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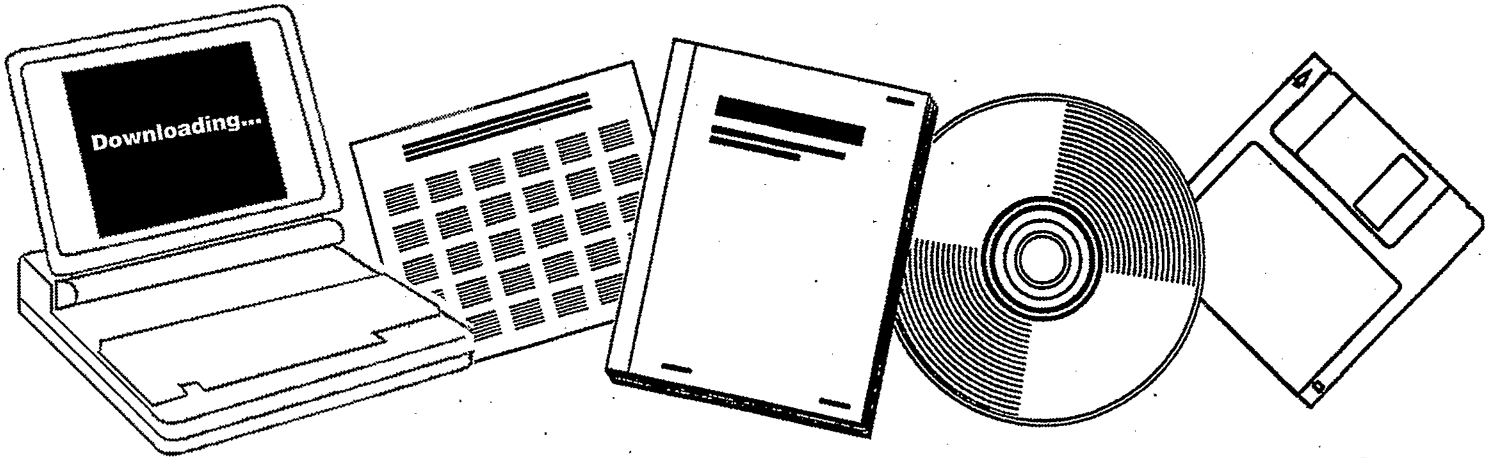

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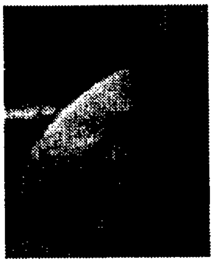

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CO + H_2 Reactions Over Nitrogen-Modified Iron Catalysts

Quarterly Technical Progress Report for the Peroid Jan 1, 1983 - March 31, 1983

W. Nicholas Delgass

Purdue University
West Lafayette, Indiana 47907

PREPARED FOR THE

U.S. DEPARTMENT OF ENERGY

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ABSTRACT

The objective of this work is to examine the effects of nitrogen, as nitride and as a gas phase reactant, on hydrocarbon synthesis reactions over iron catalysts. This is the second quarterly period of a three year program. Preliminary Mössbauer and kinetic studies of bulk iron nitrides suggest that selectivity and stability of the catalysts vary with nitride phase. Procedures for preparing pure phases will be improved so that this relationship can be studied more closely. Computer simulation of transient kinetic responses has also been initiated. The reactor and kinetic description is still oversimplified, but responses qualitatively similar to those expected have been achieved.

1. OBJECTIVES AND SCOPE

1.1 BACKGROUND

The feasibility of utilizing synthesis gas ($CO + H_2$) via the Fischer-Tropsch reaction pathway for the production of fuels and chemicals is well established. The SASOL ventures, for example, take advantage of abundant coal resources to produce both desirable synthetic automotive fuels and basic chemical feedstocks. The applicability of these chemical transformations is nonetheless limited. The present procedure requires extensive processing if the production of non-essential byproducts is to be avoided. The discovery and subsequent usage of improved catalysts would therefore be advantageous.

Experimental results published in the current literature show that nitrogen affects the performance of iron catalysts-catalysts which find widespread use in the Fischer-Tropsch synthesis route. Prenitriding of the iron catalyst has been reported to shift the product distribution to one exhibiting lower molecular weight fractions and enhanced alcohol yields (1). On the other hand, simultaneous introduction of ammonia (NH_3) with synthesis gas produces nitrogeneous compounds. Furthermore, and probably of greater importance, this addition of ammonia effects a reduction in the overall chain length of compounds in the product spectrum (2,3). It is of considerable interest, therefore, to study these and other characteristics of nitrided iron catalysts in order to gain a basic understanding of their behavior. Discovery of the new pathways in Fischer–Tropsch synthesis afforded by nitrogen will add to the fundamental knowledge from which future synthesis-catalysts can be derived.

