Nitrogen Oxide Abatement by Distributed Fuel Addition

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ABSTRACT

A screening study was performed on a laboratory scale downfired combustor to determine the effect of various variables on the effectiveness of the reburning process as a technique for NO_x abatement. The objective was to define optimum conditions under which reburning can be used and to be able to compare the reburning performance of our combustor to that reported by others. For this purpose, a statistically designed parametric investigation was conducted to determine how a set of controlled variables (primary and secondary stoichiometric ratios, location of the reburn zone and primary fuel load) would affect the reduction in NO emissions in a classical reburning configuration. Also, the effects of other variables (NO in the primary zone, temperatures in the primary, reburn and burnout zones and the residence time in the reburn zone) were also investigated.

No optimum configuration was identified in this study. Nevertheless, this study provides insight into the parameters associated with reburning.

STATISTICAL DESIGN OF EXPERIMENTS

Preliminary screening results reported in previous quarterly reports have indicated that reburning effectiveness is a function of a large number of experimental variables. In order to compare our data to those of others, it is necessary to examine the effects of each variable seperately. An efficient way to do this is to employ a statistically correct design of experiments, as described below.

Statiscical methods can be used to design experiments in which various factors are varied over an experimental region of interest. The effects of these factors and their interactions can be predicted in this region. The response can be expressed as a continuous function in terms of these factors. A factor or a controlled variable is a variable which can be changed from one level to another without a change in any other factor. A response is any measured property corresponding to a combination of levels of the controlled variables affecting that property.

One of the methods used in the statistical design of experiments is referred to as Response Surface Experimentation (1,2,4,5,7,8). This method allows the determination of an empirical relationship between a response and the controlled parameters in the experimental region based on an experiment involving a minimal number of trials. It empoloys several topics in Mathematics and Statistics, such as the theory of multiple regression and some features of Factorial Design. After a response is related to the controlled variables, the optimum conditions can be easily determined in the experimental region. If the desired optimum is outside the covered experimental region, a simple first order model relating the response to the controlled parameters in this region is derived. Then, the method of steepest ascent (maximum response) or descent (minimum response) can be used to determine the direction along which the next set of experiments should be performed (2).

The following functional relationship is proposed between a response y and factors x_1, x_2, ... , x_k affecting that response:

$$y = \sum_{i=0}^{k} b_i x_i + \sum_{i=1}^{k} \sum_{j=1}^{k} b_{ij} x_i x_j$$

x_o is a dummy variable which is always equal to unity and k is the number of the controlled variables. The coefficients, b_i and b_{ij}, can be estimated using the theory of multiple regression and the principle of least squares. In least squares model fitting, O, the sum of the squares of the difference between the measured response y_m and the response predicted by the model y_p is minimized:

$$\frac{\partial O}{\partial b_{i \text{ or } ij}} = 0$$ giving same number of equations and unknown coefficents which can be solved simultaneously

It is assumed that effects of order higher than the second can be ignored. Now consider a factorial arrangement of treatments. A factorial experiment is one in which all the levels of a given factor are combined with all the levels of every other factor in the experiment (2,4). The result is a combination of all possible treatment levels. There are some advantages to factorial experiments:

1. A factorial experimental design is more efficent because it requires fewer experiments than methods in which the factors are varied individually one

at a time.

2. All observations can be used in evaluating all effects without any necessary repititions.

3. The experiments cover the whole experimental region of interest.

The simplest form of factorial design is 2^f factorial corresponding to f factors and 2^f pssible treatment combinations. Each controlled variable has two possible levels throughout the experiment, a high level or a low level. These two levels can be the two extreme levels of the variable. On the other hand, a 3^f factorial would have three possible levels which can be labeled as low, high and intermediate. An example of a 2^3 factorial design is shown in Table 1. The low and high settings of each variable are denoted by -1 and +1 respectively and a test matix is formulated.

