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AFFDL-TR-66-56

CATALYTIC OXIDATION OF METHANE AT LOW SPACE VELOCITIES

DERRY W. MARSHALL, 1/LT., USAF

TECHNICAL REPORT AFFDL-TR-66-56

JUNE 1966

This document is subject to special export controls and each transmittal to foreign governments or foreign nationals may be made only with prior approval of the Air Force Flight Dynamics Laboratory (FDFE), Wright-Patterson Air Force Base, Ohio 45433.

AIR FORCE FLIGHT DYNAMICS LABORATORY
RESEARCH AND TECHNOLOGY DIVISION
AIR FORCE SYSTEMS COMMAND
WRIGHT-PATTERSON AIR FORCE BASE, OHIO

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FOREWORD

This report was prepared by the Vehicle Equipment Division, Air Force Flight Dynamics Laboratory. The work was performed under Project No. 6146, "Atmosphere and Thermal Control," Task No. 614613, "Trace Contaminant Oxidation Removal and Effects," and concerns the catalytic oxidation of methane to carbon dioxide and water. The work was administered by the Air Force Flight Dynamics Laboratory, Research and Technology Division, Air Force Systems Command, Wright-Patterson Air Force Base, Ohio, the Chief Investigator being D. W. Marshall, 1/Lt, USAF, with assistance from Messrs. K. Wess and A. Civetz.

This report covers work conducted from January 1965 through September 1965. The manuscript was released by the author in April 1966 for publication as an RTD technical report.

The distribution of this report is limited because it covers an area of technology that is embargoed under the Department of State International Traffic in Arms Regulations and U. S. Export Control Act of 1949.

This Technical report has been reviewed and is approved.

William C Savage
W. C. SAVAGE
Chief, Environmental Control Branch
Vehicle Equipment Division
Air Force Flight Dynamics Laboratory

ABSTRACT

The feasibility of removing methane from an airstream by catalytic oxidation at low flows was investigated by passing a one percent, by volume, methane in airstream through an in-house designed and fabricated stainless steel reactor. The reactor contained either Hopcalite, 5% V_2O_5 - 5% MoO_3 on alumina, or one of the following materials supported on alumina: 0.5% palladium, 0.5% platinum, 0.5% rhodium, and 0.5% ruthenium. Some discontinuity in the data reported in the literature on methane oxidation at or around the transition flow region prompted a closer investigation in this area. In addition, space velocities around 500 fills per hour were used to insure chemical equilibrium and for comparison with the more common value of approximately 15,000 fills per hour used in most industrial applications. Experiments performed using the above catalysts revealed no deleterious effects associated with operating in the transition flow region, but did point out the advantage of using high space velocities for maximum catalyst utilization. The palladium catalyst proved superior to all others tested and catalyzed the reaction to completion at 560°F. Conversion versus temperature curves are plotted for each catalyst flow rate combination and the effect of space velocity on conversion at constant temperature is shown for the Hopcalite and palladium catalysts. Variables which affected the accuracy of experimentation most are discussed and possible improvements suggested.

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SECTION I

INTRODUCTION

This report summarizes an in-house research program under general effort in the area of "Catalytic Trace Contaminant Oxidation." The primary objective of this program is to determine the feasibility of removing methane from the atmosphere by direct catalytic oxidation. A suitable catalytic oxidation process will complement the charcoal filters which do not satisfactorily adsorb gases of such low molecular weight. The oxidation testing is accomplished by passing the methane containing gas stream through a heated catalytic reactor and by gas analysis determining what percentage is oxidized. The degree of oxidation which takes place depends upon the catalyst material, the temperature, the flow rate through the catalyst bed and to a lesser extent the absolute pressure in the reactor.

Johns, Garwig, and Markels,* using a space velocity of 17,000 fills per hour, experienced some discontinuity among differential reaction rates measured at a Reynolds number of 50 and considered the transition flow effects to be of sufficient importance to justify redesigning the reactor. For this reason a closer look was taken at methane oxidation using flow rates in the transition region. Relatively low space velocities were used (500 fills per hour) with each catalyst for the purpose of establishing data to which other investigations can be compared. Three different space velocities were used with the Hopcalite and palladium catalysts to determine the effect of flow rate on conversion. The eventual purpose of this work, then, is to produce ideas which will be helpful in determining design parameters for a space oriented catalytic burner.

