by the end of this decade.

Rheinisch-Westfälisches Elektrizitätswerk A.G. (RWE) has a total generating capacity of more than 20,000 MWe and is the largest electric utility in the Federal Republic of Germany. One of the major fuels of RWE is brown coal which is burned in power stations of about 10,000 MWe installed capacity, with individual units sized up to 600 MWe.

The Rhine region brown coal differs distinctly from other fossil fuels such as bituminous coal or lignite. A high moisture level of 55% - 62% on a raw coal basis, a variable ash content of 2% - 20%, and a low nitrogen concentration of 0.3% - 0.4%, are typical for this fuel.

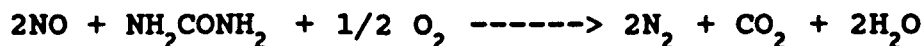
Consequently, brown coal combustion requires specifically designed fuel preparation circuits and boilers which, in conjunction with the fuel properties, result in specific flue gas characteristics. The flue gas contains about 20% water vapor resulting in maximum flame temperatures below 1200°C and fairly constant NOx concentrations well below 800 mg/Nm³ (approximately 400 ppm).

It is noteworthy that this fuel is mostly burned in tangentially fired boilers with jet burners in vertically arranged slits in the walls and/or corners of the combustion chamber. Normal operation with one burner slit always out of service at full load and more slits out of service at partial load differs from the firing mode of higher quality coals and results in asymmetric horizontal temperature distributions in the boiler, which has major consequences, as will be discussed later.

In order to comply with the NOx emission requirements, after having completed extensive large scale testing and economic evaluation of the selective catalytic reduction (SCR) technique (3), and also having reached very promising results with combustion modifications (4), RWE decided to retrofit 37 boilers with a total capacity of 9,300 MWe with alterations to the combustion chamber only. However, there were some indications that the final goal of a maximum NOx emission of 200 mg/Nm³ (approximately 100 ppm) may not be achievable for all boilers and under all operating conditions. Therefore, additional NOx removal principles would be required. This led to large scale tests of various options of injection systems.

One major option for RWE was the NOxOUT Process. The NOxOUT Process licensed through Fuel-Tech N.V., is a relatively low cost retrofit NOx control technology. The NOxOUT Process can be installed with a short downtime and is applicable to a very broad range of fuels and boilers. NOx reductions up to 80% are achievable with this technology (5). The NOxOUT Process can be installed along with combustion modifications to achieve even higher levels of reduction. Hence, the NOxOUT Process is applicable as a primary retrofit application and can be applied in conjunction with combustion modifications.

The NOxOUT Process is based upon the chemical reaction between nitrogen oxides and urea.



The reaction takes place at a temperature of about 925°C - 1,100°C. Research on the urea-based process was initiated in 1976 under sponsorship of the Electric Power Research Institute (6). Starting with the EPRI inventions (7, 8), Fuel Tech has made a number of contributions to improve the application of the technology (9, 11), including broadening and/or shifting of the optimum temperature for reaction by the use of Fuel Tech proprietary chemical enhancers (10, 12, 13, 14).

Fuel Tech patented enhancers have also been effective in the control of ammonia formation (11). Ammonia is a by-product of the reaction between NOx and urea that can be formed under certain conditions. Ammonia production is generally undesirable because of the possibility of forming ammonium sulfate and ammonium bisulfate in the presence of sulfur trioxide and in the contamination of ash, affecting ash disposal options. (Ammonium bisulfate has been known to cause fouling in the air preheater area.) The sulfur content of RWE coal (0.2% by weight) and the temperatures of the preheater area (<330°C) could provide a reactive environment for the formation of ammonium bisulfate if there is ammonia present.

TEST PROCEDURES

The introduction of the NOxOUT Process to brown coal fired boilers was carried out in various steps. The first preliminary tests had to show:

- at least 30% - 50% NOx reduction is possible with NH₃ emissions, or "slip", below 3.8 mg/Nm³ (approximately 5 ppm)
- the process is flexible enough to accommodate various combinations of burners in operation
- the process could cope with load variations down to partial loads of 60%
- the process is also effective under simulated flue gas conditions which were expected after the envisaged installation of combustion modifications.

These tests were carried out on a 150 MWe boiler (Unit D, RWE Weisweiler power station, Figure 1) during the summer of 1987.

