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AN INVESTIGATION TO DEFINE THE PHYSICAL/CHEMICAL CONSTRAINTS WHICH LIMIT NO_x EMISSION REDUCTION ACHIEVABLE BY REBURNING

Quarterly Report 5

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DISCLAIMER

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1.0 INTRODUCTION

Reburning is a combustion modification technique which removes NO_x from combustion products by using fuel as a reducing agent. Previous studies have shown that natural gas is more effective than coal as a reburning fuel. It is believed that 60 percent reduction in NO_x emission can be achieved with natural gas reburning. However, kinetic calculations indicate that emission reductions greater than 80 percent are possible using the reburning process.

The objectives of this program are to define the chemical and physical constraints which prevent the attainment of 80 percent NO_x reduction with reburning and to test improved configurations for reburning as an advanced NO_x control technique for coal-fired boilers. The program has been divided into two experimental scales. Bench scale studies are designed to screen the chemical and physical means for enhancing reburning efficiency. Subsequent pilot studies will evaluate the impacts of finite rate mixing on the effectiveness of the various concepts. These studies have been supported with chemical kinetics and boiler performance modeling to generalize the experimental data to full scale boilers. Specifically, the program consists of the following:

- Bench scale studies
    - N_2 formation in reburning zone
    - XN conversion in burnout zone
- Pilot scale studies
- Interpretation and generalization
- Final Report

This quarterly report documents the experimental results obtained in the bench scale studies. The focus is on the chemistry of N_2 formation in the reburning zone. The experiments were conducted in the bench scale Control Temperature Tower (CTT). The CTT is a downfired, refractory lined furnace which is 20 cm in diameter and 2.4 m in length. The baseline conditions for these tests were:

- Primary fuel-natural gas at 80 x 10^3 Btu/hr; primary NO_x, $(NO_x)_p$ = 240 or 600 ppm (dry, 0% O_2) by doping; SR_1 = 1.1.

- Reburning gas injection at 2600°F (T_1); reburning zone residence time (τ_2) = 400 ms.

- Burnout air injection at 2300°F (T_2), SR_3 = 1.25.

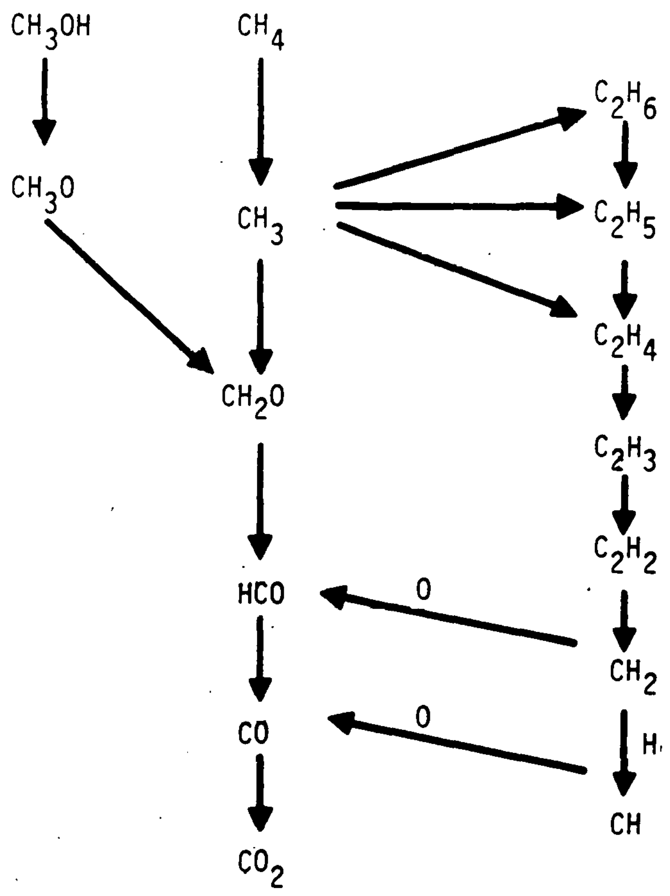
2.0 EXPERIMENTAL RESULTS

In the reburning process, NO is reduced to molecular nitrogen. The process is initiated by reaction of NO with a hydrocarbon radical, and the reaction pathway passes through intermediate cyanide and amine species prior to the evolution of molecular nitrogen. Reburning chemistry is shown in Figure 2-1. Upon introduction of burnout air, the intermediate nitrogenous species can form N_2 or be oxidized to NO. Thus, the amount of these species at the reburning zone exit and their subsequent conversion has an impact on reburning efficiency. Experimental results presented in this section focus on the characterization of reactive species, including NO, NH_3, HCN, CO and THC, at the reburning zone exit, and the effect of temperature on reburning performance.