1.2 OBJECTIVES

The scope of the program may be broken down into two main areas of concern. Firstly, consideration must be given to the role of the surface nitrogen in

i. altering the product distribution and

ii. stabilizing catalyst activity

of the synthesis reactions. *In-situ* Mössbauer studies, already in progress, will identify the various iron nitride phases and allow for examination of their stability during reaction. The Mossbauer results will form the basis for detailed kinetic tracer experiments involving transient and isotope labeling analyses. Ultrahigh vacuum work using SIMS and AES will supplement the Mössbauer and kinetic characterizations.

The second area of consideration will involve the kinetic and catalytic effects observed during the addition of ammonia to the synthesis gas stream. Transient work will be ideal for observing initial activity changes occurring as a result of NH_3 pulses. Along with ultrahigh vacuum studies, the transient kinetics of NH_3 addition will help clarify which steady state experiments would be most productive. The various analytical methods will define interactions between surface and bulk nitrogen, and their role in effecting new reaction pathways.

The primary experiments which define our route to understanding which parameters influence the selectivity and alter the activity of synthesis reactions may, therefore, be outlined as follows:

i. Mossbauer and simultaneous kinetics of prenitrided iron catalysts will be used to determine nitride phase stability and to correlate these phases to reaction selectivity. The initial pressure of one atmosphere may later be increased.

ii. Similar analysis of the effects of addition of NH_3 to the reactant stream will be performed.

iii. Transient analysis and isotope tracer studies of synthesis reactions over prenitrided catalysts will determine surface nitride stability. The stoichiometry at the surface and influence of nitriding on CO dissociation will be sought.

iv. Ultrahigh vacuum analysis will examine surface stoichiometry and reaction intermediates. Interaction between the nitrided phases and adsorption bond strengths of CO and H_2 will be investigated.

v. The effects of NH_3 addition to the reactant stream will be similarly followed by UHV and transient tracer studies to determine possible alterations in reaction pathways envoked by the presence of NH_3.

Computer modeling will be undertaken to aid in quantitative interpretation of transient data.

2. TECHNICAL PROGRESS

2.1 LITERATURE REVIEW

2.1.1 Advantages of Nitrided Iron Catalysts

The transition metal carbides and nitrides can possess certain catalytic advantages relative to their parent transition metals. Recent literature shows that both carbides and nitrides can sometimes offer better selectivities and greater resistances to poisoning by carburization. In addition, better activity maintenance and overall selectivity towards lower molecular weight products is evident in Fischer-Tropsch synthesis (1,4). Examples of these greater activities and altered selectivities have been reported in the previous quarterly report (5).

2.1.2 Mossbauer Effect Characterization of Iron Nitrides

A description of the nitride phases and methods for their preparation have been reported (5). The detailed nitride structures, although partially characterized by the X-ray studies of Jack (6,7), are still not comprehensively understood. Mössbauer effect and magnetization measurements are continuing to add to the understanding of these catalysts.

Nozik et al. (8) have studied the γ'-Fe_4N nitride while Bainbridge et al. (9) and Chabanel et al. (10) have used Mössbauer spectroscopy and magnetization measurements on the ϵ-Fe_2N/ζ-Fe_2N nitrides. Work on the ϵ-Fe_xN nitride has been performed by various authors for varying nitrogen contents. Eickel and Pitsch (11) have researched the ϵ-$Fe_{3.2}N$ phase while Chen et al. (12) have concentrated upon the ϵ-$Fe_{2.67}N$ and ϵ-$Fe_{2.47}N$ phases. The primary results of these investigators are presented in Table One. As a result of their work, Eickel and Pitsch (11) present a structural model, originally devised by Weiner and Berger (13), as an update on the model for ϵ-iron nitride formulated by Jack (6,7). Chen et al. (12) have proposed a different model for the same system.

The data from Table One indicate the discrepancies between the characterization and modeling of the ϵ-Fe_xN phase. Agreement has not been reached on the positioning of the nitrogen atoms in the nitride lattice, or on the Mössbauer characterization of structural assignments. Identification of the γ' and ζ phases does appear straightforward, however. Attempts to produce these pure phases are in progress. Analyses of the ϵ phase and possible complications of superparamagnetism will be part of our future research.

2.2 SUMMARY OF RESULTS

Both first year reseachers have now fulfilled their core course and teaching obligations and are beginning fulltime

TABLE ONE Mössbauer parameters of iron nitrides.

