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**TECHNOLOGY DEVELOPMENT FOR IRON
FISCHER-TROPSCH CATALYSIS**

Contract No. DE-AC22-⁹⁴~~91~~PC94055

Quarterly Technical Progress Report No. 8

Covering the Period July 1, 1996 to September 30, 1996

Prepared for

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2. Contract Objectives

The objective of this research project is to develop the technology for the production of physically robust iron-based Fischer-Tropsch catalysts that have suitable activity, selectivity and stability to be used in the slurry phase synthesis reactor development. The catalysts that are developed shall be suitable for testing in the Advanced Fuels Development Facility at LaPorte, Texas, to produce either low- or high-alpha product distributions. Previous work by the offeror has produced a catalyst formulation that is 1.5 times as active as the "standard-catalyst" developed by German workers for slurry phase synthesis. The proposed work will optimize the catalyst composition and pretreatment operation for this low-alpha catalyst. In parallel, work will be conducted to design a high-alpha iron catalyst this is suitable for slurry phase synthesis. Studies will be conducted to define the chemical phases present at various stages of the pretreatment and synthesis stages and to define the course of these changes. The oxidation/reduction cycles that are anticipated to occur in large, commercial reactors will be studied at the laboratory scale. Catalyst performance will be determined for catalysts synthesized in this program for activity, selectivity and aging characteristics.

The research is divided into four major topical areas: (a) catalyst preparation and characterization, (b) product characterization, (c) reactor operations, and (d) data assessment.

To accomplish the objectives of the project, these topics have been organized into the following technical tasks:

- a. Task 1.0 Development of Optimum Promoter Levels for Low- and High-Alpha Catalysts

- 1.1 Determine Optimized Synthesis Procedure for High-Alpha Iron-Based Fischer-Tropsch Catalysts
 - Role of precursor particle size on activity.
 - Role of Cu in precipitated catalysts.
 - Define attrition resistance.
- 1.2 Prepare Catalysts that can be Used to Determine the Role of Promoters for Low- and High-Alpha Catalysts
 - Define optimum SiO_2 .
 - Define optimum Al_2O_3 .
- 1.3 Prepare Catalysts that can be Used to Quantify the Role of K on Product Selectivity in both Low- and High-Alpha Catalysts.
- 1.4 Complete the Optimization of the Two Best Low-Alpha, Iron-Based Fischer-Tropsch Catalysts Developed during the Previous Contract.

b. Task 2.0 Definition of Preferred Pretreatment for both Low- and High-Alpha Fischer-Tropsch Catalysts.

- 2.1 Determine the Role of Cu in the Activation of Precipitated Low- and High-Alpha, Iron-Based Fischer-Tropsch Catalysts.
- 2.2 Determine the Effect of K Content on Activation Procedures and Determine if the Method of Addition has any Effect on Catalyst Activity and Life.
- 2.3 Determine the Physical and Chemical Changes that Occur during Catalyst Pretreatment and Use and Determine how these Changes Effect the Strength of the Catalysts.

2.4 Evaluate the Effect of Carbon Deposition during Catalyst Activation on Activity, Selectivity and Aging Characteristics.

c. Task 3.0 Catalyst Structure and Characterization.

d. Task 4.0 Catalyst Testing.

4.1 Verify the Quality of Data Obtained from the CSTR's.

4.2 Measure Catalyst Performance.

4.3 Determine the Stable Phases that Exist during Synthesis at Low and High CO Conversion Levels.

4.4 Obtain Data on the Rates Involved in the Interconversion of Iron Oxide and Iron Carbide.

e. Task 5.0 Reports.

3. Summary of Activities

A successful 300+ hour run of the CAER slurry bubble column reactor (SBCR) was completed. The run demonstrated the operability of the SBCR using a 20 wt.% catalyst slurry. Mass balances were 95-102%.

A Fe supported MgAl_2O_3 catalyst was prepared and studied in the continuous stirred tank reactor (CSTR). The physical properties for this catalyst are compiled in Table 1 and the comparison of XRD peaks for $\alpha\text{-Fe}_2\text{O}_3$ and the $\text{Fe-MgAl}_2\text{O}_4$ catalyst is given in Table 2.

Despite the high potassium loading, the activity and selectivity of the $100\text{Fe}/6.0\text{Cu}/6.1\text{K}/370\text{MgAl}_2\text{O}_4$ catalyst are typical of our base-line, low-alpha precipitated catalysts (Table 3) with much lower K:Fe ratio ($100\text{Fe}/2.2\text{Si}/x\text{Cu}/0.7\text{K}$, $x=0$ or 3.0, parts per weight). This implies that a substantial amount of the potassium in the $100\text{Fe}/6.0\text{Cu}/6.1\text{K}/370\text{MgAl}_2\text{O}_4$ catalyst is combined with the support. A new

catalyst batch with a higher iron loading and higher K:Fe ratio has recently been prepared (100Fe/6.0K/8.0/260MgAl₂O₄). The activity and selectivity of this catalyst will be reported in the next quarter. In addition, similar catalysts will be prepared with Al₂O₃ and ZnAl₂O₄ supports.

4. Status, Accomplishments, Results and Discussion

A.0. - Task 1.0. Development of Optimum Promoter Levels for Low- and High-Alpha Catalysts

The goal of this task is to identify and optimize procedure for the preparation of iron-based catalysts that combine high activity selectivity and life with physical robustness. Each of the subtasks address an area of considerable uncertainty in the synthesis of catalysts.

A.1. Determine Optimized Synthesis Procedure for High-Alpha Iron-Based Catalysts

No scheduled or further activity to report.

A.2. Prepare Catalysts that can be Used to Determine the Role of Promoters for Low- and High-Alpha Catalysts

No scheduled or further activity to report.

A.3. Prepare Catalysts that can be Used to Quantify the Role of K on Product Selectivity for both High- and Low-Alpha Catalysts

Low-alpha catalyst work has been successfully completed.

A.4. Complete the Optimization of Two Best Low-Alpha, Iron-Based Catalysts Developed During the Previous Contract

Completed.

A.5. Schedule of Activities for Next Quarter

- No scheduled activity to report.

B.0. - Task 2.0. Definition of Preferred Pretreatment for Both Low- and High-Alpha Catalysts

The goals of this task are to define the preferred treatment, to define the role of Cu and K during the pretreatment on activity and selectivity and to define the chemical and physical changes which occur during the preferred pretreatment. The subtasks address each of these goals.

B.1. Role of Cu in the Activation of Precipitated Low- and High-Alpha, Iron Based Fischer-Tropsch Catalysts

This task has been successfully completed for both high and low alpha catalysts.

B.2. Determine the Effect of K Content on Activation Procedures and Determine if the Method of Addition has any Effect on Catalyst Activity and Life.

The work on the method of addition has been completed.

B.3. Physical and Chemical Changes that Occur During Pretreatment and Use

No scheduled or further activity to report.

B.4. Effect of Carbon Deposition

No scheduled or further activity to report.

B.5. Schedule of Activities for Next Quarter

Continue to determine effectiveness of Ba and Zn as promoters. These potential promoters should, to a first approximation mimic the alkali promoters we have been studying.

C.0. - Task 3.0. Catalyst Structure and Characterization

The goal of this task is to provide basic analyses (surface area, XRD) of all catalyst prepared and to provide additional techniques as required (Mössbauer, SEM, XPS, etc.) to answer specific questions or to provide basic required characterization data for the catalysts.

C.1. Schedule of Activities for Next Quarter

Continue to provide the characterization data as required.

D.0. - Task 4.0. Catalyst Testing

The goals of this task are to operate the eight CSTR reactors, measure catalyst performance, determine the stable phases that exist during synthesis at low and high conversions and to determine the rates of interconversion of iron oxide and carbide.

D.1. Verify the Quality of Data Obtained from the CSTRs.

This task is now successfully completed.

D.2. Measure Catalyst Performance

D.2.1. Fe Supported MgAl₂O₄ Catalysts

A logical route to the development of a robust FTS catalyst, that can be easily separated from reactor wax, is to employ a large particle support. In general, supported iron catalysts have been considered to be inferior to precipitated iron catalysts because of strong interactions between iron and the most commonly used supports: alumina and silica. To neutralize the acidity of silica and alumina, a large amount of basic promoters (potassium) must be used to achieve reasonable alkene selectivity and low methane yields. Unfortunately a consequence of using a high level of potassium is that stability is generally poor. In addition, a loss of FTS activity may occur if iron reacts with the support to form an inactive species such as iron silicates

or $\text{Fe}_x\text{Al}_{3-x}\text{O}_4$ mixed spinels. The reaction of alumina with iron can be prevented by using an aluminate as a support. Toledo et al. have studied the dehydrogenation of 1-butene with iron supported on zinc aluminate and with $\text{ZnFe}_x\text{Al}_{2-x}\text{O}_4$ mixed spinels [J. A. Toledo, M. A. Valenzuela, H. Armendáriz, G. Aquilar-Ríos, B. Zapata, A. Montoya, N. Nava, P. Salas and I. Schifter, *Catal. Lett.* 30 (1995) 279]. They found that the supported catalyst was the most active because iron was located in octahedral vacancies on the surface of the ZnAl_2O_4 support and did not migrate into the bulk of the support to form $\text{ZnFe}_x\text{Al}_{2-x}\text{O}_4$ species. We have had promising attrition results with supported $\text{Fe/Cu/K/Al}_2\text{O}_3$ FTS catalysts. Catalysts with up to 30% iron loading remained intact (Figure 1) after 400 h of FTS in a CSTR; however, a large amount of potassium was needed to obtain reasonable methane selectivity. As part of our effort to develop a robust catalyst, we have decided to study the FTS of iron supported on various aluminates. Using an aluminate support such as MgAl_2O_4 or ZnAl_2O_4 may provide the structural integrity of alumina with improved activity by blocking the formation of $\text{Fe}_x\text{Al}_{3-x}\text{O}_4$ species. Reported here are preliminary results for a supported iron on MgAl_2O_4 catalyst.