In this table, the columns under x_1, x_2, x_3 represent chosen values (maximum by 1 and minimum by -1) of the independent experimentally adjustable variables, while the other columns represent values of interaction and second order terms arising from that choice. First order variables and their interactions have the properties:

$$\sum_{m=1}^{N} x_i x_j = 0 \quad \text{for } i,j = 1, 2, \ldots, k \quad \text{and} \quad i = j$$

where k is number of variables and N is number of observations.
This is a property of orthogonality. Also,

$$\sum_{m=1}^{N} x_i = 0$$

This is not the case for second order variables of the form x_i^2.
In general, orthogonality of variables can be obtained if the responses are measured at equally spaced increments of the controlled variable. The property of orthogonality has some advantages:

1. The sign and magnitude of the predicted coefficients of first order effects and their interactions would be independent of each other and thus, can be estimated independently. The Least Squares estimates of the coefficients would be orthogonal linear functions of the observations.

2. Second order effects may be added to the fit equation and would be independent of the effects already included in the model.

As seen in Table 1, the vectors of second order parameters, x_i^2, cannot be distinguished from x_o. Consequently, it is impossible to obtain estimates of the second order coefficients, b_{ii}, from factorial experiments. In other words, b_o estimate would be biased because it would include estimates of the second order coefficients. Therefore, additional experiments would be required and should be selected in such a way to preserve some of the orthogonality features of the experimental design.

If the true relationship between the response and the variables is not as predicted by the model, the difference would be due to experimental error and/or inadequate choice of a model. The experimental errors are independent of each other. They are normally distributed with a constant variance and are independent of the model used. The error variance or error mean square can be estimated from replicate observations. The regression equation should account for a large and significant part of the variation to be a useful representation of the data. The variance due to each estimated element in the model can be evaluated and compared to the error variance and the contribution of that element to the prediction of the response can be judged. If the variability about the fitted model is larger than what would be expected from the errors of measurements alone, the model would be inadequate for representing the data and it should be modified.

The formulas for computing the analysis of variance are given in Table 2 (5).

The F ratio is a test of two independent variances. It gives the probability of the variances of two normal distributions being equal. The probabilities due to F distribution are tabulated. For example, the F ratio of the mean squares due to lack of fit to that of the error of observation would give the probability of both variances being equal. Thus, this ratio would give an indication of how the variation about the model compares with the variation due to errors of observation.

One complete experimental design is known as a Central Composite Design. An example is shown in Table 3:

The factorial experiments are used to estimate the coefficients of the first order effects and their interactions. The star design experiments are used to estimate the coefficients of the second order effects. An appropriate choice of "a" is $a=2^{k/4}$ where k is the number of variables(5). That would produce a rotatable design which gives equal predictive power in all directions at a constant distance from the center of the design. Repeated experiments at the center of the design would give an estimate of the error variance that is independent of the model.

The advantage of Composite design is that it allows the work to proceed in stages. The first order model including interaction terms can first be completed. If the first order effects are fairly small and the interaction effects are large, it may be necessary to determine all the second order effects and additional points can be added to complete the design.

The derived model is a representation of the response in terms of the controlled variables. A graphical representation of the response surface can be obtained by drawing lines of equal response on a graph whose coordinates denote the levels of the factors.

To summarize, the derived model would be an empirical model and should be utilized only in that sense. It can be used for interpolation only in the experimental region that is covered in the design and should not be used for extrapolation. The model can be quite useful in locating the optimum conditions in the investigated region.

A complete second order model should be adequate for representing responses if all the relevent factors are included. If such a model proved to be inadequate in the representation of the data, a second look should be given to the data and the factors involved.

SCREENING STUDY FOR CLASSICAL REBURNING

The objective of this study is to identify the significant factors that affect the efficiency of the reburning process with the goal of developing predictive methods that correlate reburning effectiveness with the operating parameters. The independent variables that were selected for this study are: the stoichiometric ratio in the primary zone and that in the reburn zone, the location of the reburn zone and the primary zone fuel load. Response surface experimentation was used in this parametric study. Table 4 shows the experimental limits of the examined variables and the coding equations.

The low and high limits were determined by experimental limitations of the existing experimental setup. The setup was described in an earlier report (10). For example, since some existing utility ports are not equally spaced, an intermediate setting of 0 was not possible. Denoting low, intermediate and high settings of the four variables by -1, 0 and 1 respectively, the test matrix shown in Table 5 was formulated.

The first 16 tests (factorial) would be sufficient to compute linear relationships relating the response to the parameters. The additional 14 tests (star) would allow a full quadratic model (except for the coefficient of x_1^2) to be formulated as well as providing additional tests for computing the main effects and two factor interactions.