Nitride	Iron Identity	Temperature (K)	Isomer Shift (mm/s)	Quadrupole Split. (mm/s)	Hyperfine Field (koe)	References
γ'-Fe_4N	Fe0	300	.24	0.0	340.6	8.
	FeIIa	300	.30	-.22	215.5	8.
	FeIIb	300	.30	.43	219.2	8.
ε-$Fe_{3.2}N$	2nn	300	.33	0.0	238	15.
	1nn	300	.24	0.0	298	15.
ε-$Fe_{2.67}N$						
	FeII	300	.34	---	205	12.
	FeIII	300	.40	---	99.5	12.
	FeII	4.2	.48	---	258	12.
	FeIII	4.2	.54		126	12.
ε-$Fe_{2.47}N$	FeII	300	.35	---	186	12.
	FeIII	300	.41	---	84	12.
	FeII	4.2	.47	---	259	12.
	FeIII	4.2	.57/.60	---	142/66	12.
ε-$Fe_{2.20}N$	FeII	4.2	.48	---	244	12.
ε-$Fe_{2.07}N$	FeIII	4.2	.59	---	60	12.
ε-Fe_2N	3nn	78	.40	.26	0.0	9.
ξ-Fe_2N	3nn	300	.41	.28	0.0	11.
		78	.53	.25	0.0	11.
		78	.42	.27	0.0	9.
		4.2	.57	.29	0.0	11.

nn ≡ nearest neighbor

research for the summer. The upgrading of the Mössbauer Apparatus has been completed and will accelerate our Mössbauer study capabilities significantly. We discuss here preliminary Mössbauer results and initiation of computer simulations of transient kinetics. The simulations to date consider the surface reactions of CO and H_2 on a simplistic level, but will form the basis for development of quantitative analyses of our transient data.

2.2.1 Preliminary Mossbauer Studies

The bulk iron nitride catalysts were prepared from a physical mixture of 30 mg of α-Fe_2O_3 with 270 mg of SiO_2 as a diluent. The mixed powder was dampened with distilled water to facillitate mechanical pressing into a wafer. The wafer was then dried in the Mössbauer cell for half an hour at 120°C in flowing H_2. Further reduction in pure H_2 was performed at 400°C for an additional 4 hours. The nitriding procedure consisted of exposing the wafer to various flowrates of NH_3 and H_2 at temperatures of between 350 and 400°C. Room temperature Mössbauer spectra were then obtained upon completion of the nitriding steps and subsequent cell cooling. The Fischer-Tropsch synthesis reactions were then started with a switch to an He carrier gas which contained the H_2/CO reactant mixture. The total reaction gas flowrate was maintained at 150 ml/min. while that of CO was at 10% of this total. H_2/CO ratios were changed by altering the relative amounts of H_2 and He in the feed stream. After the reaction study was complete, the catalyst was cooled and the Mössbauer spectrum again recorded.

The preparative methods used to attempt to produce a pure ε-iron nitride were those presented by Bouchard et al. (14). The reduced wafer was nitrided for 3.25 hr at 400°C with ammonia and hydrogen flowrates at 200 ml/min and at 15 ml/min respectively. We expected to obtain an iron nitride high in nitrogen content ($Fe_{2.2}N$ to $Fe_{2.5}N$), but the Mössbauer spectrum of this catalyst did not exhibit the anticipated room temperature magnetic splitting. This may be indicative of the very high nitrogen containing ζ-nitride, or may arise from small size, superperamagnetic particles. Figure 1a shows the apparent single peaked spectrum obtained from this wafer. Fischer-Tropsch synthesis conditions of 250°C for 6 hrs in a $CO:H_2:He = 1:3:6$ flow were then instituted. The Mössbauer spectrum, shown in Figure 1b, was obtained at room temperature upon completion of the synthesis reaction. This spectrum shows a magnetically split pattern which may be due to the formation of a carbonitride structure. The characterization of the carbonitride phases has not yet been started.

According to Anderson (1), carrying out the nitriding at 400°C for only one hour in a flow of 280 ml NH_3/hr and 50 mlH_2/hr should produce a low-nitrogen ε-nitrite. The room temperature Mössbauer spectrum for the sample receiving this treatment, Figure 1c, shows the presence of limited quantities of the γ′-Fe_4N in addition to the ε phase. The presence of ε-nitride is indicated by the sextet with an H = 245 kOe, and two small shoulders