Preparation of MgAl_2O_4 . A small scale batch of Mg-Al hydroxides was prepared by precipitation. An aqueous solution (645 mL) of 55.15 g $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (215 mmol) and 161.25 g $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (430 mmol) was stirred vigorously with a mechanical stirrer while 195 mL of 29.5% ammonium hydroxide was added rapidly (final pH = 9.8). The resulting slurry was stirred for 10 minutes and then filtered. The filter cake was reslurried with a volume of distilled, deionized water equal to the filtrate volume and then filtered again. A second wash following the same procedure was repeated. The filter cake was then dried in a furnace at 125°C for 24 h.

Five calcination procedures were conducted to convert the Mg-Al hydroxides to MgAl_2O_4 : 500°C, 600°C, 700°C and 800°C for six h and according to a variation of the procedure reported by Rennard and Freel [R. J. Rennard and J. Freel, *J. Catal.* 98 (1986) 235], 400°C for 16 h followed by 800°C for an additional 16 h. A 600 g batch of MgAl_2O_4 was prepared by following the latter calcination procedure.

Preparation of 100Fe/6.0Cu/6.1K/370MgAl₂O₄ 275 g of $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, 8.72 g of $\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ and 6.00 g of KNO_3 were placed in a 600 mL beaker and melted at 70°C by heating in an oil bath. MgAl_2O_4 (78.89 g) which had been calcined at 400°C for 16 h and 800°C for 16 h was screened to 325 X 60 mesh (45 to 250 μm) and also placed in a 600 mL beaker in the oil bath. The melt was slowly added to the MgAl_2O_4 with a pipet followed by thorough mixing with a spatula. This was continued until the MgAl_2O_4 appeared to be saturated. The mixture was then transferred to a ceramic dish and calcined at 350°C for 2 h. The same impregnation procedure was repeated with the Fe/Cu/K/ MgAl_2O_4 powder followed by calcination at 350°C for 4 h. The nominal iron content of the catalyst following the second impregnation was 18.5 wt% (100Fe/6.0Cu/6.1K/370MgAl₂O₄ parts per weight).

Activation and FTS Conditions. 40 g of 100Fe/6.0Cu/6.1K/370MgAl₂O₄ and 290 g of Ethylflo 164 oil were mixed in a one L CSTR. Hydrogen flow was started at 120 SLPH and 0.10 MPa. The temperature of the reactor was increased to 270°C at 2°C min⁻¹ and held there for 24 h. After the 24 h activation, carbon monoxide flow was started and the flow rates were adjusted to 3.1 NL h⁻¹g-Fe⁻¹. Pressure was then increased to 1.31MPa and FTS was started.

XRD of Mg-Al hydroxides following various calcination procedures are shown in Figure 2. Calcination at 500, 600, 700 and 800°C for 4 h produced materials which had XRD peaks corresponding to the spinel structure; however, peak intensities increased with increasing calcination temperature so it was not clear if the Mg-Al hydroxides had been completely converted to MgAl₂O₄. The XRD of γ -Al₂O₃ and MgAl₂O₄ are very similar so to ensure complete formation of MgAl₂O₄, a calcination procedure similar to that of Rennard and Freel was utilized: 400°C for 16 h followed by 800°C for an additional 16 h. XRD following this calcination procedure showed the sharpest peaks corresponding to MgAl₂O₄. The unit cell volume of this material was 528 Å³ which is identical to the literature value (JCPDS, 21-1152) of synthetic MgAl₂O₄. The surface areas of the Mg-Al hydroxide material following the different calcination procedures are shown in Table 1. As expected, the surface areas decreased with increasing calcination temperature; however, it was decided that the surface area following the 400°C and 800°C calcination procedure was sufficient to be used as a support. XRD of the 100Fe/6.0Cu/6.1K/370MgAl₂O₄ catalyst is compared to the MgAl₂O₄ support in Figure 3. In addition to the obvious peaks corresponding to the spinel structure of MgAl₂O₄, peaks corresponding to α -Fe₂O₃ are clearly visible. The unit cell volume of the MgAl₂O₄ did not change with iron impregnation and the major peaks of α -Fe₂O₃ are similar to the literature values (Table 2) thereby indicating that there was little or no reaction between the support and the impregnated iron.

Activity and Selectivity. FTS activity of the 100Fe/6.0Cu/6.1K/370MgAl₂O₄ catalyst is very promising. Operating under our base-line conditions of 270°C, 1.31 MPa, H₂:CO=0.7 and 3.1 NL h⁻¹g-Fe⁻¹ carbon monoxide conversions were as high as 90% before leveling off at just under 80% after 200 h of FTS (Figure 4). After 200 h the

catalyst is stable with a deactivation rate of only 0.38% carbon monoxide conversion per week. Hydrocarbon production mirrored the carbon monoxide conversion leveling off at 140 g m⁻³ of syngas (Figure 5). FTS activity in terms of g of hydrocarbon produced per g of Fe per h was between 0.53 and 0.45 (Figure 6). Methane selectivity was ~10% of the carbon monoxide converted to hydrocarbons and the combined methane and ethane selectivity was ~15% (Figure 7). The alkene selectivity for the C₂, C₃ and C₄ fractions is shown in Figure 8. Alkene selectivities for the C₃ and C₄ fractions were identical at 75% while the ethylene selectivity was only 20% for the C₂ fraction. Approximately 79% of the C₄ alkene was 1-butene which indicates that isomerization was low (Figure 8).

Carbon dioxide selectivity was approximately 49% of the converted carbon monoxide (Figure 9) which is characteristic of high water-gas shift activity. A H₂:CO usage ratio of 0.6 (Figure 9) and a high reaction quotient, K_p, greater than 30 (Figure 10) also indicate high water-gas shift activity.

$$K_p = \frac{[H_2][CO_2]}{[H_2O][CO]}$$

Despite the high potassium loading, the activity and selectivity of the 100Fe/6.0Cu/6.1K/370MgAl₂O₄ catalyst are typical of our base-line, low-alpha precipitated catalysts (Table 3) with much lower K:Fe ratio (100Fe/2.2Si/xCu/0.7K, x=0 or 3.0, parts per weight). This implies that a substantial amount of the potassium in the 100Fe/6.0Cu/6.1K/370MgAl₂O₄ catalyst is neutralized by the support. A new catalyst batch with a higher iron loading and higher K:Fe ratio has recently been prepared (100Fe/6.0Cu/8.0K/260MgAl₂O₄). The activity and selectivity of this catalyst

will be reported in the next quarter. In addition, similar catalysts will be prepared with Al_2O_3 and ZnAl_2O_4 supports.

D.2.2. Slurry Bubble Column Reactor

The third run of the SBCR was successfully complete. The catalyst (RLS 4.4 Si/150 cyclone) was slurried (20 wt.%) with the C_{30} startup oil and pretreated with CO plus H_2 ($\text{H}_2/\text{CO} = 0.7$) using a GHSV = 5.3 SL/h-gFe ($U_g = 35.7$ cm/sec.). The internal reactor temperature was ramped at a rate of 20°C/hr from 100°C to 270°C during the pretreatment step. The reactor was held at 270°C for 21 hours. Synthesis was done using a reactor temperature of 270°C , 175 psig, $\text{H}_2/\text{CO} = 0.7$ and a GHSV = 5.3 SL/h-gFe ($U_g = 3$ cm/sec.). The conversions obtained during this run are shown in Figure 11. The initial conversions increased to a maximum of 53% and then over the next 80 hours decreased to around 35%. The conversions decreased slowly until the scheduled shutdown after 310 hours of operation.

Figure 12 shows the gas, H_2O and oil + wax product distributions during the run. The SBCR data shows a better selectivity when compared to the data obtained in the CSTR using the same catalysts, pretreatment and synthesis conditions. Significantly less gas is produced and significantly more oil + wax and H_2O are produced in the SBCR. Figures 13-16 show the product distributions in more detail. The mass balances were within the range of 95-102%.

D.3. Determine Stable Phases that Exist During Synthesis at High and Low CO

Conversion Levels

No scheduled or further activity to report.

D.4. Obtain Data on Rates Involved in the Interconversion of Iron Oxide and Carbide

No scheduled or additional activity to report.

D.5. Schedule of Activities for Next Quarter

- Continue the evaluation of the wax/catalyst separation.
- Continue to make additional supported Fe catalysts and test their activity, selectivity and productivity rate.
- Make an addition run in the CAER SBCR with a high-alpha catalyst.
- We will continue to study the deactivation of the catalysts to determine if this deactivation effect is general or catalyst specific.

Table 1

Surface Area and Pore Volume of MgAl_2O_4 Following
Different Calcination Procedures

Calcination Temp., °C	Surface Area, m^2g^{-1}	Pore Volume, cm^3g^{-1}
500	153	0.22
600	122	0.21
500	104	0.21
800	110	0.23
400 for 16 h/800 for 16 h	85	0.23

Table 2

Comparison of XRD Reflections for α -Fe₂O₃ and
100Fe/6.0Cu/6.1K/370MgAl₂O₄

d (Å)/ α -Fe ₂ O ₃ ^a	d (Å) 100Fe/6.0Cu/6.1K/370MgAl ₂ O ₄	hkl
3.686	3.682	012
2.703	2.702	104
2.519	2.518	110
2.208	2.209	113
1.8428	1.841	202
1.6966	1.695	116
1.4873	1.484	214

a. JCPDS, 24-72.