Coal was burned at the desired feed rate and the inlet air was adjusted to obtain the desired SR_1. After all the instuments indicated a steady state baseline condition (stable temperatures and stable exhaust concentrations), reburn fuel (methane gas) was introduced at the desired location to reduce the stoichiometric ratio to the desired level (SR_2). Additional air was introduced downstream of the reburn zone to complete the combustion process. The final stoichiometric ratio (SR_3) was maintained at 1.1 for all the tests. Measurements were taken after all the instuments indicated steady state operation. A total of 44 tests were examined including 10 replicates. The results of these tests are shown in Table 6:

A quadratic response surface model was fitted to the data using SPSS multiple regression procedure. The quadratic model included linear and quadratic dependences on each variable as well as interaction terms between pairs of variables. Table 7 shows the final step in the regression analysis using STEPWISE method.

The analysis yielded the following model relating the response to the controlled variables:

$$y=53.6-13.3*SR_2+9.28*X_1-6.19*SR_2*X_4-2.32*X_4+2.67*SR_2*SR_1$$

where y is the desired response expressed as the percentage of NO reduction due to reburning. It is calculated from the following equation:

$$y=100-100*(NO_{ex}/NO_p)$$

where

NO_{ex}=ppmv NO in exhaust (corrected)

NO_p =ppmv NO (corrected) in the primary zone, measured at port 3 (before reburn fuel was introduced)

Both NO measurements were measured on a dry basis and corrected to molar flue gas rates for coal burned at SR = 1, i.e.

ppm NO (corrected) = ppm NO (measured) * Dilution Correction Factor

where

Dilution Correction Factor= (actual moles/h flue gas at measuring point/ moles/h flue gas for coal only burned at SR=1.0)

Thus, all dilutuion effects were eliminated.

The model allowed for 91% of the variation among the 44 data points to be accounted for by the variation of the controlled variables. Only terms significant at the 2% level were included. The quadratic terms had no statistical significance in the experimental range that was covered. The results can be best interpreted through response surface plots as shown in Figure 1.

The dependence of the response on the coal feed rate diminished in the vicinity of SR_2 of 0.81 regardless of the location of the reburn zone. Also, at that point the response appears to have little dependence on SR_1 with values of about 68%±1 and 49%±1 for reburn fuel injection at ports 3 and 5 respectively. Lower coal feed rates would be more desirable at $SR_2 > 0.81$ and less desirable at SR2 < 0.81. As expected, better results were obtained when the reburn fuel was introduced at port 3 (x_1=1) which corresponded to higher reburn zone residence times and hotter reburn zone temperatures, as compared to port 5 injection (x_1=-1). The analysis showed that the linear term in SR1 was insignificant but it is important through its interaction with SR2. Overall, SR_1 has little effect on the reduction in NO. This is in agreement with the results of Greene et al.(3). However, as suggested in that study, it would be more desirable to operate the primary zone under low excess air level to reduce the amount of reburning fuel required to reach the desired level of reburn zone stoichiometric ratio.

The reduction in NO due to reburning increased as SR2 decreased and no optimum was observed. An optimum SR2 in the vicinity of 0.9 was identified by several researchers(3,6,9). This difference in observation may be explained by the high levels of NO that were observed in this study (950 - 1210 ppm) which may have resulted in continuous reduction in NO as more reburn fuel was added. Reductions, as high as 82% in NO were observed.

Other derived variables were measured along with the desired response. These variables are the NO cocentration in the primary zone (NOp), the peak temperatures in the primary zone (Tp), the reburn zone (Tr) and the burnout zone (Tb), and the reburn zone residence time (RTr). The effects of these variables on the desired response were analysed qualitatively using SPSS multiple regression procedure. Tables 8, 9 and 10 show the last steps in these analysis, corresponding to SR_2 of 0.73, 0.855 and 0.98 respectively. Tp and NOp did not have any statistical significance and will be assumed to have little effect on reburning effectiveness. This conclusion regarding NOp applies only in the range of values that was observed (950-1210 ppm). As expected, Tr and RTr contributions were very significant. More reductions in NO could be achieved at higher residence times and the contribution of RTr increased with SR_2. The analysis show that hotter reburn zone temperatures would improve the reburning effectiveness. However, cooler temperatures may be more desirable if the reburn zone is close to being fuel lean (as seen in Table 10 where the coefficient of Tr is negative). Furthermore, residence time and temperature interaction in the reburn zone appear to be significant especially as SR_2 decreases. The contribution of Tb seems to be significant only in the upper range of SR_2 and will be assume trivial under practical reburning conditions.