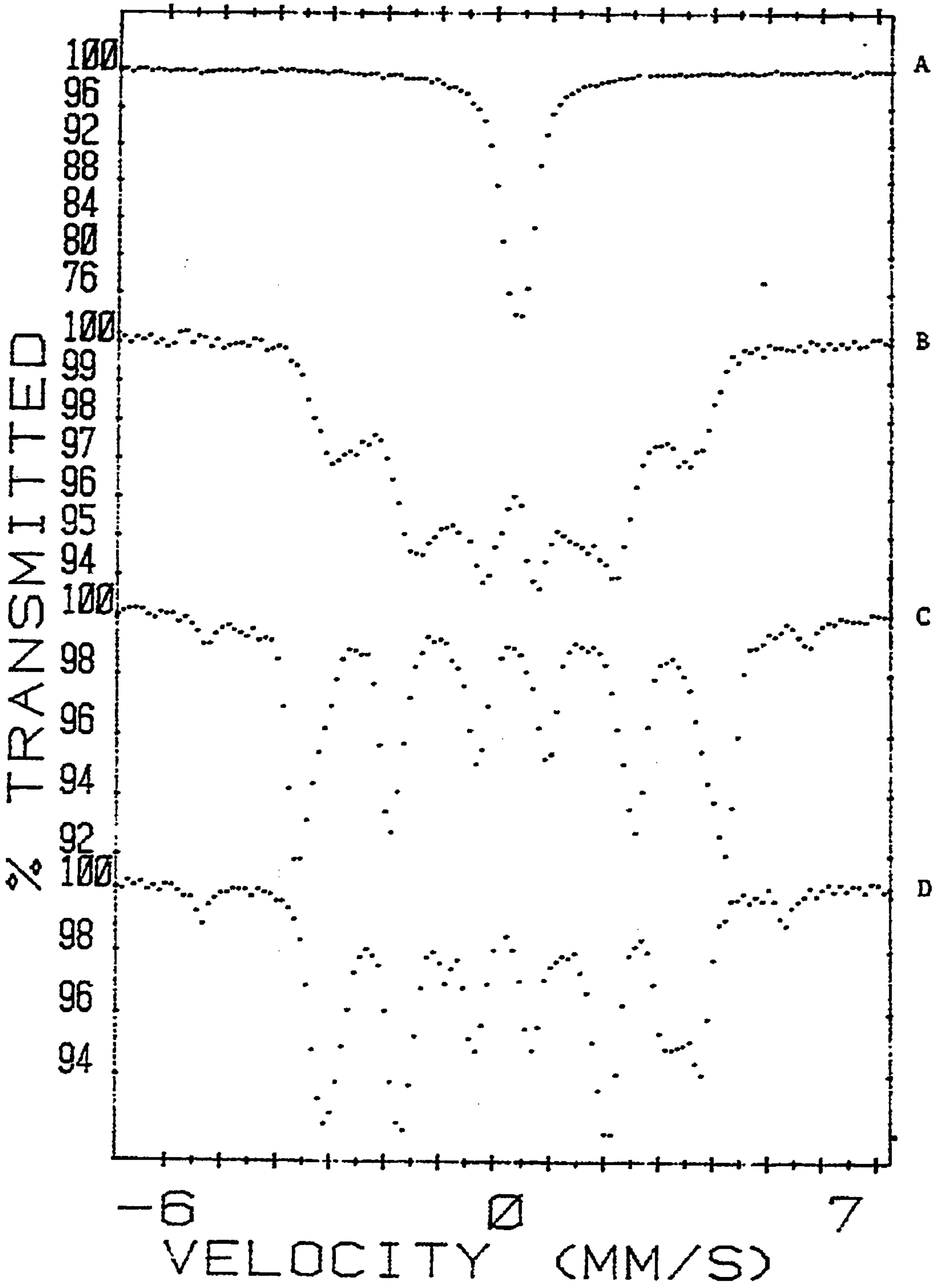

Figure 1. Room Temperature Mössbauer Spectra of $\alpha Fe_2O_3/SiO_2$, reduced 4 hrs in H2 at 400°C. A) after 3.25 hrs nitriding at 400°C and B) after 6 hrs FTS at 250°C C) after 1 hr nitriding at 350°C and D) an additional 2 hr nitriding at 400°C and then 6 hrs FTS at 250°C.

at 300 kOe. The approximate nitride phase compositions are estimated to be 15-20% γ'-Fe_4N and 80-85% ϵ-Fe_xN. A further nitriding for 2 additional hours at identical conditions produced little change in the Mössbauer spectrum of this catalyst. The specrum after FTS, shown in Figure 1d, deviates only slightly from that obtained after the extended nitriding. Carbonitrides may; however, be responsible for the peak occuring at between 3-3.5 mm/s in this Figure.

The Mössbauer results to date confirm that various bulk iron nitrides can be identified. The characterization of the carbonitrides encountered during Fischer Tropsch synthesis may; however, be more difficult. The preparation of the pure nitride phases appears not to be as facile as reported by others (1,14). Thus, some adjustments in preparation procedure will be made to improve phase purity so that reactivity studies of the individual phases can be undertaken.