Table 3

Comparison of Activity and Selectivity of 100Fe/6.0Cu/6.1K/370MgAl₂O₄
Catalyst and 100Fe/2.2Si/3.0Cu/0.7K Catalyst

Catalyst	Fe/Cu/K/MgAl ₂ O ₄	100Fe/4.4Si/2.6Cu/1.0K
Time of Synthesis	284	670
Conversion/%		
CO	79	84
H ₂	67	77
CO + H ₂	74	81
HC Production		
Total/ g/m ³ syngas	147	157
C ₃ +/ g/m ³ syngas	119	124
g h ⁻¹ g-Fe ⁻¹	0.45	0.46
WGS		
K _p ^a	32	36
CO ₂ selectivity/C basis %	0.49	0.50
H ₂ :CO usage ratio	0.60	0.64
Selectivity/wt. %		
CH ₄	11.9	13.7
C ₂ -C ₄ (alkene wt. %)	30.3 (62)	32.8 (54)
C ₅ -C ₁₁ (alkene wt. %)	46.6 (57)	46.2 (53)
C ₁₂ +	11.2	7.2
$a. K_p = \frac{[H_2][CO_2]}{[H_2O][CO]}$		

Figure 1. Scanning electron microscopy micrograph of a 30 wt% iron on alumina catalyst following 452 h of FTS in a CSTR.



Figure 2. X-ray diffraction results for Mg-Al hydroxides following calcination at (a) 500 °C, (b) 600 °C, (c) 700 °C and (d) 800 °C for 4 h and (e) 400 °C for 16 h followed by 800 °C for 16 h. Cu K α =1.5418 Å.

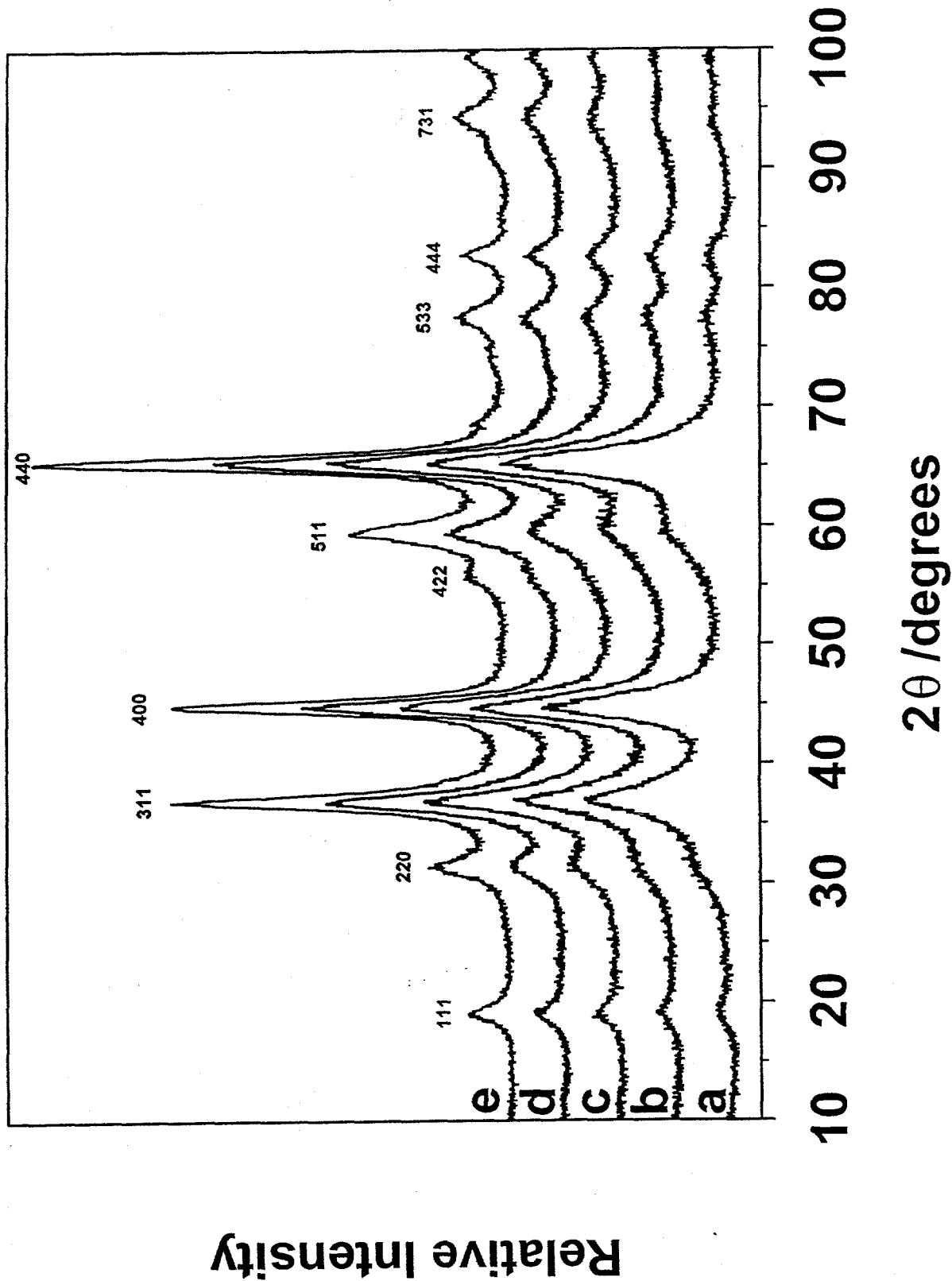


Figure 3. X-ray diffraction results for (a) MgAl_2O_4 and (b) $100\text{Fe}/6.0\text{Cu}/6.1\text{K}/370\text{MgAl}_2\text{O}_4$, where the most intense $\alpha\text{-Fe}_2\text{O}_3$ reflections are labeled.

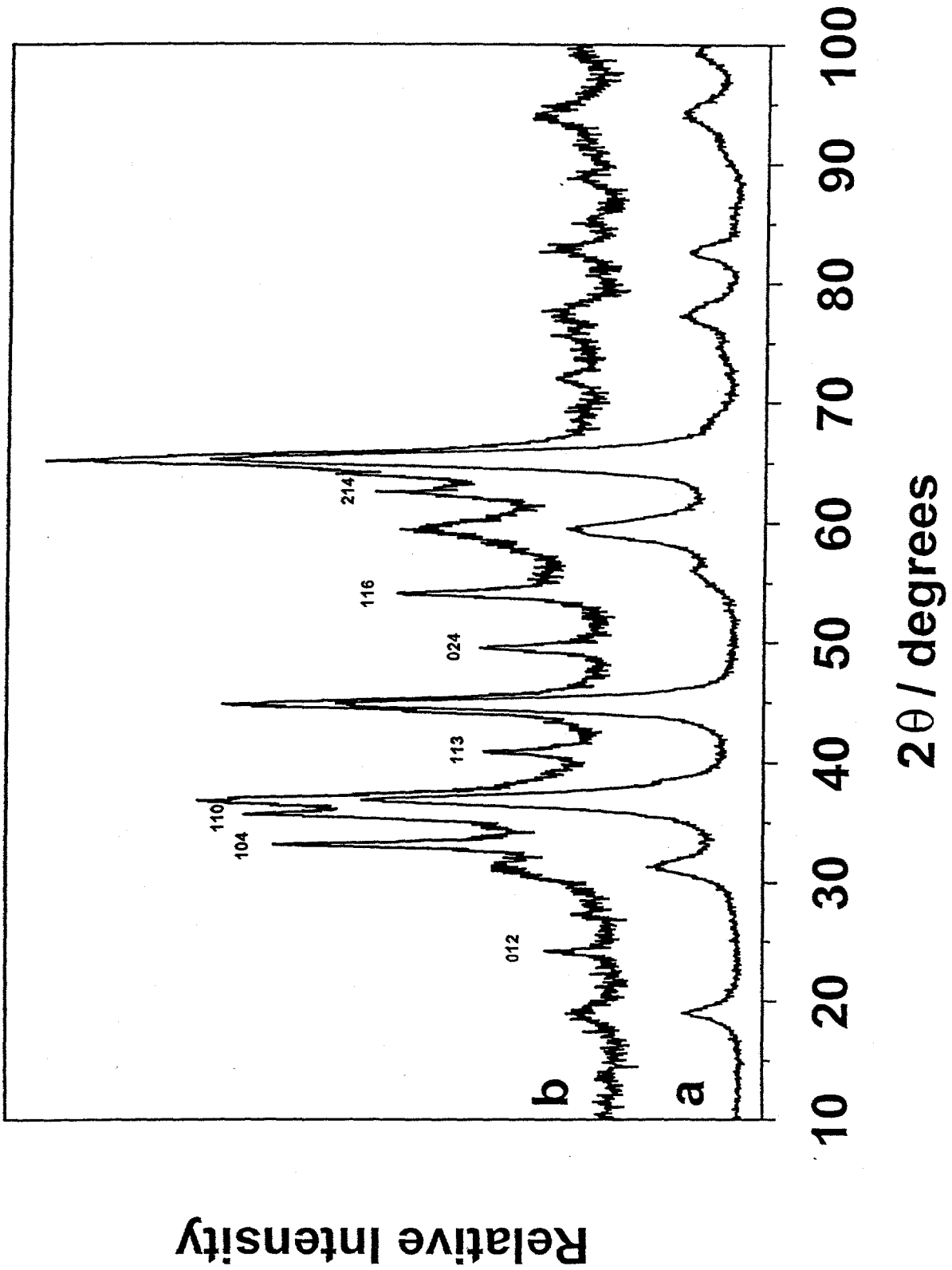


Figure 4. CO conversion (O), H₂ conversion (□) and CO+H₂ conversion (◇) as a function of time on stream for the 100Fe/6.0Cu/6.1K/370MgAl₂O₄ catalyst. FTS conditions: 270 °C, 1.31MPa, H₂:CO=0.7, 3.1 NL h⁻¹ g-Fe⁻¹.