2.1 Reactive Species

Speciation at the reburning zone exit was characterized by sampling 400 msec downstream of the reburning fuel injection location. This is the usual location for burnout air addition. Nitrogen species, measured at three rich zone stoichiometries, are shown in Figure 2-2. The concentration of NO decreased with stoichiometry, while the concentration of intermediate species rose. When SR_2 was decreased from 0.9 to 0.7, the amount of HCN rose dramatically, while NH_3 levels exhibited little change. This is an indication of a lack of oxidizing radicals available at the richer stoichiometry for processing HCN through NCO to NH_3. Optimum reburning efficiency was previously established at a fuel rich zone stoichiometry of 0.9. Since a richer stoichiometry did not result in a more efficient process, a significant amount of rich zone HCN must have been oxidized to NO in the burnout zone at an SR_2 of 0.7. Primary NO_x concentration had only a minor impact on the percent conversion of NO to nitrogenous intermediates.

In Figure 2-3, CO and total hydrocarbon concentrations at the reburning zone exit are presented for two reburning zone temperatures. The reburning zone temperature was varied by changing its location in the furnace. A 400 msec residence time was maintained for all of the temperatures tested.



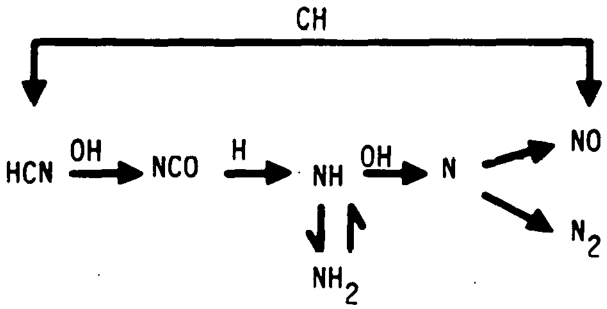


Figure 2-1. Hydrocarbon and nitrogen chemistry relevant to reburning.