CONCLUSIONS

A screening study for classical reburning was performed and the effects of various variables that are associated with reburning were analysed. A predictive model correlating reburning effectiveness with the controlled variables was derived. Although no clear optimum was identified, reburning can be performed under conditions that would minimize the effects of some parameters and thus allow greater control of the reburning process. Reburning at a secondary stoichiometric ratio of 0.81 would minimize the contributions of the primary zone stoichiometric ratio and the coal feed rate.

FURTHER WORK

The next step in this study would be to investigate the effect of multiple distributed fuel addition on the reduction of NO by reburning. It is expected that by distributing the reburn fuel down the combustor, further reduction in NO can be achieved by slowing down the consumption of the reburn fuel and the generation of free radicals that cause the destruction of the nitrogenous species.

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Table 1.
EXAMPLE OF 2^3 FACTORIAL DESIGN

	first order terms				interaction terms			second order terms		
trial	x_o	x_1	x_2	x_3	x_1x_2	x_1x_3	x_2x_3	x_1^2	x_2^2	x_3^2
1	1	-1	-1	-1	1	1	1	1	1	1
2	1	1	-1	-1	-1	-1	1	1	1	1
3	1	-1	1	-1	-1	1	-1	1	1	1
4	1	1	1	-1	1	-1	-1	1	1	1
5	1	-1	-1	1	1	-1	-1	1	1	1
6	1	1	-1	1	-1	1	-1	1	1	1
7	1	-1	1	1	-1	-1	1	1	1	1
8	1	1	1	1	1	1	1	1	1	1

Table 2.
ANALYSIS OF VARIANCE

TERM	SUM OF SQUARES SS	DEGREES OF FREEDOM, df	NOTE
(1) response, y	sum(y^2)	N	N=# observations
(2) coefficient, b b=b_i, b_{ii} or b_{ij}	b*sum(u*y)	1	u=effect x_i, $x_i x_j$ or x_i^2
(3) residual	difference (1)-sum(2)	N-sum(2)	k=# factors
(4) error	y^2-[sum(y)]^2/n	n-1	n replicates of 1 experiment
	sum(d^2)/2	n	2 replicates of n experiments d=[y_{test1}-y_{test2}]
(5) lack of fit	difference (3)-(4)	difference (3)-(4)	

The mean squares, MS=SS/df.

TABLE 3.

COORDINATES OF A CENTRAL COMPOSITE DESIGN, k=3

	x_1	x_2	x_3
	--	--	--
(1) 2^3 facorial	-1	-1	-1
	1	-1	-1
	-1	1	-1
	1	1	-1
	-1	-1	1
	1	-1	1
	-1	1	1
	1	1	1
(2) star design	-a	0	0
	a	0	0
	0	-a	0
	0	a	0
	0	0	-a
	0	0	a
(3) center points (replicates)	0	0	0
	.	.	.
	.	.	.
	0	0	0

TABLE 4
CONTROLLED VARIABLES

Variable
1. location of reburn fuel injection, distance from burner in cm
2. primary zone stoichiometric ratio
3. reburn zone stoichiometric ratio
4. primary fuel load (Utah Bituminous #2 coal), lb/hr

Variable	Code	Low Limit	High Limit	Coding Equation
1	x_1	99.1 (port 5)	53.4 (port 3)	$x_1=(var-76.25)/-22.85$
2	SR_1	1.1	1.35	$SR_1=(var-1.225)/0.125$
3	SR_2	0.73	0.98	$SR_2=(var-0.855)/0.125$
4	x_4	2.5	4.5	$x_4=(var-3.5)/1.0$

y is the desired response which is the percentage in NO reduction after reburning