Temporary equipment was used for the D boiler tests which consisted of plastic day tanks for chemical storage, a portable pumping skid, mixing headers for combination of NOxOUT-A solution, enhancers and water, rubber hoses, and distribution headers to split the flow between multiple wall injectors at each of three levels of injection. NOxOUT-A is an aqueous solution of urea and proprietary chemicals.

The first level of injection was into the upper furnace region at 25 meters. The second level was located immediately before the secondary superheater. The third level was located at the top of the backpass after the primary superheater. A number of different injectors were tried and the final arrangement consisted of sixteen external mix injectors at Level 1 (two per penetration), nine pin jet injectors at Level 2, and four internal mix injectors at Level 3. Steam was used for cooling and atomization for injectors at Level 1. Air was used for cooling and atomization at Levels 2 and 3 due to a limited availability of steam.

After the successful completion of the preliminary tests a full scale demonstration was conducted at the same power station on a 75 MWe boiler (Unit C2, Figure 2) during the Summer of 1988. Combustion modifications, installed during the Spring of 1988, reduced the NOx emissions from about 450 mg/Nm³ (approximately 225 ppm) to about 275 - 325 mg/Nm³ (approximately 140 - 165 ppm) at full load. The major goal of the Unit C2 demonstration was the successful performance of the NOxOUT Process for four weeks of continuous operation, during which the NOx emissions must not exceed the guaranteed level of 200 mg/Nm³ (approximately 100 ppm). In addition, the process was required to meet this emission limit while maintaining ammonia emissions below 5 ppm at all operating conditions.

Based on experience from the D boiler, NOxOUT injectors were installed at three levels; at 19 meters immediately above the burners but below the flue gas recycle and overfire air ports, at 25 meters immediately before the secondary superheater, and at 36 meters at the top of the backpass after the primary superheater. There were twelve penetrations at Level 1, twelve at Level 2, and four at Level 3. External mix injectors were used at all levels. Steam was used as the cooling and atomizing fluid for all injectors.

TEST RESULTS - D BOILER

A. Temperature and Composition Profiles

Prior to the commencement of NOxOUT injection, temperatures were measured at each of the three levels of injection. Temperature measurements were made using water-cooled suction pyrometers. Analyses for O₂, CO and NOx were carried out at the same time as the temperature measurements. Typical temperature measurements for 100% load were found to be 1021°C (Level 1), 825°C (Level 2), and 580°C (Level 3).

As expected, in the upper furnace region temperatures near the wall tended to be lower than temperatures towards the center of the boiler. However, the overall temperature profiles tended to be fairly uniform. The temperatures at Levels 1 and 2 were within the temperature window for enhanced urea chemicals. The temperature at Level 3 however, was too cold for injection of chemicals available at RWE.

Gas composition profiles for NOx, CO and O₂ were recorded at the three levels of injections. The NOx, CO and O₂ concentrations differed at the three levels. NOx values were highest at Level 2. Higher NOx concentrations recorded at Level 2 were associated with higher O₂ concentrations at this level. The CO levels were found to be significantly higher at Level 1 than other levels. The higher concentration of CO at Level 1 indicates that complete burnout has not occurred until after this point.

B. NOx Reduction, Level 1 Injection

Results obtained from injection of mixtures of NOxOUT-A and enhancer at Level 1 in the D boiler are shown in Figure 3. The Normalized Stoichiometric Ratio (NSR) is defined as the ratio of NH₃ species injected divided by the initial moles of NOx. Thus one mole of urea per mole of NOx represents an NSR of 2.0. The initial NOx level for this series of experiments was approximately 550 mg/Nm³ (6% O₂). The results show an essentially linear response to increasing amounts of urea injection.

The effect of enhancer is to shift the temperature window for NOx reduction activity to a lower temperature (5). Thus at a specific operating temperature, the addition of enhancer could result in either increased NOx reduction, decreased NOx reduction or no change in NOx reduction. A decrease in NOx reduction implies that the reaction temperature (in this case approximately 1020°C) is such that the operation is on the high side of the reaction temperature window. There is a benefit in operating on the high side of the reaction temperature window in that by-product NH₃ formation is suppressed while NOx reduction is only slightly diminished (11).