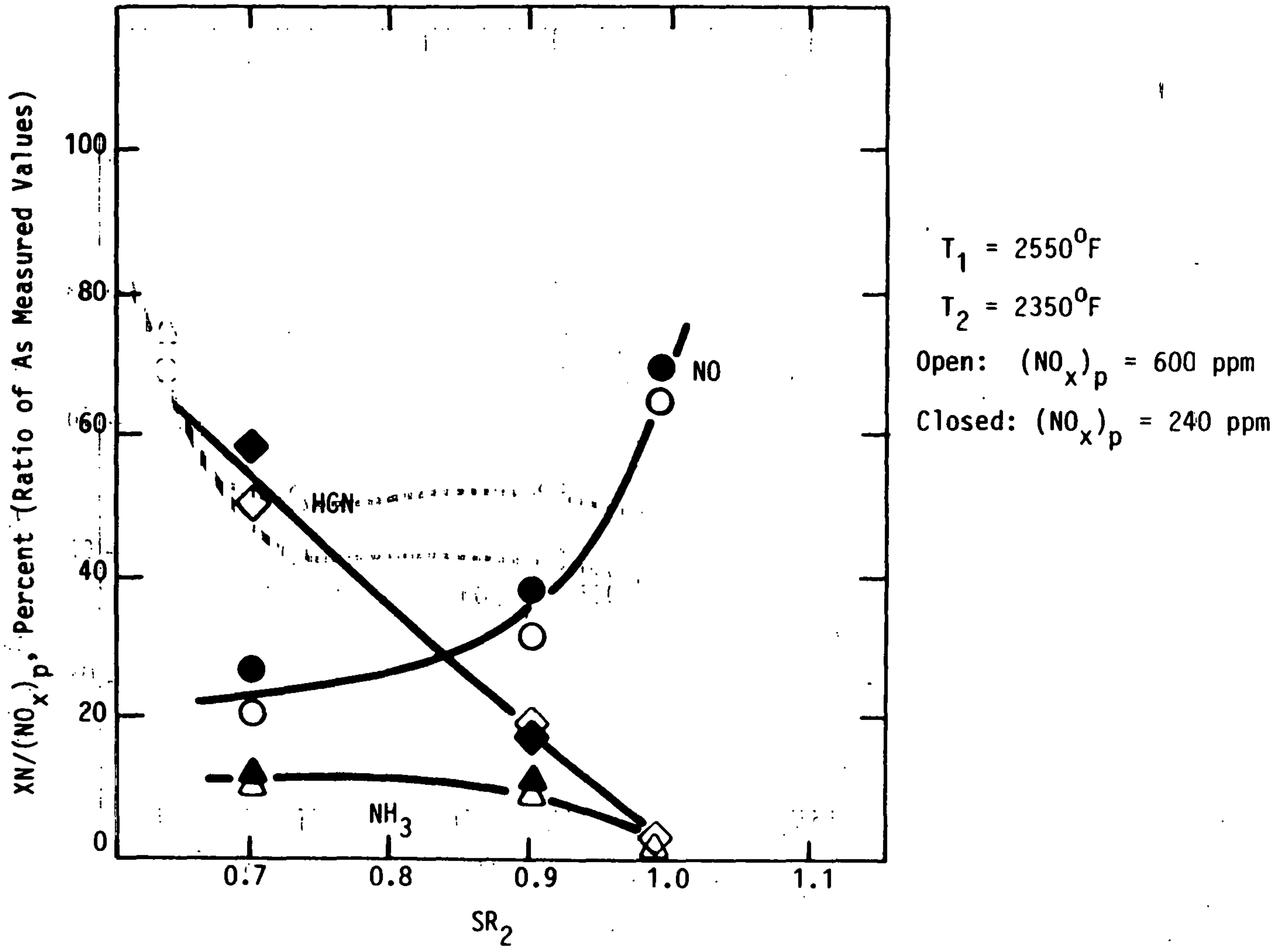


Figure 2-2. Nitrogen speciation at the reburning zone exit.