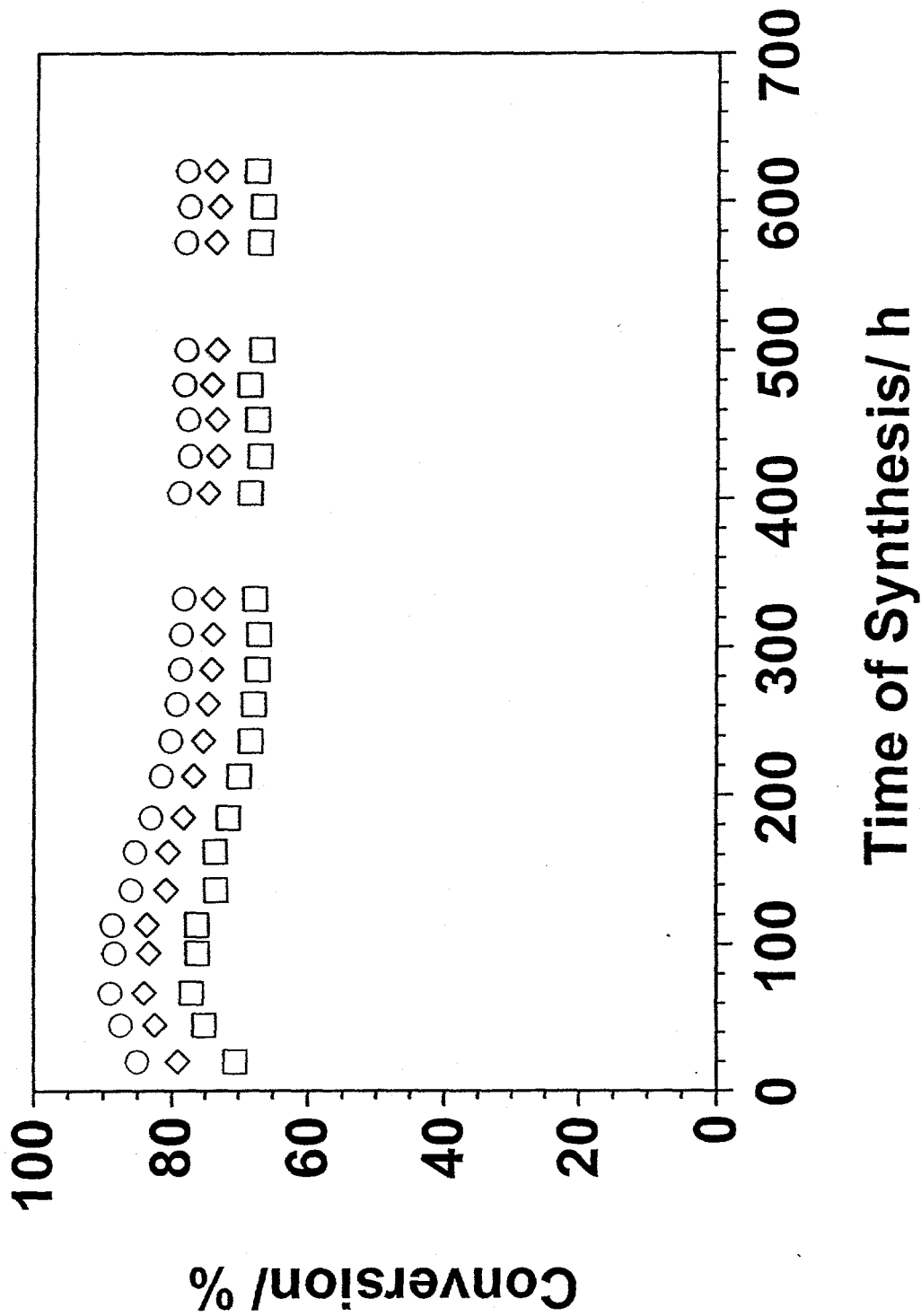


Figure 5. Hydrocarbon production for the 100Fe/6.0Cu/6.1K/370MgAl₂O₄ catalyst as a function of time on stream. Total hydrocarbon (O) and C₃+ hydrocarbon (□), g m⁻³ of syngas feed. FTS conditions: 270 °C, 1.31MPa, H₂:CO=0.7, 3.1 NL h⁻¹ g-Fe⁻¹.

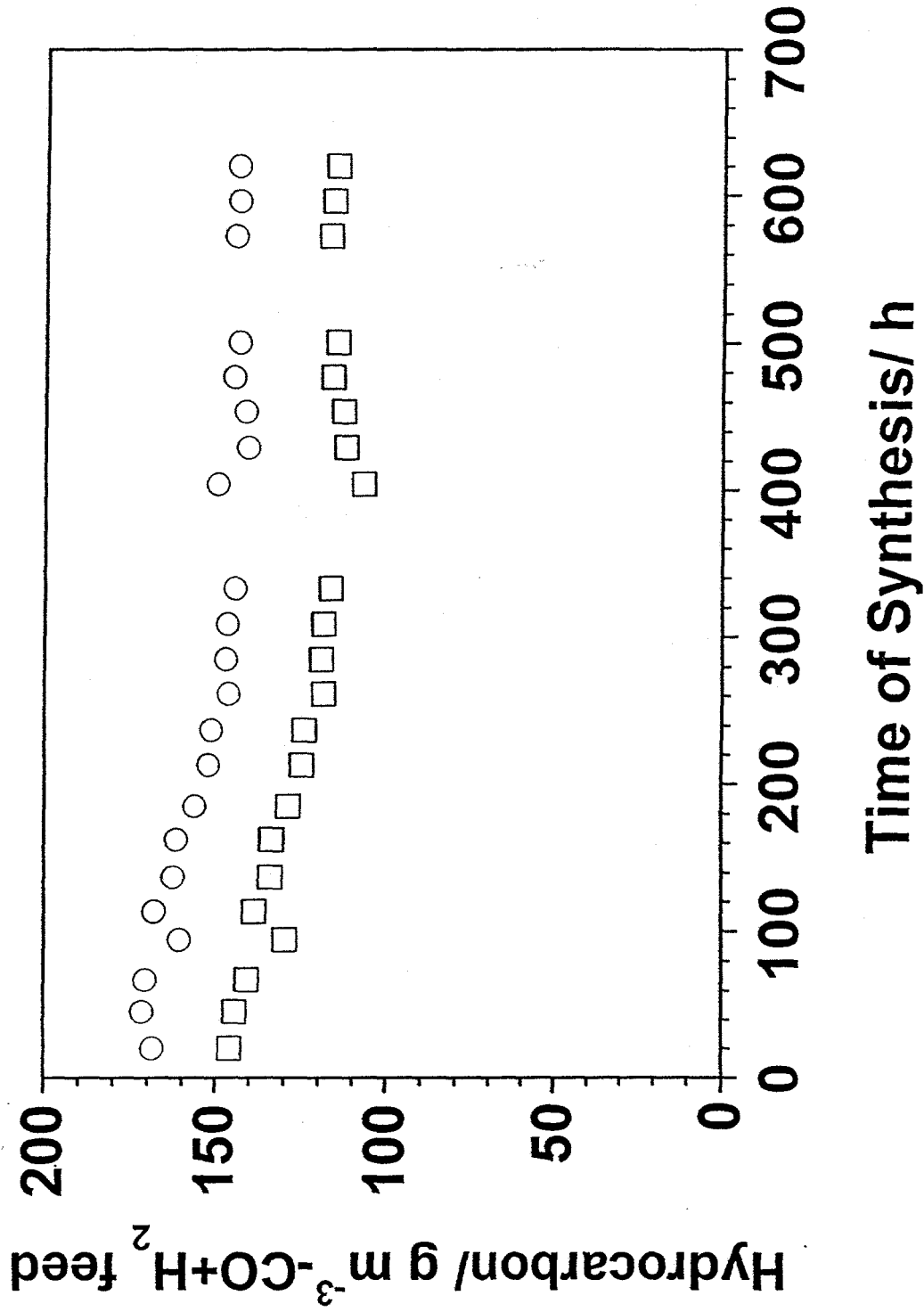


Figure 6. FTS activity as a function of time on stream, $\text{g h}^{-1} \text{g-Fe}^{-1}$. FTS conditions: 270°C , 1.31MPa , $\text{H}_2:\text{CO}=0.7$, $3.1 \text{NL h}^{-1} \text{g-Fe}^{-1}$.