The amount of NOx reduction achieved in these experiments implies a urea utilization in the range of 27% with the low enhancer ratio, shifting to 20% for the high enhancer ratio (assuming that each urea molecule is theoretically capable of reducing two molecules of NOx). As Figure 3 indicates, the response of utilization to increasing NSR is essentially flat over the range of NSR investigated. Other studies have shown that utilization will tend to decrease with increasing NSR (5).

C. Enhancer/Urea Ratio, Control of NH₃ Slip

The enhancer/urea (E/U) ratio is one of the most significant process variables in the NOxOUT Process. As previously noted, the enhancer/urea ratio can affect the level of NOx reduction but it is particularly important in the control of NH₃ slip. The amount of NH₃ produced is a function of various process variables and tends to increase with:

- decreasing temperature
- increasing the reagent-to-NOx mole ratio (increasing NOx reduction)
- decreasing enhancer/urea ratio.

The amount of NH₃ formed tends to increase with increases in NOx reduction and therefore a trade off occurs when NOx reduction has to be forfeited to maintain low NH₃ levels.

Figure 4 shows a comparison of ammonia slip versus NOx reduction at two different E/U ratios. At the higher E/U ratio less ammonia was generated per increment of NOx reduction. The higher E/U ratio enables NOx reduction to be increased from about 30% to about 50% at 5 ppm of NH₃. NOx reduction can be further increased to about 65% with an NH₃ slip of less than 15 ppm.

D. NOx Reduction, Two Level Injection

Urea and enhancer injection was performed simultaneously at Levels 1 and 2 to determine if multiple level injection would significantly (A) increase chemical utilization, (B) reduce NH₃ slip, and (C) increase the NOx reduction.

Results from injection of urea and enhancer at Level 1 versus Levels 1 and 2 at comparable test conditions are contained in Table 1.

TABLE 1
SINGLE VS. TWO LEVEL INJECTION

Experiment	A	B	C	D	E	F
No. of Levels	1	2	1	2	1	2
NSR (1)	1.2	1.0	2.7	2.6	3.0	3.3
E/U (2)	0.1	0.15	0.2	0.56	0.03	0.2
% Reduction	31.4	44.6	51.0	64.0	44.0	50.5
% Utilization	26.0	45.0	19.0	24.0	14.5	15.5
NH ₃ , ppm	13.0	14.0	20.5	--	--	12.9

(1) NSR=ratio of NH_i moles injected divided by the initial moles of NO_x.

(2) Weight ratio of Enhancer to Urea.

Generally, it can be concluded that urea utilization increases when urea is split between two levels at the same overall NSR. These results were achieved despite a slightly higher overall E/U ratio with two-level injection. Although it has previously been found that a higher E/U ratio leads to lower NH₃ slip, NH₃ slip was as high with two level injection as with one. It is believed that NH₃ slip from Level 2 was the result of chemical impingement on the superheater platens which were within five feet of the point of injection at Level 2. Efforts were made to inject the chemicals between the platens, but it was virtually impossible to completely eliminate direct chemical impingement.

E. Burners Out of Service

During the demonstration, extensive data collection was undertaken to determine if differences in NO_xOUT Process performance could be attributed to a specific burner slit that was out of service. Temperature measurements were taken, as previously described, prior to injection with different configurations of burner slits in service. Only slight variations in daily average temperatures indicated that there was no significant temperature difference with any specific burner slit out of service.

In addition, process results obtained with the four different burner slits out of service indicate no significant difference in process performance. The amount of urea required to achieve a given level of NO_x reduction and the level of NH₃ slip were essentially unchanged.

F. Low Excess Air and Low Load

Two specific objectives of the demonstration were to determine the flexibility of the NOxOUT Process to respond to reduced load and to low excess air (simulated combustion modification).

Temperature profiles and gas composition analyses obtained at Level 1 revealed significant differences for the modified conditions (Table 2).

TABLE 2
EFFECT OF BURNERS OUT OF SERVICE
ON TEMPERATURE AND GAS COMPOSITION

<u>Operation</u>	<u>Conventional Excess Air 100% Load</u>	<u>Low Excess Air 100% Load</u>	<u>Conventional Excess Air 60% Load</u>
Level 1			
Avg. Temp, °C	1,008	1,065	954
Avg. NOx mg/Nm ³	460	321	362
Avg. CO, ppm	47	700	38
Avg. O ₂ , vol. %	5.2	2.2	5.7
Level 2			
Avg. Temp, °C	811	878	804
Avg. NOx mg/Nm ³	513	328	347
Avg. CO, ppm	11	330	0
Avg. O ₂ , vol. %	6.3	3.1	5.5
Stack			
NOx, mg/Nm ³ (1)	584	403	427
CO, ppm	6	3	0
O ₂ , vol. %	7.1	4.5	7.6
(1) Corrected to 6% O ₂ .			