2.2.2 Activity and Selectivity

These initial kinetic studies have shown the iron nitride phases prepared to have differing activities. The combined ϵ and γ' phase recorded a maximum activity of 7.7×10^{-5} gmol CO converted/min.g Fe after 0.65 hours of FTS. This later decreased by more than 50% to 3.6×10^{-5} gmol CO converted /min.g Fe after 3.55 hr FTS. The catalyst obtained after a 3.25 hour nitriding with lower H_2 partial pressure also appeared to reach maximum activity after 0.65 hours of FTS. This catalyst exhibited an

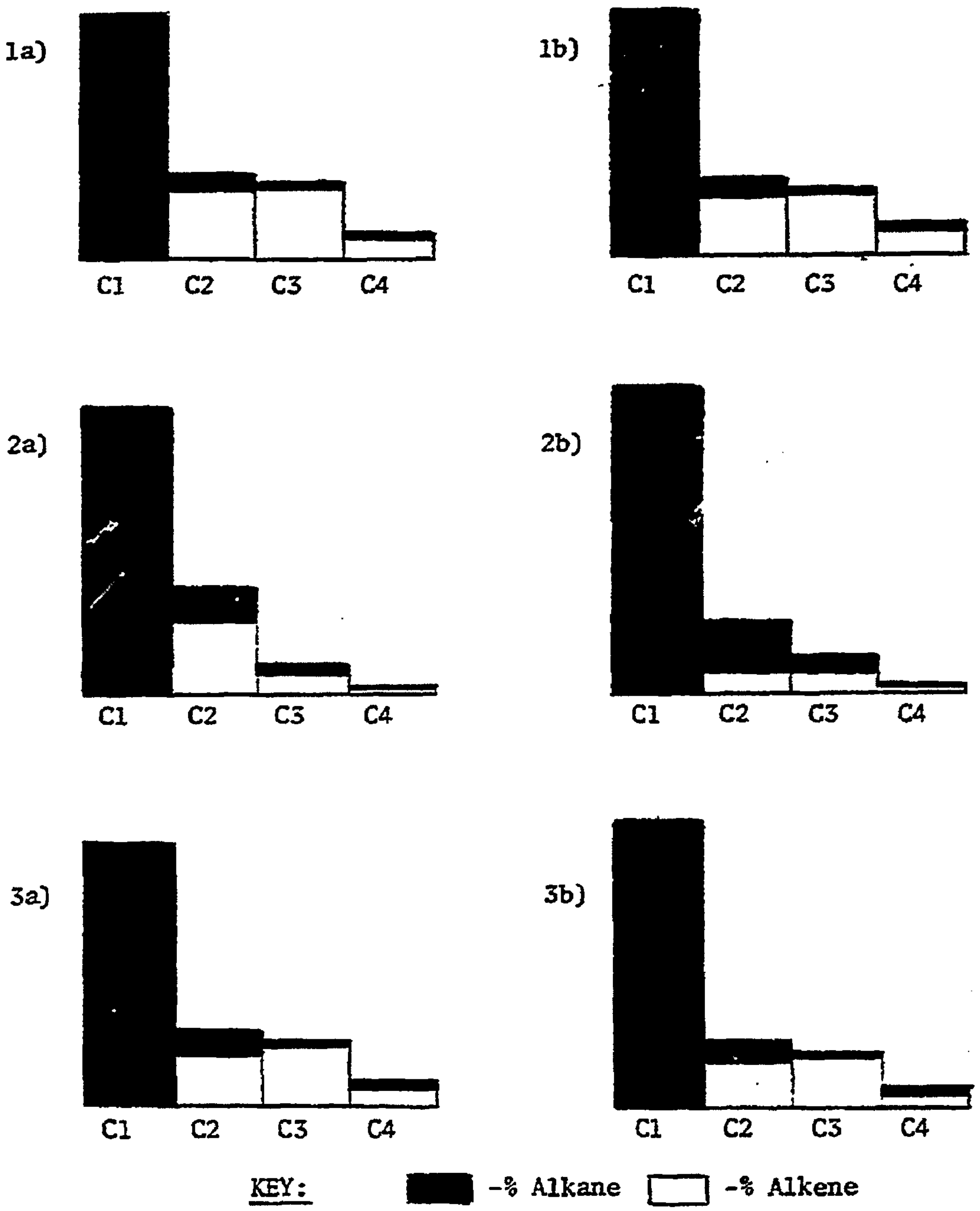

Figure 2. Selectivities of nitrided iron catalysts toward alkane and alkene production. All catalysts $\alpha Fe_2O_3/SiO_2$ reduced in H_2 4 hr at 400°C, nitrided, then subjected to $3CO/H_2$ at 250°C 1) nitrided 3.25 hr in 13.3 NH_3/H_2 at 400°C then FTS for a) .067 hr and b) .65 hr 2) nitrided 4 hr in 4 NH_3/H_2 at 400°C then FTS for a) .067 hr and b) 3.55 hr 3) nitrided in 4 NH_3/H_2 for 3 hr at 350°C and 2.5 hr at 400°C then FTS for a) .067 hr and b) 0.65 hr.

activity of 3.7×10^{-5} gmol CO converted /min.g Fe at that time.

The nitride phases prepared so far give distinct shapes for the conversion of CO versus time curves. A maximum activity peak appears to be present for those catalysts containing mostly ϵ-Fe_xN. The catalyst characterized as containing mostly γ'-Fe_4N, on the other hand, exhibits an exponential type decay in activity. This aspect of kinetic characterization will be examined more closely in future work.