TABLE 5
THE PROPOSED EXPERIMENTAL DESIGN

Test	x_1	SR_2	SR_1	x_4
1	-1	-1	-1	-1
2	1	1	-1	-1
3	1	-1	1	-1
4	-1	1	1	-1
5	1	-1	-1	-1
6	-1	1	-1	-1
7	-1	-1	1	-1
8	1	1	1	-1
9	1	-1	-1	1
10	-1	1	-1	1
11	-1	-1	1	1
12	1	1	1	1
13	-1	-1	-1	1
14	1	1	-1	1
15	1	-1	1	1
16	-1	1	1	1
17	-1	1	0	0
18	-1	-1	0	0
19	-1	0	1	0
20	-1	0	-1	0
21	-1	0	0	1
22	-1	0	0	-1
23	1	0	0	0
24	-1	0	0	0
25	1	1	0	0
26	1	-1	0	0
27	1	0	1	0
28	1	0	-1	0
29	1	0	0	1
30	1	0	0	-1

TABLE 6
DATA FOR SCREENING STUDY

Test	X_1	SR_2	SR_1	X_4	Y,exp	Response NOp	Tp	Tr	Tb	RTr
21B	-1	-1	-1	-0.915	58.4	1000	1473	1335	1294	0.224
22A	1	1	-1	-0.915	45.3	995	1503	1435	1209	0.851
16A	1	-1	1	-1.132	65.7	965	1418	1411	1297	0.569
18A	1	-1	1	-0.934	69.7	1070	1433	1425	1317	0.521
17B	-1	1	1	-1.132	42.2	1000	1430	1327	1306	0.278
19B	-1	1	1	-0.934	49.4	1100	1426	1338	1319	0.256
21A	1	-1	-1	-0.915	69.6	1000	1488	1439	1278	0.634
22B	-1	1	-1	-0.915	40.5	995	1505	1313	1227	0.331
16B	-1	-1	1	-1.132	49.4	965	1407	1319	1301	0.197
18B	-1	-1	1	-0.934	55.1	1070	1424	1345	1307	0.18
17A	1	1	1	-1.132	62.8	1000	1436	1427	1289	0.74
19A	1	1	1	-0.934	55.4	1100	1432	1429	1306	0.685
7A	1	-1	-1	0.865	77.8	1130	1658	1617	1468	0.329
8B	-1	1	-1	0.865	22.4	1020	1661	1550	1490	0.162
1B	-1	-1	1	1.13	57.1	1140	1653	1581	1544	0.085
3B	-1	-1	1	1.13	60.6	1090	1549	1562	1547	0.085
4A	1	1	1	1.13	43.3	1090	1620	1637	1536	0.327
7B	-1	-1	-1	0.865	56.4	1130	1683	1576	1503	0.113
8A	1	1	-1	0.865	30.7	1020	1656	1635	1487	0.432
1A	1	-1	1	1.13	79.4	1140	1667	1645	1564	0.245
3A	1	-1	1	1.13	80.3	1090	1614	1622	1546	0.25
4B	-1	1	1	1.13	30.3	1090	1546	1529	1517	0.122
10B	-1	1	0	0.005	31.9	985	1577	1501	1482	0.185
12B	-1	1	0	-0.074	33.3	1020	1458	1362	1338	0.208
9B	-1	-1	0	0.005	57.0	975	1572	1491	1451	0.131
14B	-1	0	1	-0.074	43.7	1040	1471	1434	1418	0.153
15B	-1	0	-1	-0.074	36.1	950	1559	1458	1393	0.192
6B	-1	0	0	0.865	38.4	1090	1598	1539	1518	0.124
20B	-1	0	0	-0.934	48.5	1090	1434	1337	1323	0.24
11A	1	0	0	0.005	66.1	1030	1569	1570	1511	0.432
13A	1	0	0	-0.074	67.5	1030	1513	1495	1363	0.475
11B	-1	0	0	0.005	43.8	1030	1569	1505	1477	0.146
13B	-1	0	0	-0.074	44.7	1030	1503	1406	1362	0.174
10A	1	1	0	0.005	47.7	985	1572	1569	1475	0.492
12A	1	1	0	-0.074	43.3	1020	1461	1453	1335	0.555
9A	1	-1	0	0.005	76.2	975	1552	1534	1437	0.388
14A	1	0	1	-0.074	62.1	1040	1475	1500	1423	0.428
15A	1	0	-1	-0.074	73.1	950	1534	1505	1360	0.523
6A	1	0	0	0.865	65.7	1090	1628	1621	1474	0.344
20A	1	0	0	-0.934	73.6	1090	1474	1443	1307	0.655
0A	1	-0.7	1	1.13	78.6	1210	1667	1645	1564	0.246
0B	-1	-0.7	1	1.13	54.5	1200	1653	1581	1544	0.094
2A	1	0	-0.2	1.13	64.5	977	1694	1677	1542	0.312
2B	-1	0	-0.2	1.13	36.7	977	1686	1588	1550	0.127