Low excess air resulted in a higher temperature and a lower NOx content at Level 1. Oxygen content was significantly lower and carbon monoxide (CO) was higher. When load was reduced to 60% the temperature and NOx content dropped. Substantial variation in CO content was observed from point to point at Level 1 during the experiment with low excess air. These localized concentration variations of CO could be the result of slow burnout of char. Process results obtained with low excess air and low load are shown in Table 3 and compared with data obtained at normal 100% load conditions.

TABLE 3
EFFECT OF MODIFIED OPERATION ON NOXOUT PROCESS

<u>Operation</u>	<u>100% Load</u>	<u>Low Excess Air</u> <u>100% Load</u>	<u>60% Load</u>
Experiment	G	H	I
Baseline NOx, mg/Nm³	580	403	427
Level 1			
NSR	2.4	3.0	3.0
E/U	0.14	0.03	0.5
Level 2			
EWR (1)	0.33	0.13	0.42
% Reduction	49	45	48
% Utilization	20	16	18
NH ₃ , ppm	4.5	3.5	5.0

(1) Weight ratio of enhancer to baseline NOx (as NO₂).

During the experiments at higher NSR, enhancer alone was injected at Level 2 to provide an additional level of control over NH₃ slip.

Experiments at low excess air were initially carried out (Exp. H) at the same chemical flow rates as had previously been determined to be the optimum for 100% load conditions (Exp. G). The NSR increased from 2.4 - 3.0 because of the lower baseline level of NOx and reduction declined from 49% - 45%.

At 60% load conditions (Exp. I) the temperature at Level 1 is lower than at 100% load (954°C vs. 1,021°C). At these conditions a three-fold increase in enhancer rate was required to maintain the NH₃ slip at 5 ppm. The fact that an NSR of 3.0 was required to achieve comparable NOx reductions for both low load and low excess air conditions is most likely due to the lower initial NOx concentration in the flue gas.

Overall, these data clearly demonstrate that the amount of enhancer required to maintain NH₃ slip below a target level depends upon the temperature at the point of injection. The average temperature at Level 1 varied over the range of 954° - 1,065°C. Within this temperature range, the amount of enhancer required to meet NH₃

limits increased from 1 part enhancer to 30 parts urea at 1,065°C up to 1 part enhancer to 2 parts urea at 954°C.

TEST RESULTS - C2 BOILER

A. Start-up Tests

Prior to the start-up of NOxOUT injection and soon after the C2 boiler returned to stable conditions with the newly installed combustion modifications, Fuel Tech measured baseline temperature, NOx, O₂, and CO. These measurements were carried out in a grid at each level of injection.

Temperature and gas composition data gathered at 80% and 100% load are shown in Table 4.

TABLE 4 C2 BOILER - TEMPERATURE AND GAS COMPOSITION				
Load Level	100%		80%	
	1 (19 m)	2 (25 m)	1 (19 m)	2 (25 m)
Temperature, °C	1040	995	992	980
NOx, mg/Nm ³	250-400	250-350	140-400	150-400
CO, ppm	500-10000	70-3000	1000-3000	250-1000
O ₂ , %	3.2	4.1	1.2	4.2

The overfire air which diverted a part of the combustion air to the ports above the 19 meter level had the effect of extending the primary combustion zone. The first injection level was within this extended combustion zone. As a result of these conditions, the values for both NOx and CO were extremely erratic and averages were considered meaningless.

Although temperature levels at 19 meters would appear to be conducive to good NOxOUT reagent utilization, the high CO levels existing at Level 1 have the impact of shifting the temperature window in which urea is effective (15). Early injection tests at Level 1 were not productive for NOx reduction. It was apparent that the "effective" temperature at these high CO conditions was too high for reasonable urea utilization. Further testing was based on injection at the second level (25 meter). The third level of injection (36 meter) was not required because the total amount of NOx reduction required to achieve <200 mg/Nm³ proved to be only about 35%, which could be readily achieved at Level 2.