For the short Fischer Tropsch synthesis runs reported above, the selectivities of the ζ-Fe_2N and γ'-Fe_4N nitrides appear to favor greater olefin production compared to the ϵ-Fe_xN. The latter nitride has a product distribution shifted towards lower hydrocarbons compared to the former two. These selectivity comparisons are detailed in Figure 2, where data was taken at approximately equivalent CO conversions. These kinetic indications are encouraging as they suggest the existence of different catalytic properties for the individual nitrides.

2.2.3 Initial Kinetic Simulations

In our initial kinetic modeling, it was our purpose to create a computer program to model the transient methanation reaction

$$CO + 3H_2 \longrightarrow CH_4 + H_2O,$$

the simplest of the complex Fischer-Tropsch reactions. As developed up to now, the program models the reactor as a CSTR (or differential PFR), and determines the concentration of all species during a transient pulse. A number of CSTRs in series can approximate the behavior of an integral PFR, which is more applicable to very fast reactions such as CO chemisorption. The next phase of our computer modeling will include this capability.

The current model involves the well-known equations that describe a CSTR, with the reaction rate modeled by a series of elementary kinetic steps. Writing equations for the material balance on each kinetic species generates differential equations describing the time derivatives of all species concentrations. These equations are numerically integrated on the computer by means of the Gear method, which is particularly useful for stiff systems of differential equations.

The difficult task of this procedure is choosing the particular sequence of mechanistic steps for the model. Bell's (16) review of the catalytic synthesis of hydrocarbons discusses various methanation mechanisms, and he supports the formation of CH_4 via successive addition of surface hydrogen to a CH_x adsorbed intermediate. The mechanism is proposed as follows:

$$CO + S \rightleftharpoons COS \tag{1}$$

$$COS + S \rightleftharpoons CS + OS \tag{2}$$

$$H_2 + 2S \rightleftharpoons 2HS \tag{3}$$

$$HS + OS \rightleftharpoons OHS + S \tag{4}$$

$$OHS + HS \rightleftharpoons H_2O + S \tag{5}$$

$$CS + 3HS \rightleftharpoons CH_3S + 3S \tag{6}$$

$$CH_3S + HS \rightleftharpoons CH_4 + S \qquad (7)$$

Note that this model requires that CO dissociate on the surface to lead to the CH_x intermediate; a step considered likely over many catalysts (Fe, Co, Ni) (16).

The following assumptions were used to estimate rate constants for the simulation results presented here:

1. CO adsorbs as often as a molecule hits an empty site.
2. steps (4) and (6) are in equilibrium
3. the rate of hydrogenation of C-S is about equal to the rate of CO-S dissociation.

The magnitudes of the rate constants used for this simulation are presented in Table 2.

Table 2

Values of Rate Constants for Kinetic Simulation

The rate equations

$$r_1 = k_1(CO)(S) - k_{-1}(COS)$$

$$r_2 = k_2(COS)(S) - k_{-2}(CS)(OS)$$

$$r_3 = k_3(H_2)(S)^2 - k_{-3}(HS)^2$$

$$r_4 = k_4(HS)^2(OS)/(S) - k_{-4}(H_2O)(S)$$

$$r_5 = k_5(S)(HS)^4/(S)^3 - k_{-5}(CH_4)(S)$$

All values in ( ) are mole fractions or fractions of surface covered; are rates in units of 1/sec.

The rate constants

$k_1 = .4 \times 10^7$

$k_{-1} = 2.4 \times 10^3$

$k_2 = 1.2 \times 10^1$

$k_{-2} = 2.4 \times 10^4$

$k_3 = 5.3 \times 10^7$

$k_{-3} = 1.6 \times 10^4$

$k_4 = 2.8 \times 10^1$

$k_{-4} \sim 0$

$k_5 = 1.8 \times 10^{-2}$

$k_{-5} \sim 0$

Rates (4) and (5) contain (S) in the denominator because of the assumption of equilibrium in steps (4) and (6) in the model. Since (S) is always small, the relative magnitudes of rate (2) (CO dissociation) and rate (5) (hydrogenation of C-S) are made approximately equal by the choice of the rate constants k_2 and k_5. Note that the adsorption of hydrogen is limited by the requirement of two empty sites; and its rate of adsorption is small compared to that of CO.

Figure (3) shows the result of a step to pure H_2 feed from a steady state reaction mixture with 3:1:2 CO/H_2/He feed. The plot shows outlet conditions of the CSTR. The removal of He denotes the step change in the inlet stream. Before the step, COS nearly completely covered the surface (see Figure 4). With the removal of CO from the gas phase, H_2 has an opportunity to adsorb, thus increasing the rate of methane production until the carbon is used up. These results reflect qualitatively what has been observed experimentally on Co and Rh catalysts in our laboratory.

Figure (4) shows the surface coverage values that are also predicted by the computer simulation. Once rates of individual steps are determined via transient and isotopic experiments, the simulation can give quantitative information on surface coverage and may suggest new tests of a particular kinetic description.