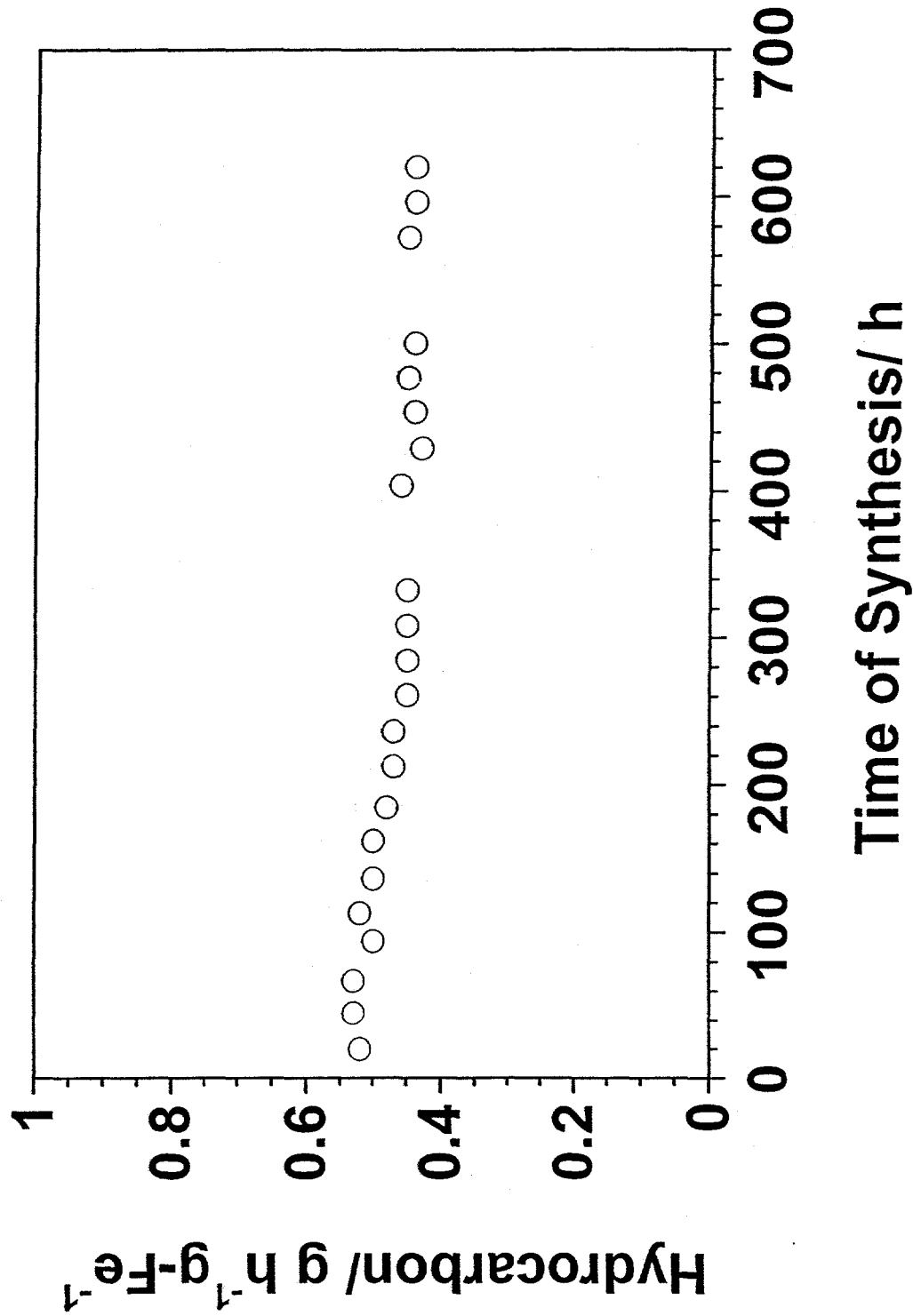


Figure 7. Methane selectivity (O) and methane and ethane selectivity (\square) as a function of time on stream. Selectivity is presented as percent of carbon which is converted to hydrocarbons. FTS conditions: 270°C , 1.31MPa, $\text{H}_2:\text{CO}=0.7$, 3.1 NL h^{-1}

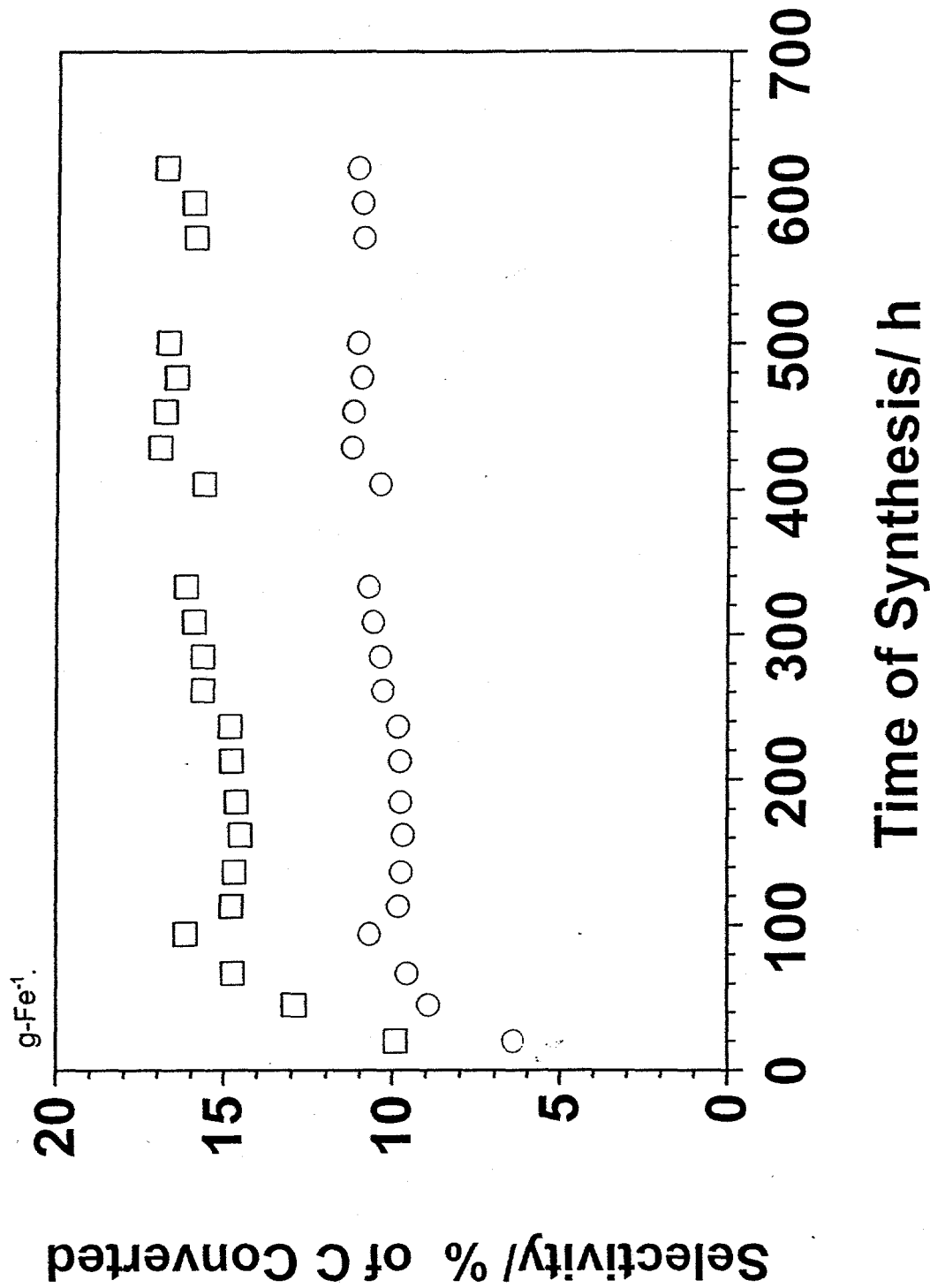


Figure 8. Alkane fraction of (O) C₂, (□) C₃ and (◇) C₄ products as a function of time on stream. Fraction of 1-butene (△) in the C₄ linear alkenes as a function of time on stream. FTS conditions: 270 °C, 1.31MPa, H₂:CO=0.7, 3.1 NL h⁻¹ g-Fe⁻¹.