B. Twenty-Five Meter Level Injection

NOx reductions in the range of 40% - 50% were repeatedly achieved at relatively low NSR values from baseline NOx levels of 275 - 325 mg/Nm³. Figure 5 shows the reductions and utilizations observed at varying NSR values. Although the majority of the tests were performed with straight injectors, some comparative tests were performed using angled injectors. NOx reduction and utilization were not sensitive to different types of injectors.

These tests were carried out at an E/U ratio in the range of 0.3 to 0.6. This range was based on results previously determined to be optimum in the D boiler tests. Specific tests to optimize the quantity of enhancer for the C2 boiler remain to be undertaken.

Tests at 80% load with each one of the four separate burner slits out of service showed that the process results were independent of the specific burner slit that was out of service. Baseline NOx levels at 80% load were typically in the range of 250 - 275 mg/Nm³ compared to 275 - 325 mg/Nm³ at 100% load. The NSR required to achieve <200 mg/Nm³ was about 1.5 and reagent utilization is lower than that achieved at 100% load. The reason for this is postulated to be the higher CO concentration at 80% load. As previously noted, CO shifts the temperature window to lower temperatures, and at the upper end of the window this results in decreased reagent utilization.

C. Ammonia Slip

The Unit C2 contract between RWE and Fuel Tech specified that after the initial optimization portion of the demonstration, the "recipe" for achieving the necessary NOx reductions over a range of operating conditions would be turned over to RWE to test its ability to operate continuously for four weeks. Ammonia slip was a very important consideration for RWE. During the four week acceptance period, 64 analyses of ammonia were undertaken to determine compliance with the 5 ppm limit. Fuel Tech performed considerable ammonia sampling and computer modelling to gather information on the potential for ammonia formation.

Samples of flue gas were withdrawn from four locations in the flue gas ducts upstream of the air heater. The flue gas temperature at this point is approximately 325°C. The baseline level of NH₃ was determined before the start of injection to be 0.7 ppm. There was no stratification of NH₃ in the flue gas. Following the start of injection at the second level (25 meters) it was found that the overall specification of <5 ppm of NH₃ could be maintained at both 100% and 80% load while also maintaining NOx at <200 mg/Nm³.

A computer model of the C2 boiler was utilized in an effort to get a better interpretation of the flow and temperature regimes affecting ammonia formation within the boiler at the second level of injection. Figure 6 shows a computer generated temperature profile at 25 meters and the location of the 12 injectors. The model showed that velocity and temperature are influenced by the boiler throat which starts right above the 25 meter level. This throat shifts the swirl in the flue gas which is characteristic of a tangentially fired boiler. This shift, combined with the location of the overfire air ports, results in cooler areas in the front right and back left corners away from the core of the swirl. This suggested that injection into these cooler regions might be leading to NH_3 slip.

Elimination of five of the twelve injectors at Level 2 combined with an enhancer/urea ratio of about 0.6 led to the extremely low levels of NH_3 slip. A total of 64 individual analyses for NH_3 were carried out over a one month period; 24 at 100% load and 40 at 80% load. The overall average of 1.5 ppm is less than 1 ppm above baseline. The overall amount of slip is well below the target of 5 ppm.

CONCLUDING REMARKS

The results of the above described projects at the two brown coal fired boilers can be summarized as follows.

- A NO_x reduction of 40% - 50% can be achieved with less than 5 ppm of NH_3 slip. Higher NO_x reductions are possible depending on the ability to achieve a good distribution of the chemicals and/or on the availability of multiple levels for injection.
- In particular, with baseline NO_x emissions of 300 mg/Nm^3 (approximately 150 ppm) or less, which was established by the application of combustion modifications, the intended emission value of less than 200 mg/Nm^3 (approximately 100 ppm) can be reached with NH_3 slip well below 5 ppm.
- The use of enhancer is effective in the control of NH_3 slip over various boiler loads especially when load changes result in temperature shifts within the boiler.
- The process is effective at various loads and operating conditions, without having to change injectors.
- Reagent utilization increases with multiple-level injection of urea where more than one level is available at appropriate temperatures.

- The use of enhancer permits the chemical utilization to be optimized for a specific reaction temperature.
- The process can be easily retrofitted on an existing boiler with only small disruption to on-going operations.
- RWE's performance objectives for NOx reduction were met by the NOxOUT Process. Further optimization of operating costs can be achieved as the technology evolves by optimizing chemical treatment and injection parameters for specific boiler applications.