Our goal is to combine carefully planned experiments with computer simulations to give quantitative estimates for the rate constants in a proposed sequence of elementary steps. Comparison

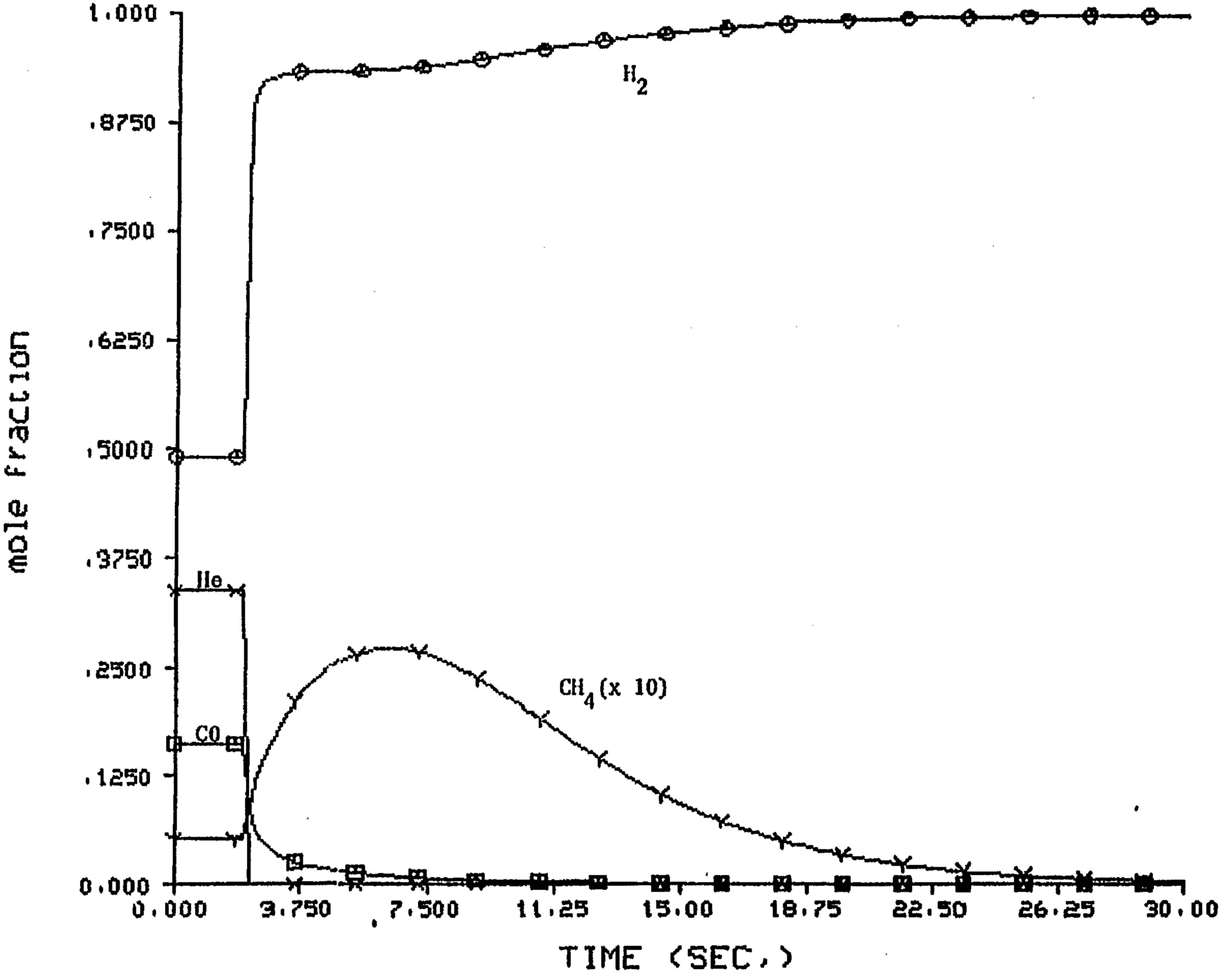

Figure 3. Simulation of transient response of gas phase mole fractions to a stop from $3H_2/CO/2He$ to H_2.