TABLE 7
RESPONSE IN TERMS OF CONTROLLED VARIABLES
* * * * MULTIPLE REGRESSION * * * *

Multiple R	.954	Analysis of Variance			
R Square	.909		DF	Sum of Squares	Mean Square
Adjusted R square	.897	Regression	5	9183.8	1836.76
Standard Error	4.907	Residual	38	915.0	24.08

F= 76.28 Signif F= .0000

--------------Variables in the Equation--------------------------

Variable	B	SE B	95% confdnce Intrvl B		F	Sig F
SR_2	-13.27	.952	-15.19	-11.34	193.99	.0000
X_1	9.28	.740	7.78	10.77	157.20	.0000
SR_2X_4	-6.19	1.026	-8.26	-4.11	36.39	.0000
X_4	-2.32	.908	-4.16	-.48	6.55	.0146
SR_2SR_1	2.67	1.075	.49	4.84	6.14	.0178
(Constant)	53.63	.750	52.11	55.15	5111.20	.0000

--------------Variables not in the Equation---------------------

Variable	F	Sig F
SR_1	3.152	.0840
X_1SR_2	3.856	.0571
X_1SR_1	.020	.8879
X_1X_4	2.863	.0990
SR_1X_4	1.499	.2286
SR_2^2	2.029	.1627
SR_1^2	.494	.4865
X_4^2	.174	.6786

TABLE 8
RESPONSE IN TERMS OF DERIVED VARIABLES
SR_2=0.73

Multiple R	.873	Analysis of Variance			
R Square	.762		DF	Sum of Squares	Mean Square
Adjusted R Square	.718	Regression	2	1050.1	525.07
Standard Error	5.464	Residual	11	328.5	29.86
		F= 17.58		Signif F= .0004	

---------------Variables in the Equation---------------

Variable	B	F	Sig F
RTr	37.0	18.588	.0012
Tr	.068	24.827	.0004
(CONSTANT)	-47.5	4.983	.0473

---------------Variables not in the Equation-----------

Variable	F	Sig F
NOp	.005	.9453
Tp	.507	.4926
Tb	.085	.7760

TABLE 9
RESPONSE IN TERMS OF DERIVED VARIABLES
SR_2=0.855

Multiple R	.952	Analysis of Variance			
R Square	.906		DF	Sum of Squares	Mean Square
Adjusted R Square	.889	Regression	3	2332.8	1166.4
Standard Error	4.694	Residual	11	242.3	22.03
		F= 52.94		Signif F= .0012	

---------------Variables in the Equation---------------

Variable	B	F	Sig F
RTr	79.6	98.920	.0000
Tr	.032	4.780	.0513
(CONSTANT)	-16.7	.579	.4627

---------------Variables not in the Equation-----------

Variable	F	Sig F
NOp	.632	.4452
Tp	2.118	.1763
Tb	.495	.4976

TABLE 10
RESPONSE IN TERMS OF DERIVED VARIABLES
SR_2=0.98

Multiple R	.885	Analysis of Variance			
R Square	.783		DF	Sum of Squares	Mean Square
Adjusted R Square	.718	Regression	3	1203.2	401.07
Standard Error	5.772	Residual	10	333.2	33.32
		F= 12.038		Signif F= .0012	