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Fig.1. R.W.E. D BOILER

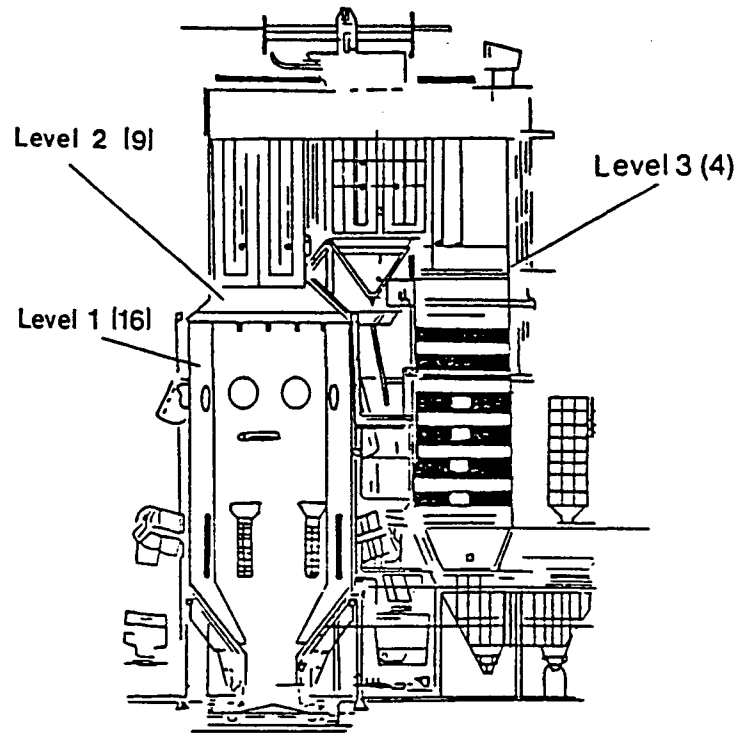


Fig. 2. R.W.E. C2 BOILER

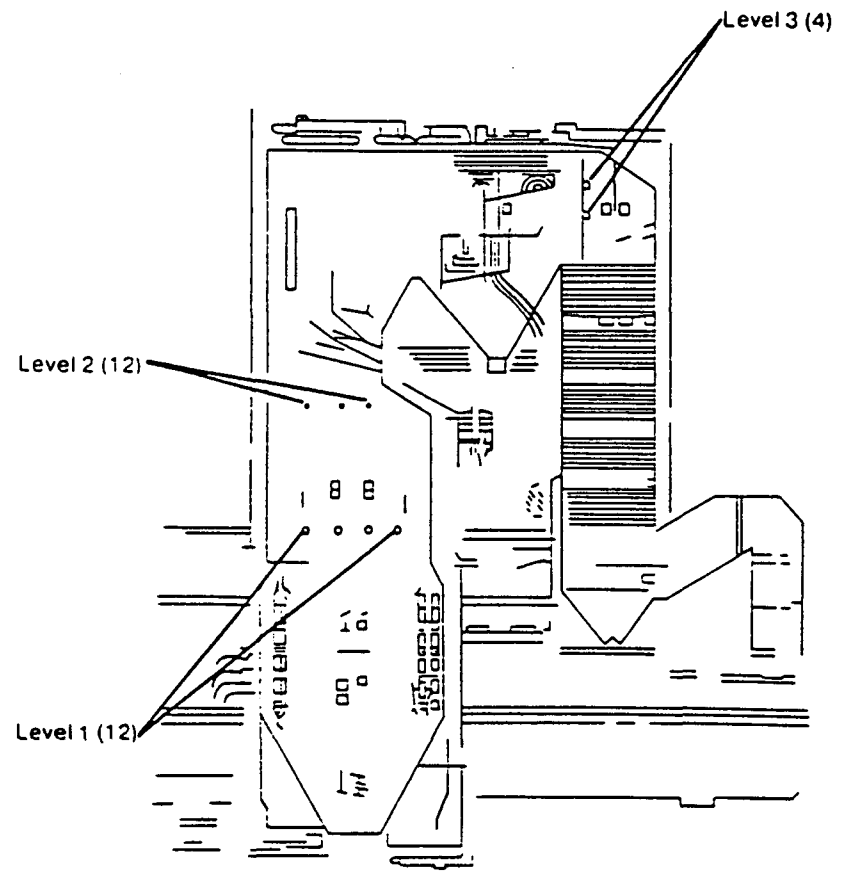