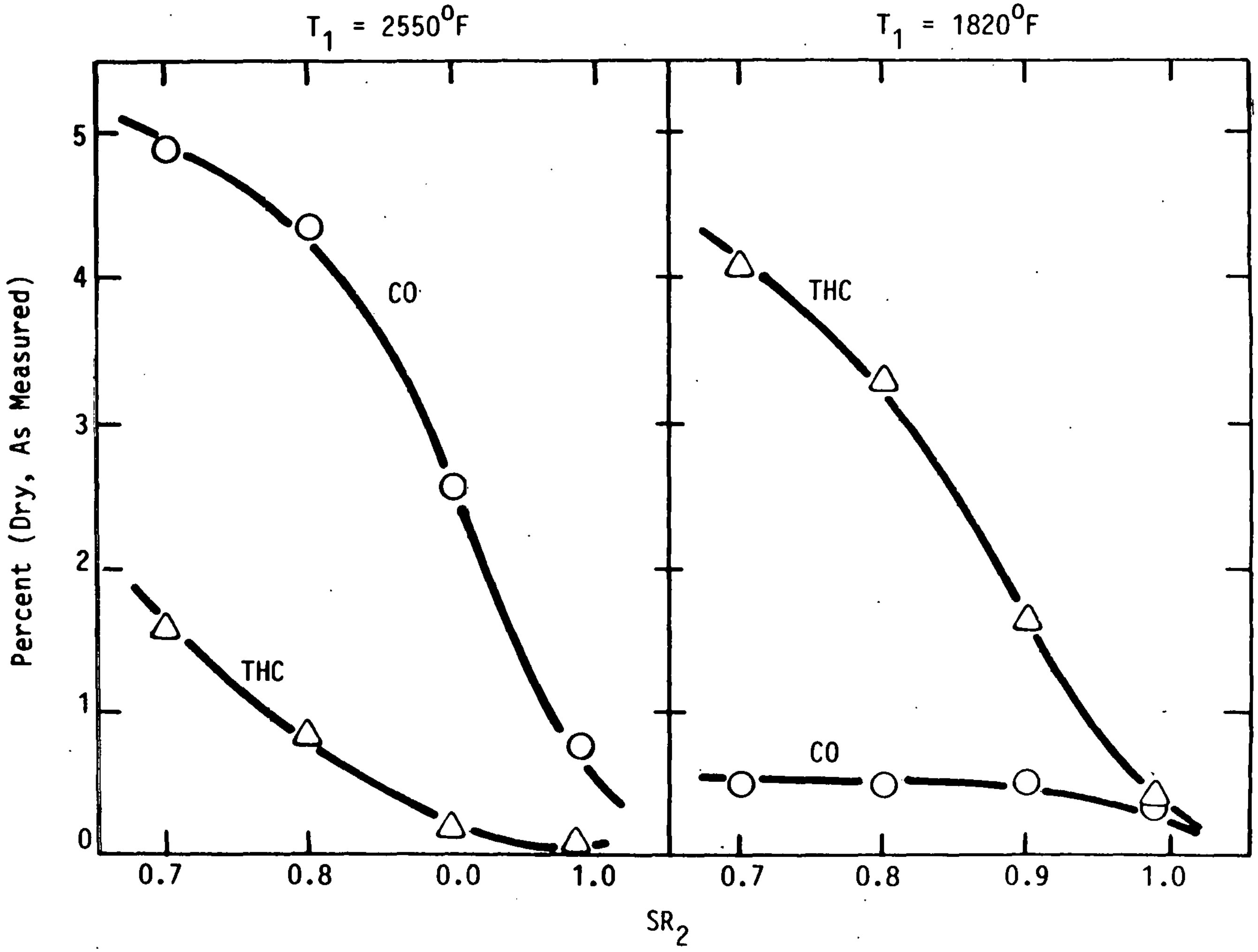


Figure 2-3. Influence of temperature on reburning fuel burnout.

These data had a rich zone entrance temperature (T_1) of 1800°F. At the higher temperature, both species increased in concentration as the zone became more fuel rich. The amount of CO was considerably higher than THC at all stoichiometries, which indicated extensive destruction of the natural gas CH constituents. At the lower temperature, THC levels rose rapidly as SR_2 decreased, while CO concentration was virtually independent of stoichiometry. These high THC concentrations were indicative of incomplete natural gas combustion, as the theoretical concentration of CH_4 (natural gas was approximately 93 percent CH_4) would range from 1 to 4 percent for the stoichiometries tested. The slow decomposition of natural gas at this temperature apparently limited NO destruction by CH radicals at an SR_2 of 0.9.

2.2 Temperature Dependence

Figure 2-4 shows the influence of temperature on rich zone NO reduction. At a stoichiometry of 0.9, the NO reduction efficiency of the reburning zone decreased with temperature, with NO concentration measured at the highest temperature case approximately half of that seen at a T_1 of 1800°F. However, when the rich zone was just fuel rich ($SR_2 = 0.99$), the NO reduction efficiency of the reburning zone was virtually independent of temperature from 1800 to 2600°F.

A comparison of the two stoichiometries indicates that at 2600°F NO reduction is more efficient at the richer stoichiometry, while reduction at 1800°F was similar for both stoichiometries. A similar amount of NO destruction at 1800°F may well be attributed to the fact that the concentration of reactive hydrocarbons were similar at both stoichiometries. the THC measurements indicated that only a small portion of the natural gas combusted at either stoichiometry at 1800°F. Figure 2-4 also shows that varying the primary NO level had no effect on reduction efficiency at low temperature, with data at higher temperatures indicating a measurable influence of primary NO. This implied that NO was not the limiting reactant at low temperature. At higher temperatures, when hydrocarbon availability