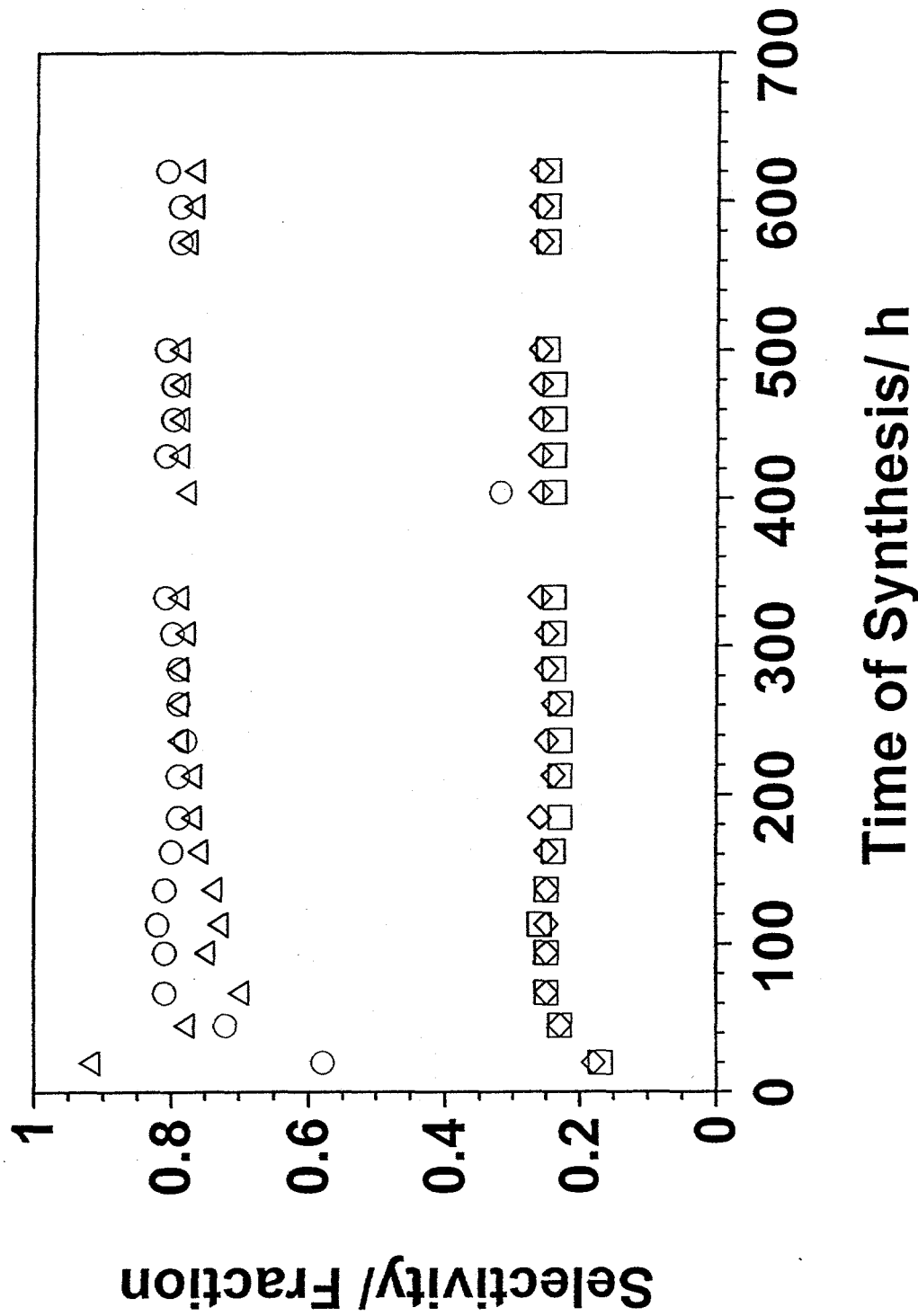


Figure 9. Percent of carbon converted to carbon dioxide (O) and H₂:CO usage ratio

(□) as a function of time on stream. FTS conditions: 270 °C, 1.31MPa, H₂:CO=0.7, 3.1

NL h⁻¹ g-Fe⁻¹.

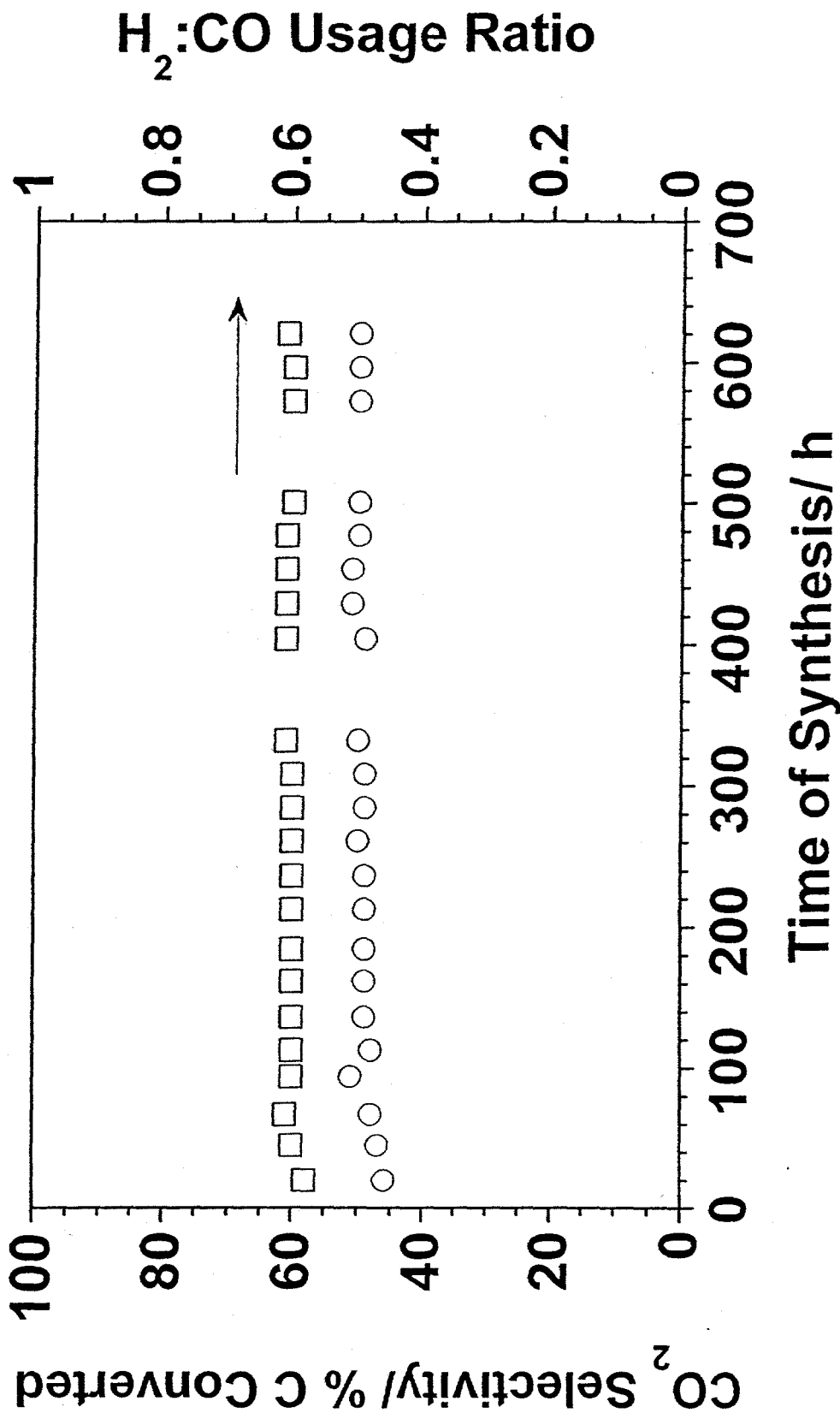


Figure 10. Water-gas shift reaction quotient, $K_p = \frac{[H_2][CO_2]}{[H_2O][CO]}$, as a function of time on stream. FTS conditions: 270°C, 1.31MPa, $H_2:CO=0.7$, 3.1 NL h^{-1} g-Fe⁻¹.