Fig. 3 **NO_x REDUCTION AND UTILIZATION vs. NSR
R.W.E. UNIT D**

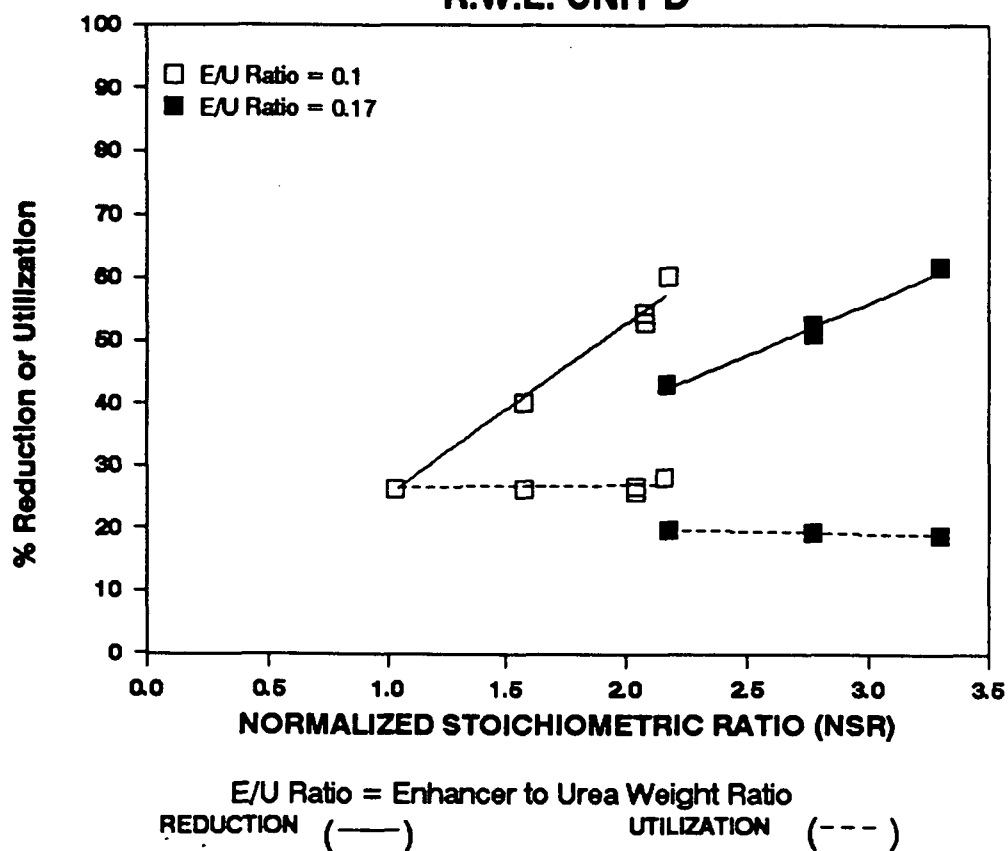


Fig. 4. **AMMONIA vs. PERCENT NO_x REDUCTION
R.W.E. - UNIT D**

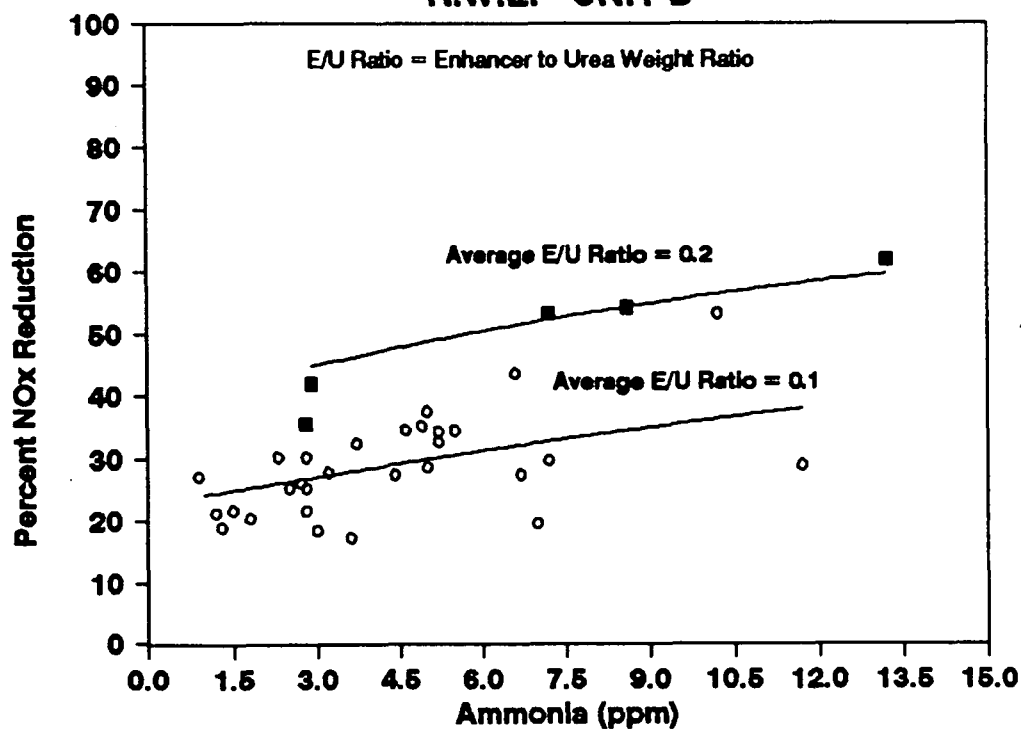