---

*Johns, Richard R., Garwig, Paul L., and Markels, Michael, Jr., Catalytic Combustion of Atmospheric Contaminants in Space Vehicle Atmospheres, AFFDL-TR-65-46, Air Force Flight Dynamics Laboratory, Wright-Patterson Air Force Base, Ohio, Contract No. AF 33(615)-1019, Atlantic Research Corporation, May 1965.

SECTION II

EXPERIMENTAL PROGRAM

DESCRIPTION OF TEST SETUP

A diagram showing the flow of gas through the experimental test setup is presented in Figure 1. A one percent by volume methane in air mixture was prepared by metering 100 percent methane and bottled air together in the proper proportions. Flowmeters calibrated at five psig served as visual proof that the correct flows were being maintained. The gas mixture then passed into the preheater tube (Figure 2) which is concentric to and surrounded by the catalyst. Five thermocouple wires lie inside 3/16 inch OD tubes which run parallel to the reactor wall and then penetrate both the wall and catalyst bed. A threaded cap on one end of the reactor facilitated changing of catalyst and inspection. Although the reaction involved is exothermic, large heat losses from the reactor ends made it necessary to continue heating the reactor after start-up. This heat was furnished by an asbestos embedded nichrome wire operating at 110 VAC and from zero to fifteen amperes. To prepare the product stream for the hydrogen flame detector, a drying tube ($CaSO_4$) and an air cooled heat exchanger (coiled copper tubing) were used. Figures 3 and 4 show, pictorially, the overall setup and the reactor. The reactor system was operated inside a hood to prevent any possible build-up of methane.

FLOW CALCULATIONS

In order to compare conversion efficiencies of given catalysts at various flow rates a modified Reynolds number was computed using the following expressions (Reference 1):

$$N_{Re} = \frac{D_p G}{\mu(1-\epsilon)} \quad \text{or since } G = V_o \rho$$

$$N_{Re} = \frac{D_p V_o \rho}{\mu(1-\epsilon)}$$

where

D_p = diameter of catalyst particle in terms of an equivalent volume sphere (ft)

V_o = velocity thru empty reactor (ft/sec)

ρ = density of gas (lbs/ft^3)

μ = viscosity of gas (lb_f/ft-sec)

G = mass velocity (lb/ft^2-sec)

ϵ = catalyst bed porosity

Catalyst bed dimensions are

L = 11.0 in. (length)

r_1 = 1.25 in. (outside radius)

r_2 = 0.25 in. (inside radius)

volume $= \pi (r_1^2 - r_2^2) L = 52.0 \text{ in.}^3$

cross-section area $= \pi (r_1^2 - r_2^2) = 4.71 \text{ in.}^2$

$A = 0.0327 \text{ ft}^2$

Size of cylindrical catalyst particle is

diameter $= \frac{1}{8}$ in.

length $= \frac{1}{8}$ in.

volume $= \pi r^2 L = \pi (\frac{1}{16})^2 (\frac{1}{8}) = 0.00153 \text{ in}^3$

where

r_s = radius of equivalent volume sphere

r_s = 0.0715 in.

therefore

$$D_p = 0.143 \text{ in.} = 0.0119 \text{ ft}$$

Now let F = flow rate of gas mixture (99% air - 1% CH_4) cc/min

$$V_o = (F \frac{cc}{min})(\frac{min}{60 sec})(\frac{1 ft^3}{28,317 cc})(\frac{1}{0.0327 ft^2})$$