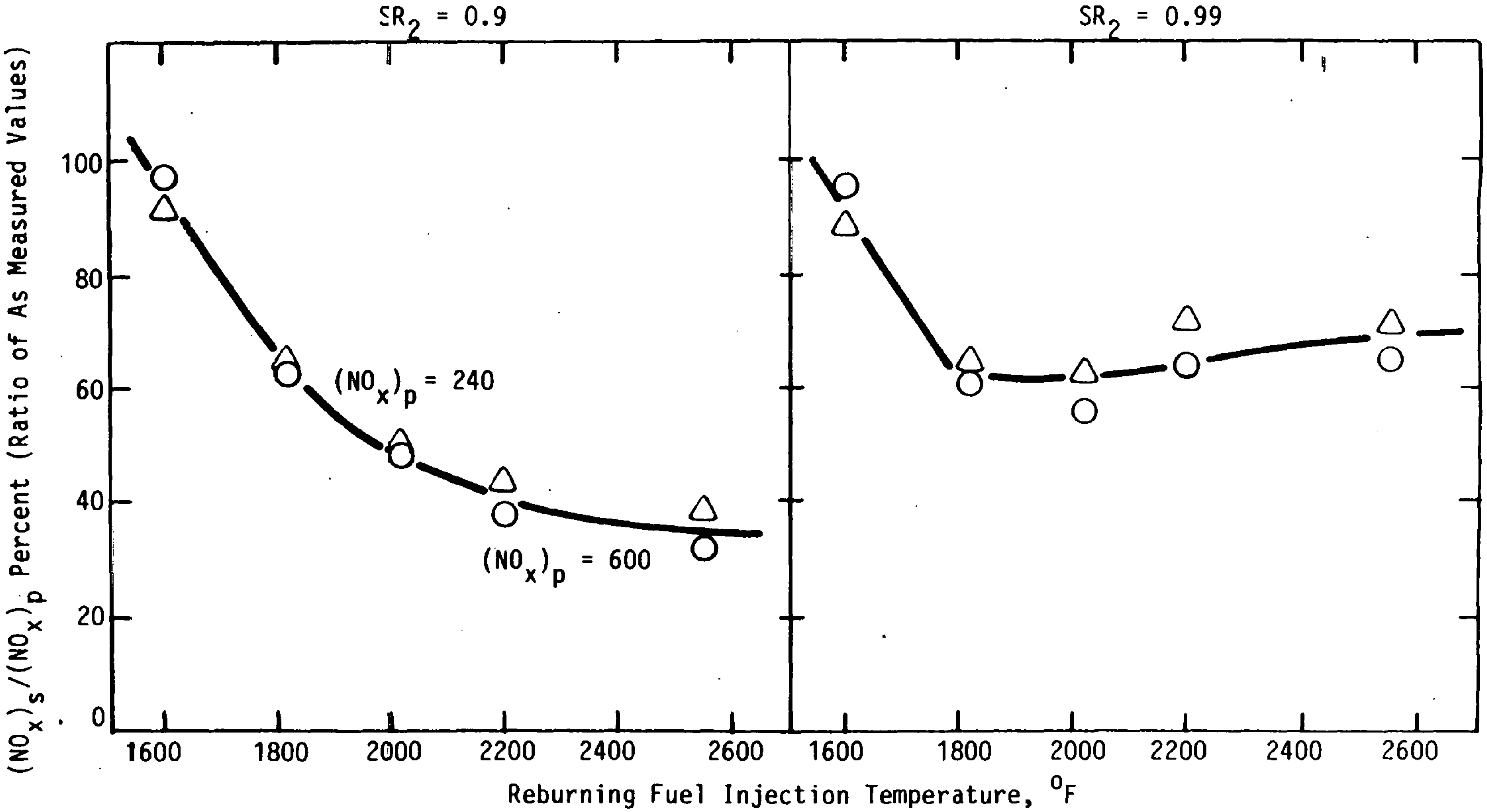


Figure 2-4. Influence of temperature on reburning zone efficiency.

increased due to faster natural gas combustion, the availability of NO apparently did affect the process efficiency.

In Figure 2-5, the influence of temperature on the overall reburning process is presented. These data were obtained at an SR_2 of 0.9 with burnout air included in the system. By comparison of NO reduction efficiencies for the rich zone (Figure 2-4), with these data, the effect of temperature on burnout zone efficiency can be discerned. The data indicate that the efficiency of the overall reburning process was constant from 1800 to 2200°F. A further increase in temperature resulted in a more efficient process. When compared to Figure 2-4, the data show that the rich zone temperature dependence on NO reduction was much stronger than the dependence of the overall process on temperature. Thus, at lower burnout zone temperatures, the processing of intermediate nitrogen species to molecular nitrogen must have been more efficient. Careful scrutiny of the data shows that at higher temperatures, NO is actually formed in the burnout zone, while at 1800°F, NO was reduced to molecular nitrogen in the final zone. For all of the data, reburning at or below 1600°F was ineffective. Application of reburning at these temperatures was not practical due to the limited combustibility of natural gas.