Fig. 5 **NO_x REDUCTION AND UTILIZATION vs. NSR**
R.W.E. - UNIT C2

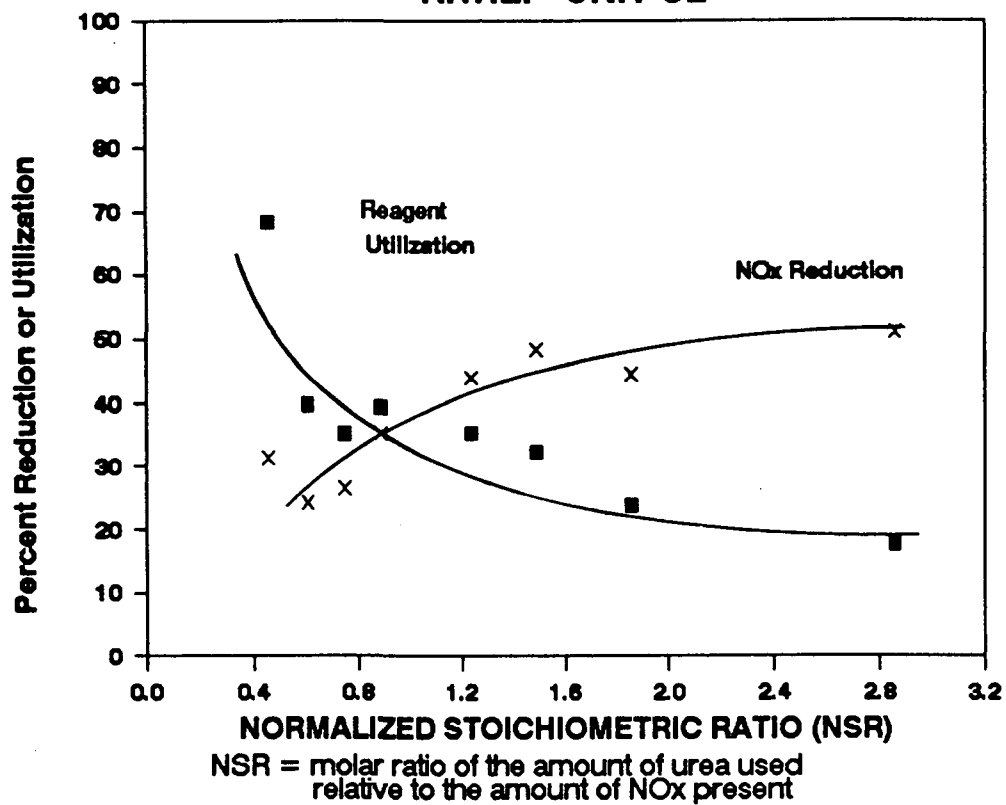


Fig. 6. **FURNACE MODELLING - TEMPERATURE PROFILE**
R.W.E. C2 BOILER - 25m LEVEL