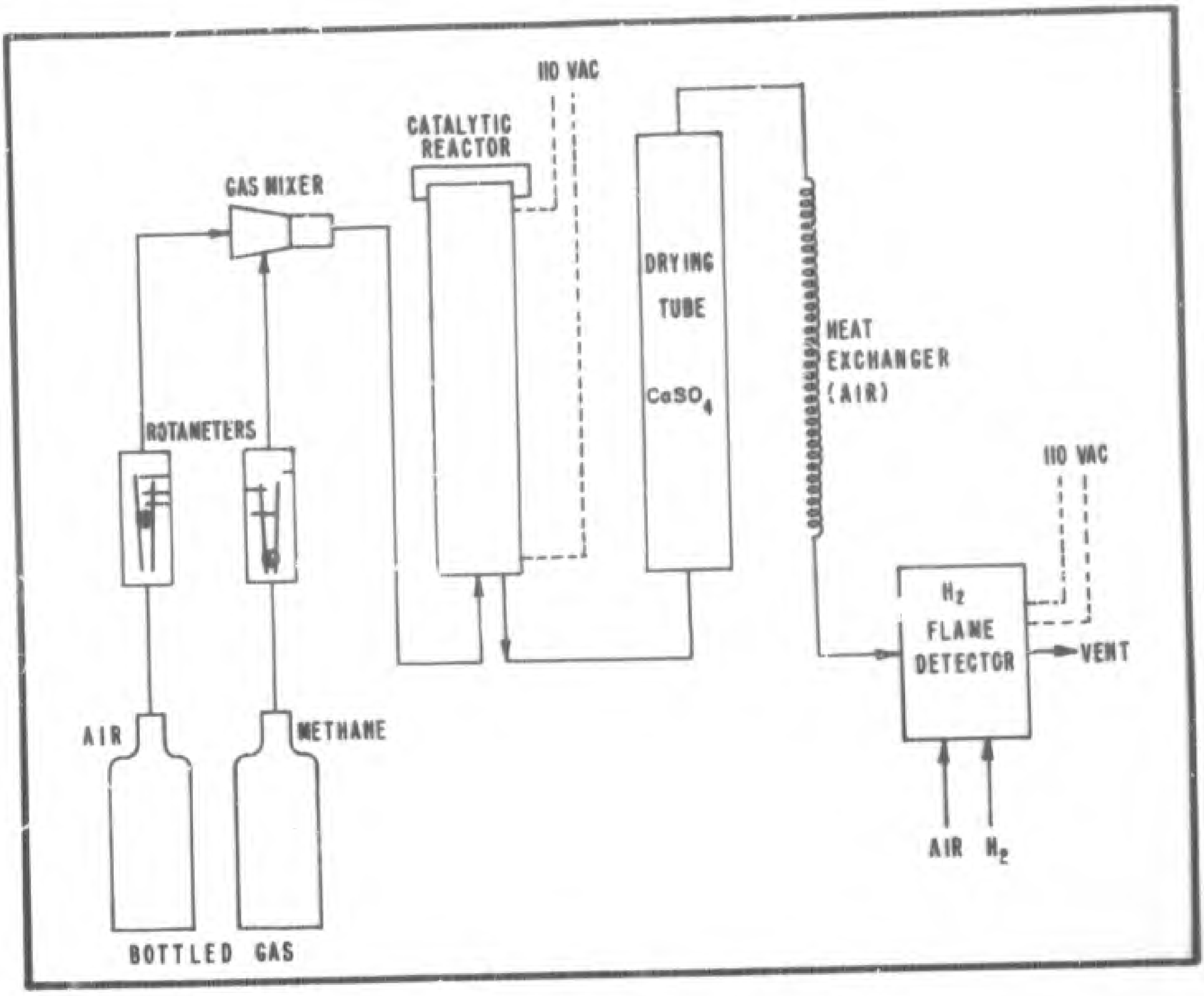

Figure 1. Flow Diagram

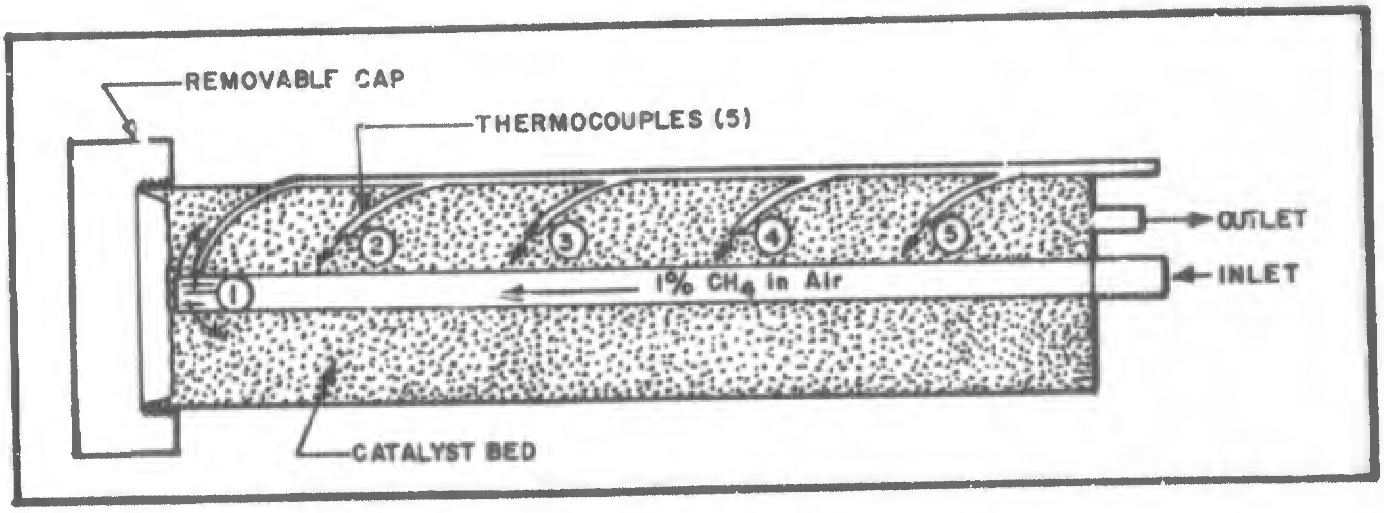

Figure 2. Reactor Schematic

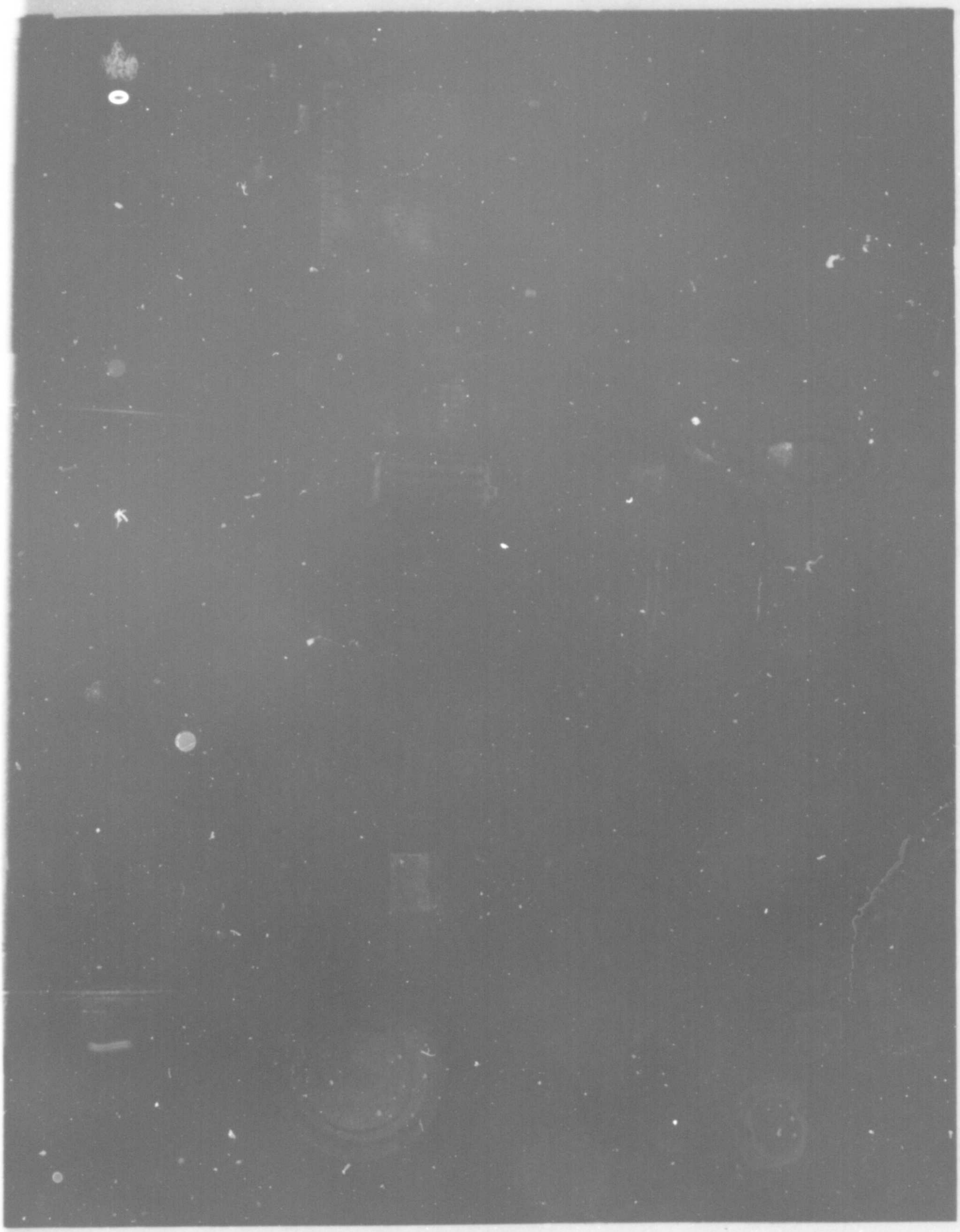

Figure 3. Overall System

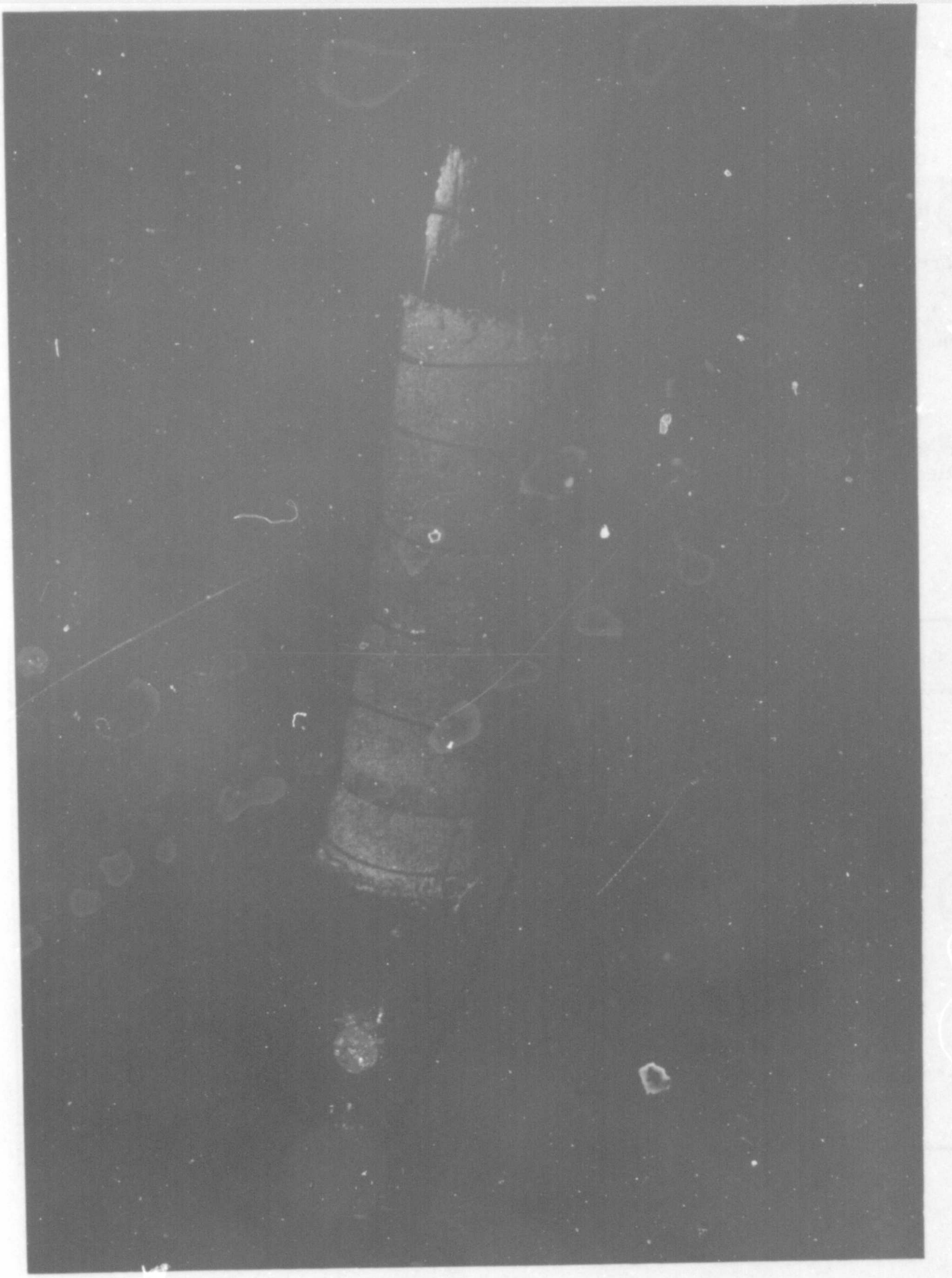

Figure 4. Reactor

$V_0 = 0.18 \times 10^{-4} F \text{ ft/sec}$

$\rho = 0.075 \text{ lb/ft}^3 \text{ (air)}$

$\epsilon = 0.68$

$\mu = 0.018 \text{ cp or } (0.018 \text{cp})(6.72 \times 10^{-4} \left(\frac{\text{lb}_f/\text{ft-sec}}{\text{cp}}\right)$