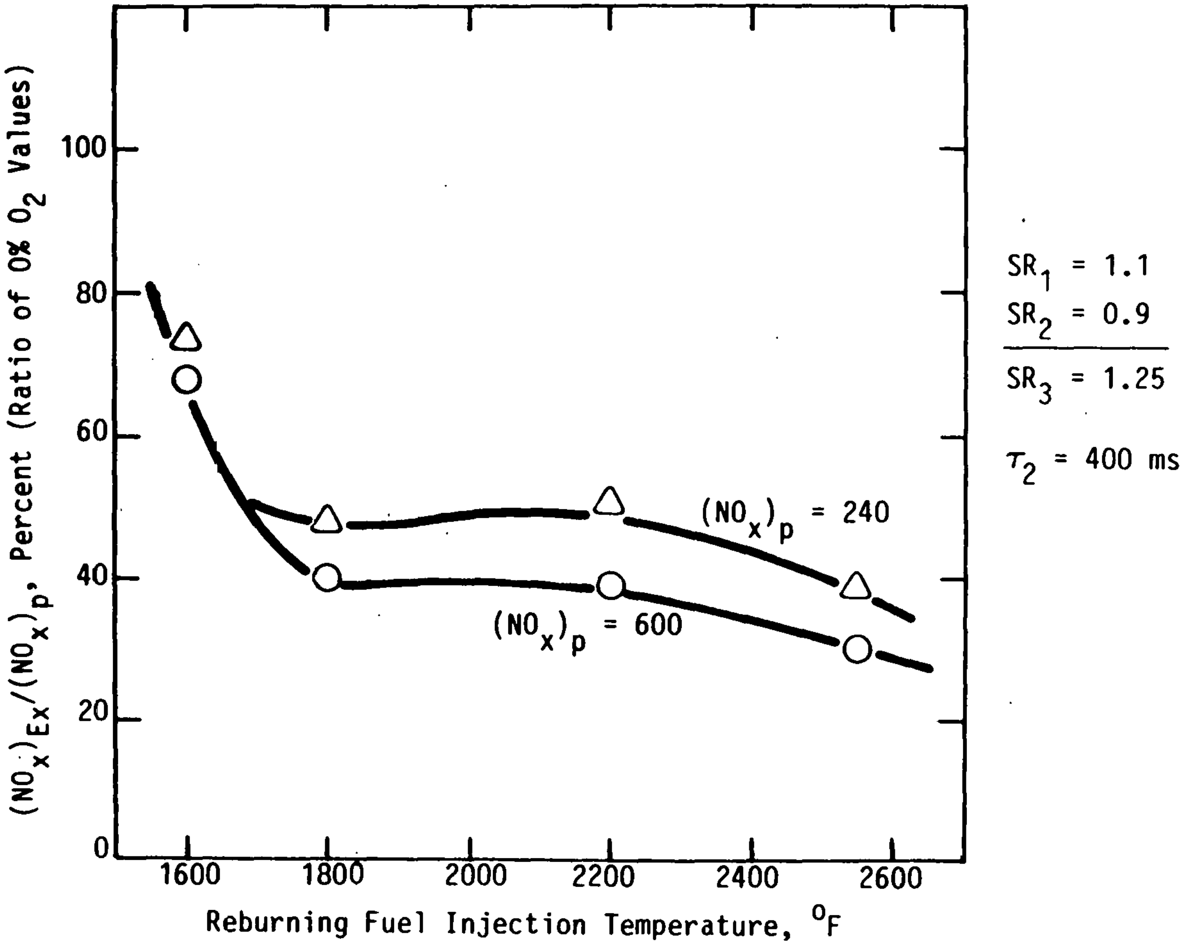


Figure 2-5. Influence of temperature on reburning process efficiency.

3.0 FUTURE WORK

In the next reporting period, the bench scale experimental work will focus on the chemistry of XN conversion in the burnout zone. Specifically, the effect on NO_x reduction in the burnout zone by NH_3 and HCN will be investigated. Also, the impact of THC and CO on NO_x reduction effectiveness will be examined.