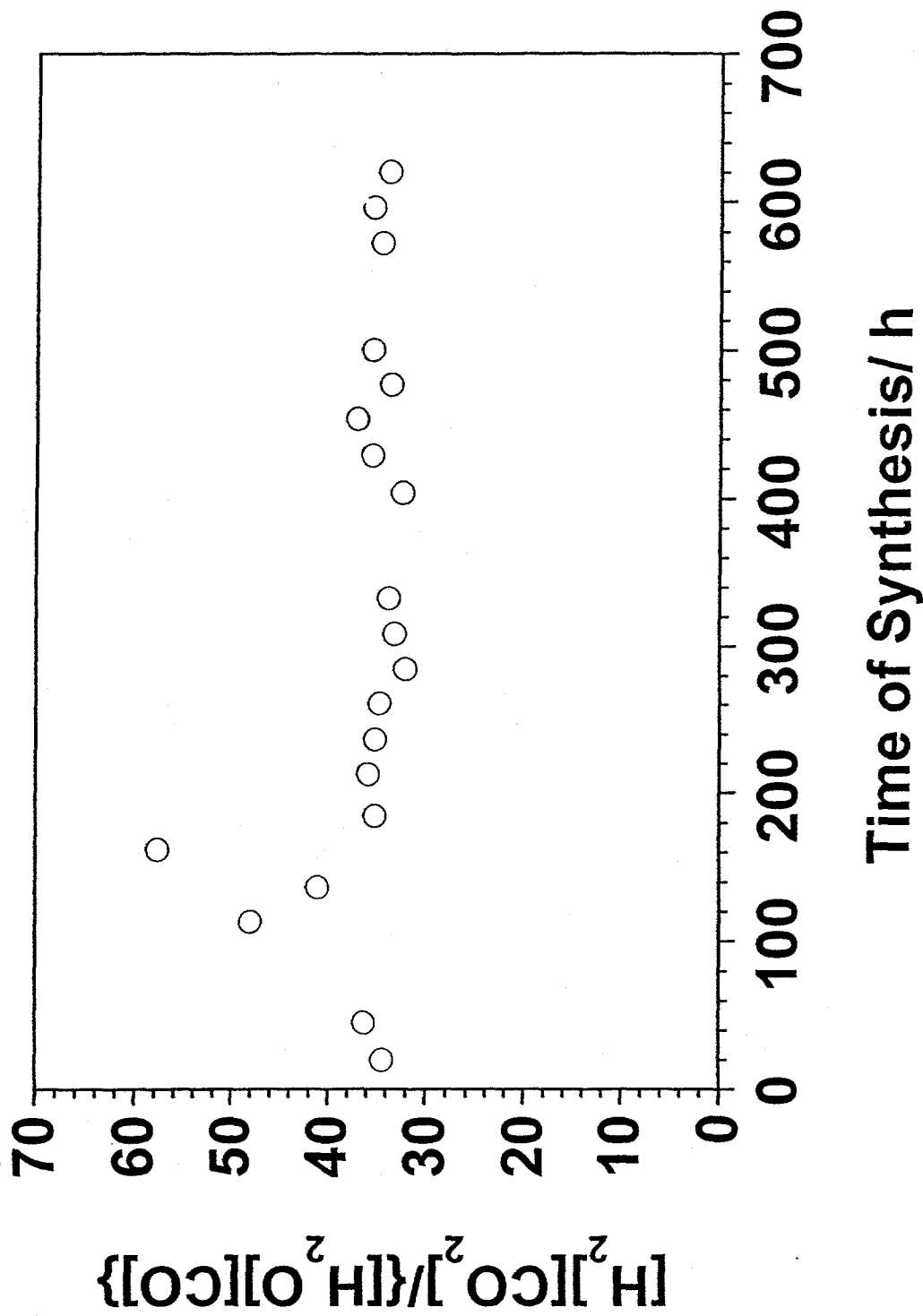


Figure 11.
SBCR503 Conversion vs Syn. Time

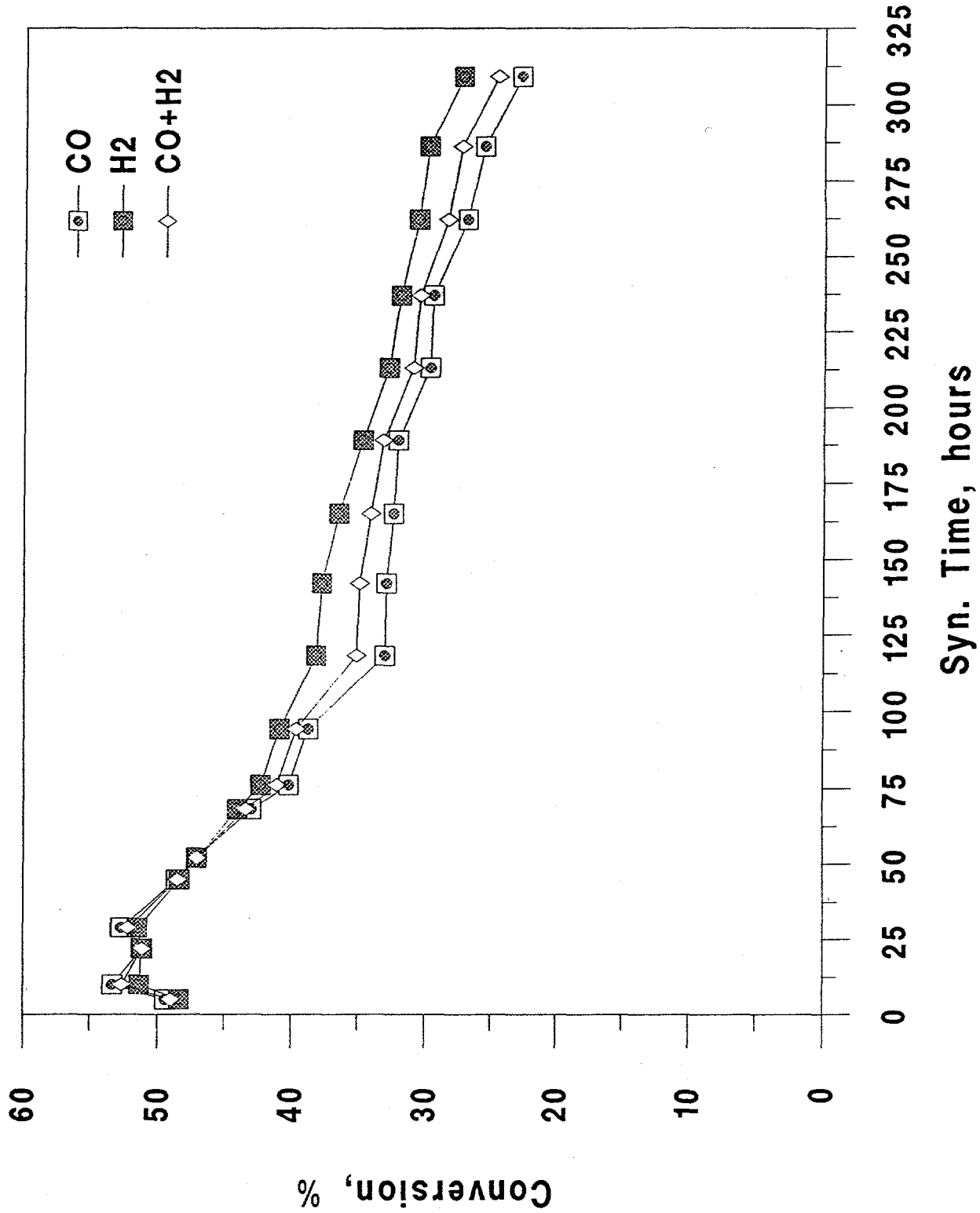


Figure 12.
 Products vs Syn. Time (SBCR-503)

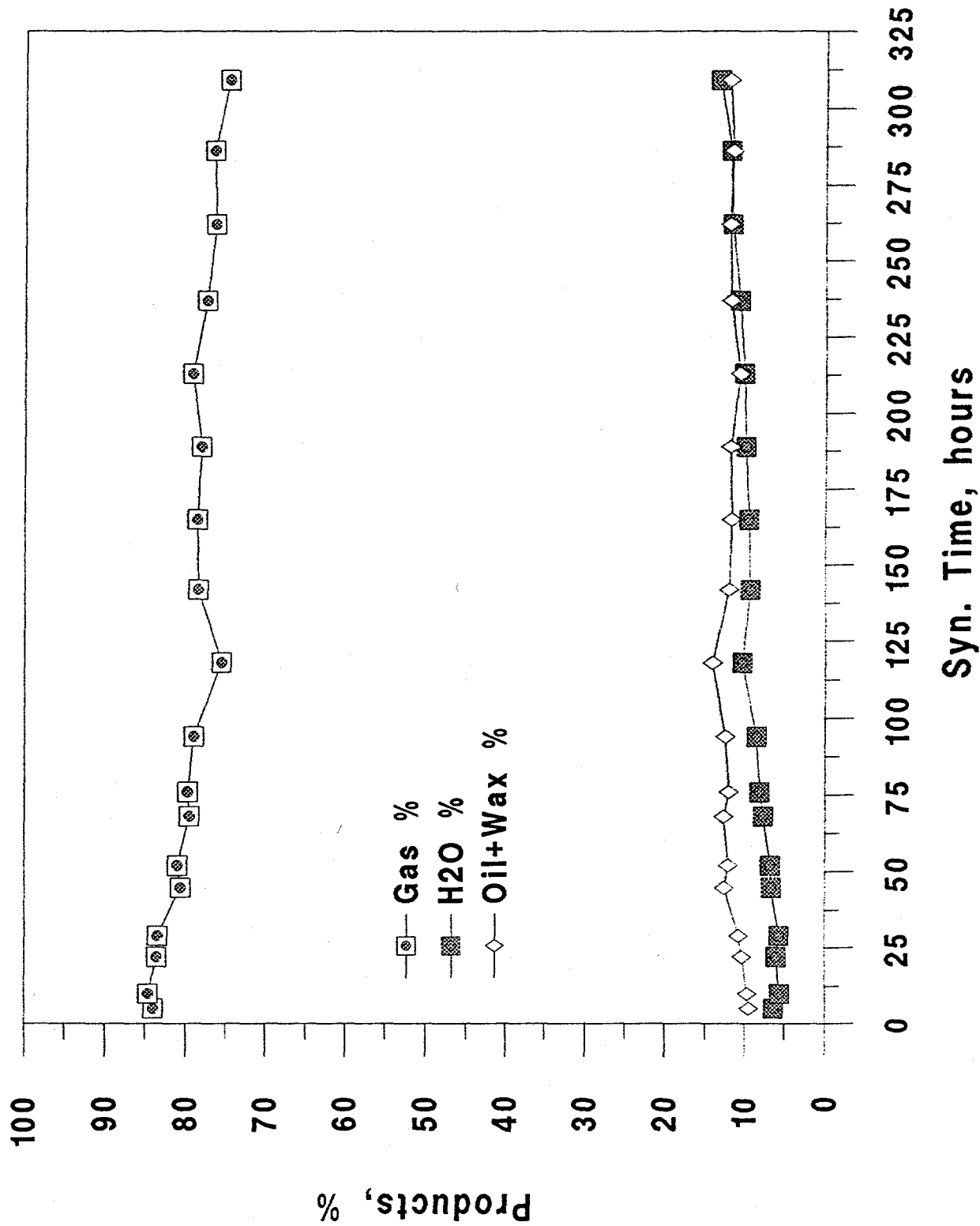


Figure 13.
CO₂ Mole%, CH₄% and CO₂/CO ratio vs Syn. Time
(SBCR-503)