$\mu = 0.121 \times 10^{-4} \text{ lb}_f/\text{ft-sec (air)}$

Therefore

$$N_{Re} = \frac{(0.0119 \text{ ft})(0.18 \times 10^{-4} \frac{\text{ft}}{\text{sec}}) F (0.075 \frac{\text{lb}}{\text{ft}^3})}{(0.121 \times 10^{-4} \frac{\text{lb}_f}{\text{ft-sec}})(1 - 0.68)}$$

$$= 0.00415 F$$

For simplicity this linear relationship is plotted in Figure 5.

In order to calculate space velocities the porosity of the catalysts was obtained by both water displacement and powdered calcium carbonate displacement. The void fraction for all catalyst materials studied was found to be 0.68.

Therefore if

$\text{reactor volume} = 52.0 \text{ in.}^3 = 851 \text{ cc}$

$\text{void volume} = 579 \text{ cc}$

and at a flow rate of 4800 cc/min

$$\text{Space velocity} = \left(\frac{4800 \text{ cc/min}}{579 \text{ cc}}\right)\left(\frac{60 \text{ min}}{\text{hr}}\right) = 500 \text{ hr}^{-1}$$

DESCRIPTION OF CATALYSTS

Table I lists all catalysts evaluated during this program.

TABLE I

DESCRIPTION OF CATALYSTS

Code	Name/Composition	Shape and Size	Porosity
A.	0.5% Palladium on Alumina	1/8" cylindrical pellets	0.68
B.	0.5% Rhodium on Alumina	1/8" cylindrical pellets	0.68
C.	Hopcalite 60% CuO 40% MnO_2	4 - 12 mesh	0.68
D.	0.5% Platinum on Alumina	1/8" cylindrical pellets	0.68
E.	0.5% Ruthenium on Alumina	1/8" cylindrical pellets	0.68
F.	Vanadia Catalyst 5% V_2O_5 on Alumina 5% MoO_3 on Alumina	6 mesh	0.68
G.	0.2% Palladium on Granular Carbon	4 - 12 mesh	0.68