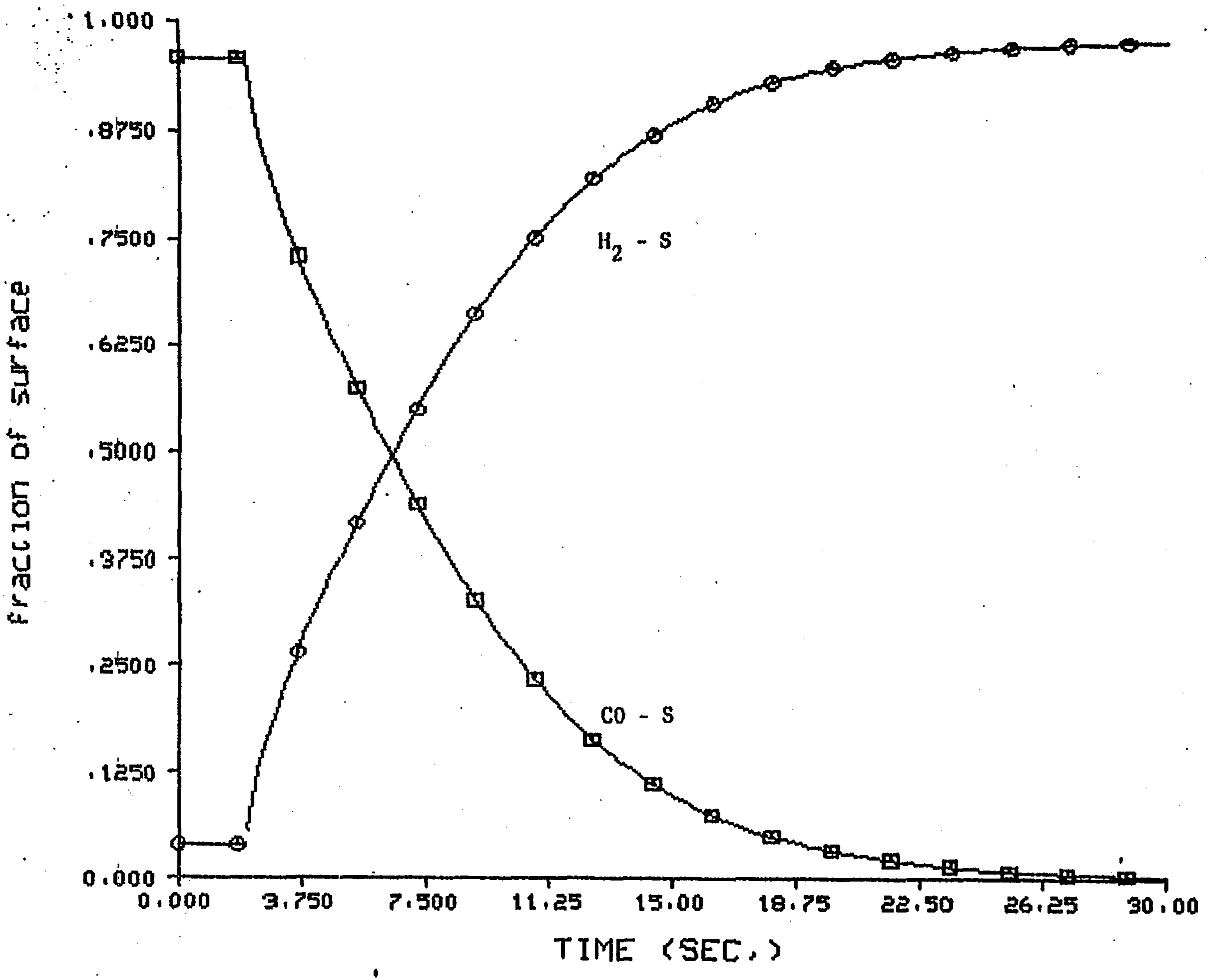

Figure 4. Simulation of transient response of fractional surface coverage to a step from $3H_2/CO/2He$ to H_2.

of these constants for different catalysts will then give an indication of the kinetic consequences of chemical changes in the catalyst surface.

3. FUTURE RESEARCH

Research in the next 3 month period will emphasize Mössbauer characterization of the iron nitride phases and detailed kinetic analyses of FTS reactions occurring on these catalysts. In this next phase the nitride stability and kinetic studies will deal initially with the single phase iron nitrides (γ', ϵ and ζ), rather than combinations of these individual phases. Exact preparative procedures will have to be investigated, as difficulties have been experienced in this field. We will utilize the Mössbauer effect in both this characterization, and in the subsequent stability analyses. The duration of the FTS runs will be increased from the present six hour maximum to provide further stability determinations. The scope of using the Mössbauer effect may be increased to include partial characterization of the carbonitride system, as greater surface carbon deposition is expected to occur with these extended Fischer Tropsch syntheses. These *in situ* kinetic measurements will be supplemented by parallel experiments on the transient equipment.

The transient experiments will begin with studies of surface nitride stability and effects of surface nitride stoichiometry on catalytic selectivity. Long term stability studies will be aided by the recent addition of a gas chromatograph to analyze the effluent of the transient reactor. We expect to continue and elaborate on our capability to do computer "experiments". These will allow us to test various approaches to studying nitride catalyst systems and will permit a more streamlined program of actual experiments. This capability should be especially valuable in future mechanistic studies where the responses of trial sequences of elementary steps can be tested to show the most sensitive experiments.

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5. APPENDICES

5.1 Distribution List

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