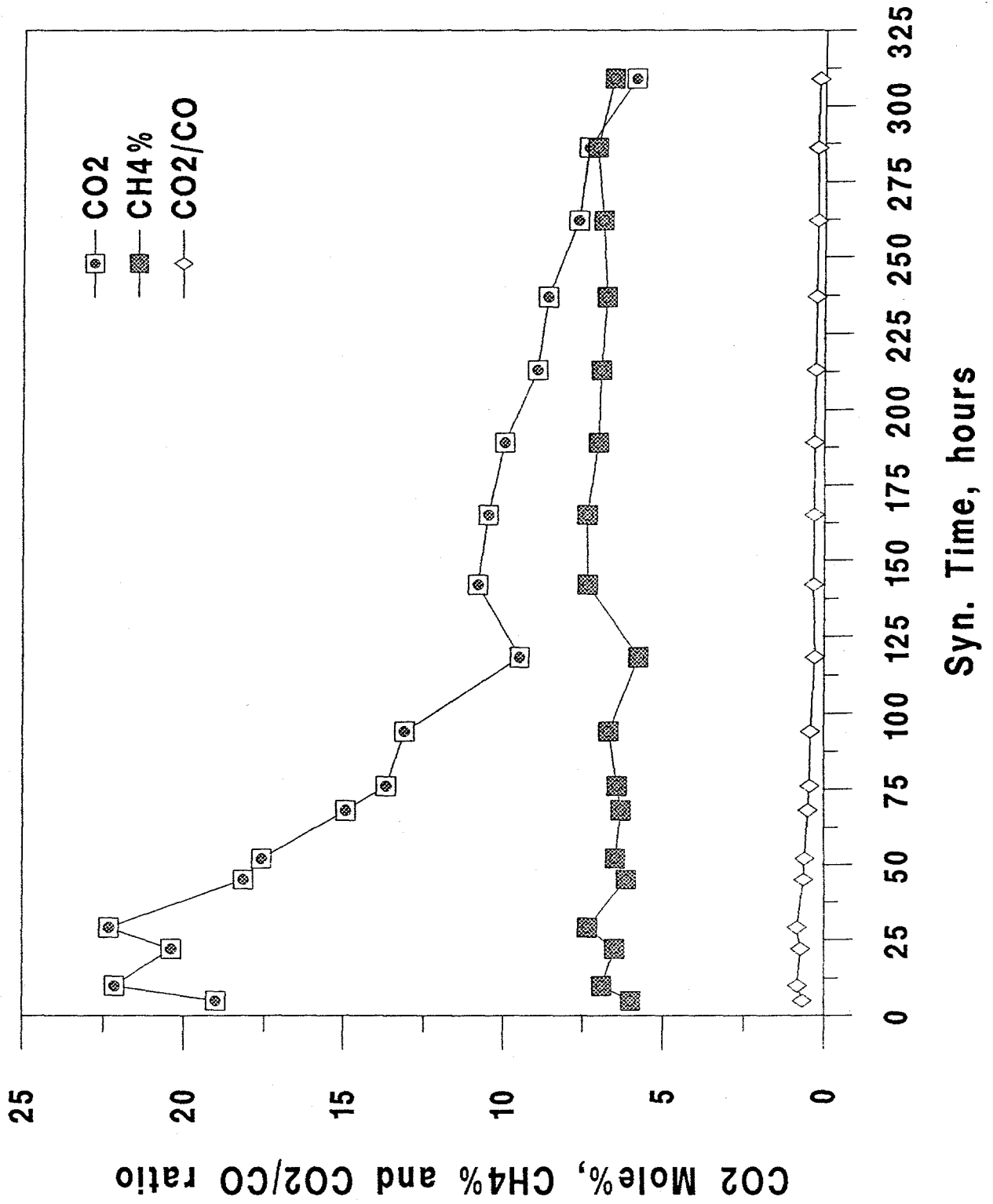


Figure 14.

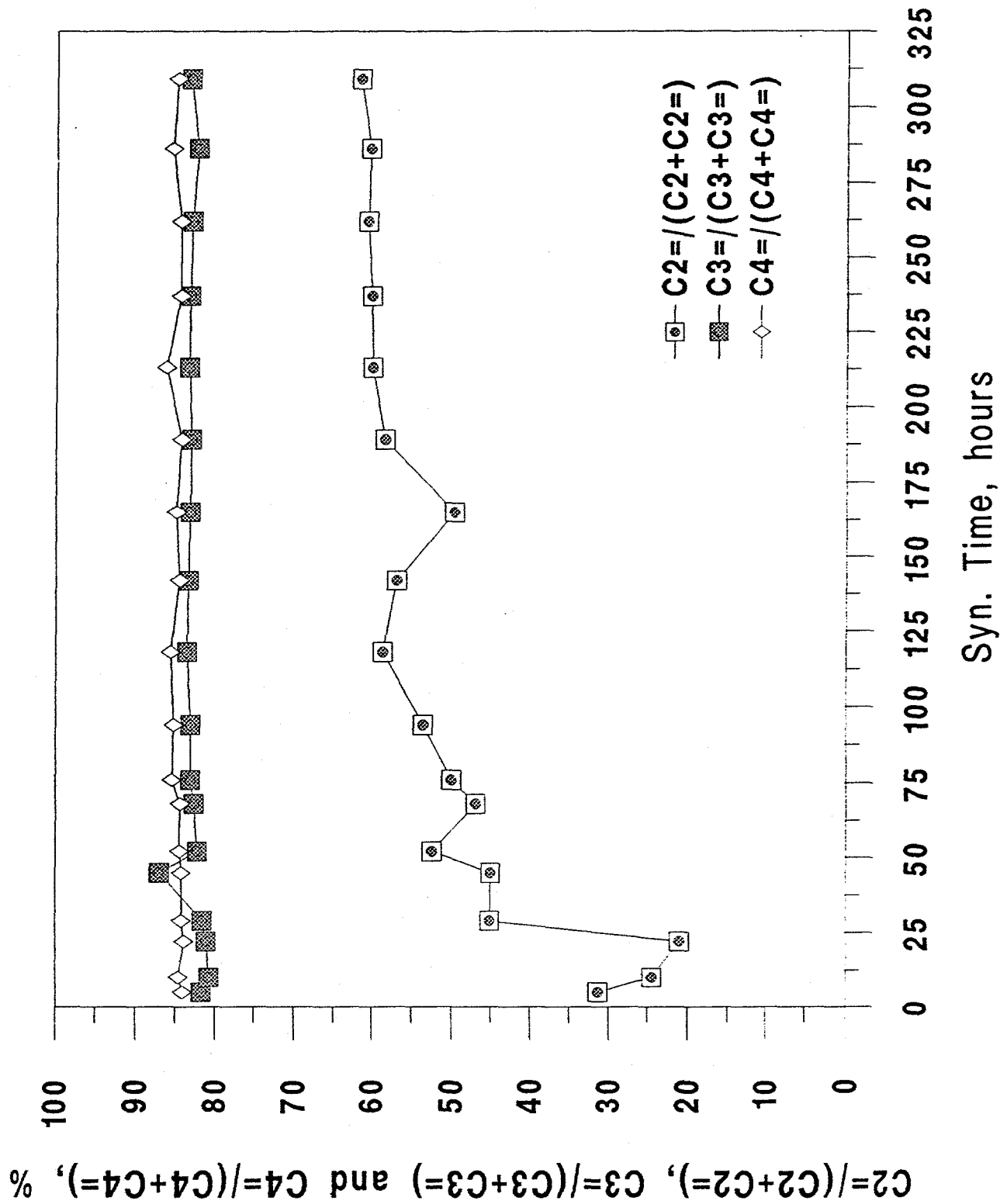


Figure 15.
Oxygenate in Aqueous vs Syn. Time
(SBCR-503)

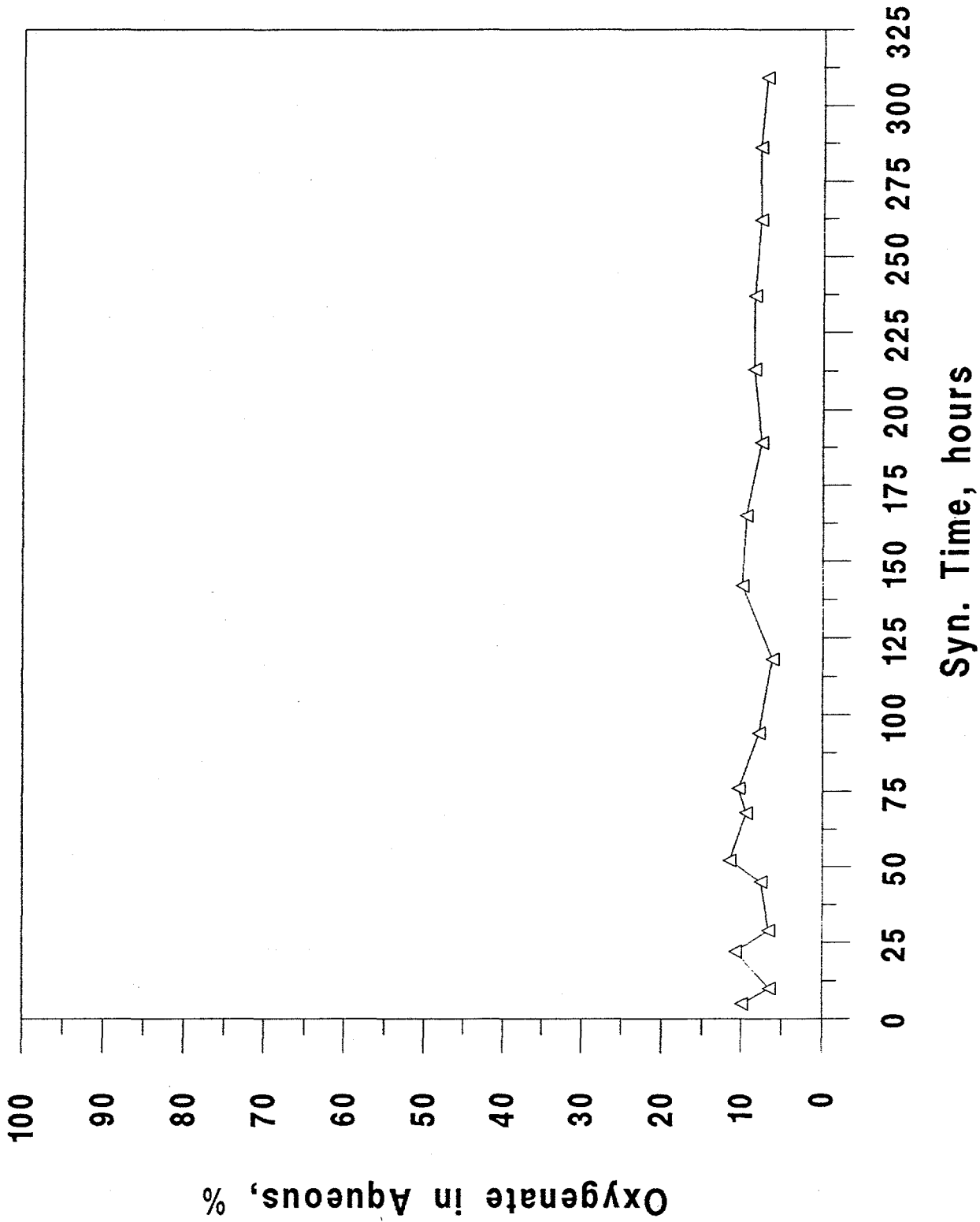


Figure 16. Alpha Plot for SBCR 503.