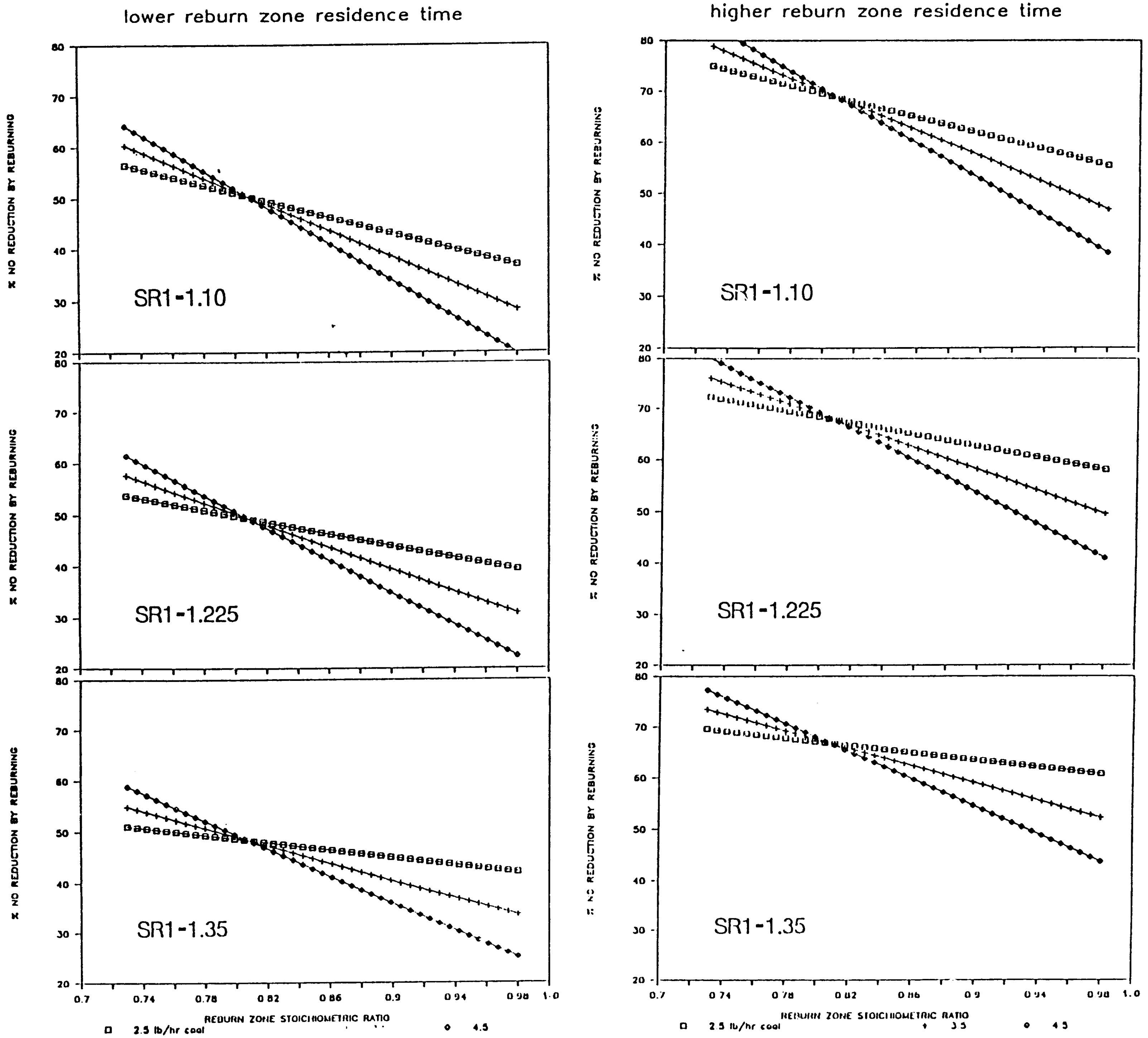
---------------Variables in the Equation---------------

Variable	B	F	Sig F
RTr	81.3	20.330	.0011
Tr	-.195	10.977	.0078
Tb	.196	8.432	.0157
(CONSTANT)	22.7	.680	.4289

---------------Variables not in the Equation-----------

Variable	F	Sig F
NOp	1.678	.2274
Tp	.366	.5603

Figure 1. SCREENING STUDY FOR CLASSICAL REBURNING
PRIMARY FUEL: UTAH BITUMINOUS #2 - REBURN FUEL: METHANE GAS



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