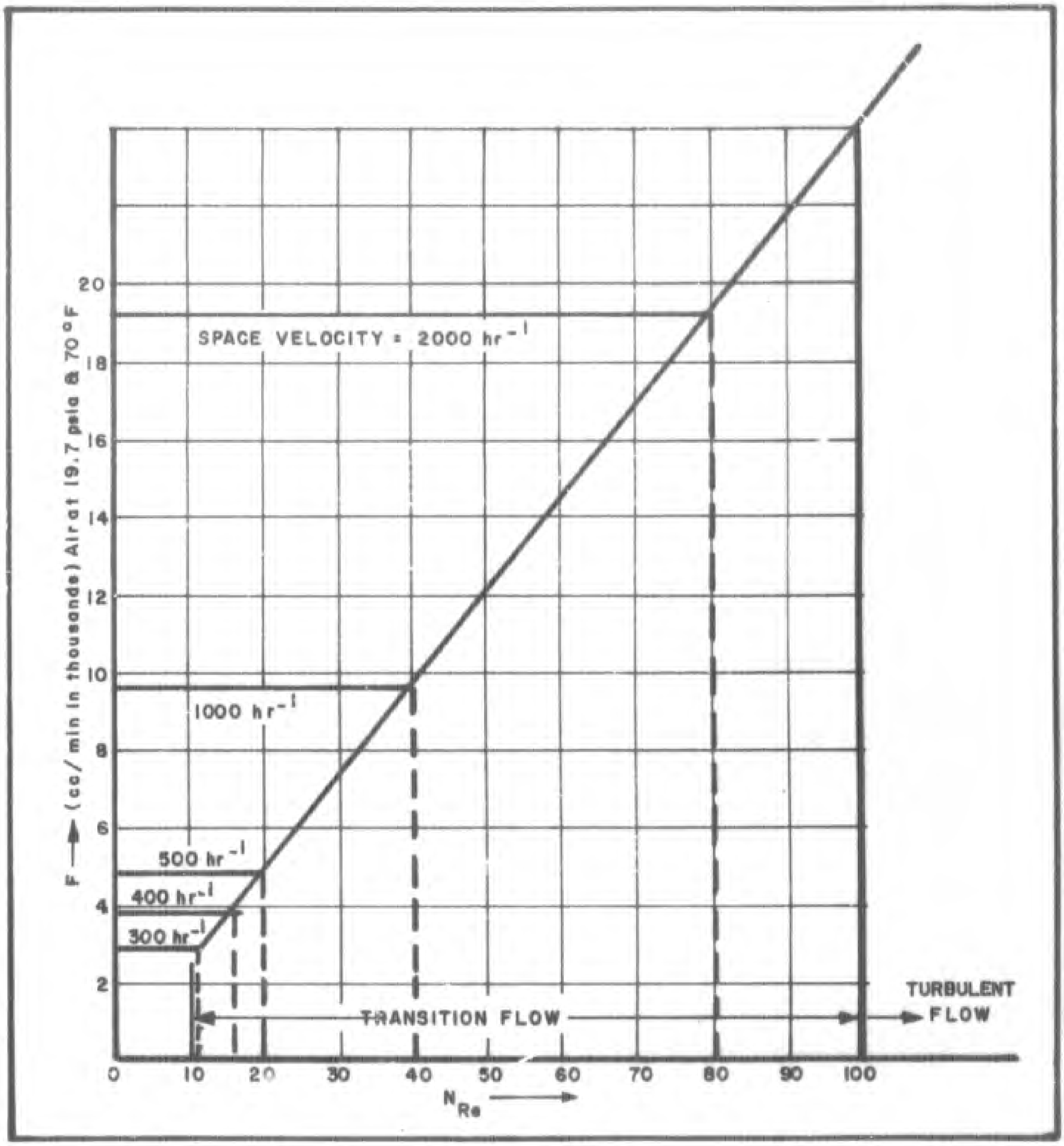

Figure 5. Reynolds Number vs Flow Rate

TESTING PROCEDURE

The following steps represent a typical outline for the testing of a single catalyst.

a. Determine catalyst porosity as previously explained.

b. Fill reactor with known weight of catalyst.

c. Determine flow rate necessary for desired operating space velocity.

d. Warm up and calibrate hydrogen flame detector (approximately 30 minutes required).

e. Set all flow rates at predetermined values.

f. Take meter readings on detector as temperature is elevated slowly from ambient to desired upper limit.

SECTION III

RESULTS

TABULATION OF EXPERIMENTAL DATA

Table II lists the numerical results of experiments performed during the program.

Figures 6, 7, and 8 show the complete temperature versus conversion curves for each of the above experiments. Raw data for these curves may be found in the Appendix. In all instances the reactant stream was composed of one percent methane in dry air.

Table III shows the effect of flow rate on conversion at constant temperature for the Hopcalite catalyst. These data were taken from Figure 7, and are discussed more fully in Section IV.

TABLE II

PERFORMANCE OF CATALYSTS

Catalyst*	Space Velocity (hr^{-1})	100% Conversion Temperature (°F)	Flow Rate (cc/min)
A	500	560	4800
A	1000	635	9600
A	2000	625	19,200
B	500	635	4800
C	300	625	2880
	400	650	3840
	500	665	4800
D	500	695	4800
E	500	800	4800
F	500	No Conversion	4800
G	500	No Conversion	4800

*See Table I for name and composition

TABLE III

EFFECT OF SPACE VELOCITY VARIATION OF HOPCALITE CATALYST

Space Velocity (hr^{-1})	Total Flow cc/min	CH_4 Flow cc/min	% Conversion at 550 °F	CH_4 Oxidized cc/min
300	2880	29	82	23.6
400	3840	38	65	25.0
500	4800	48	55	26.4

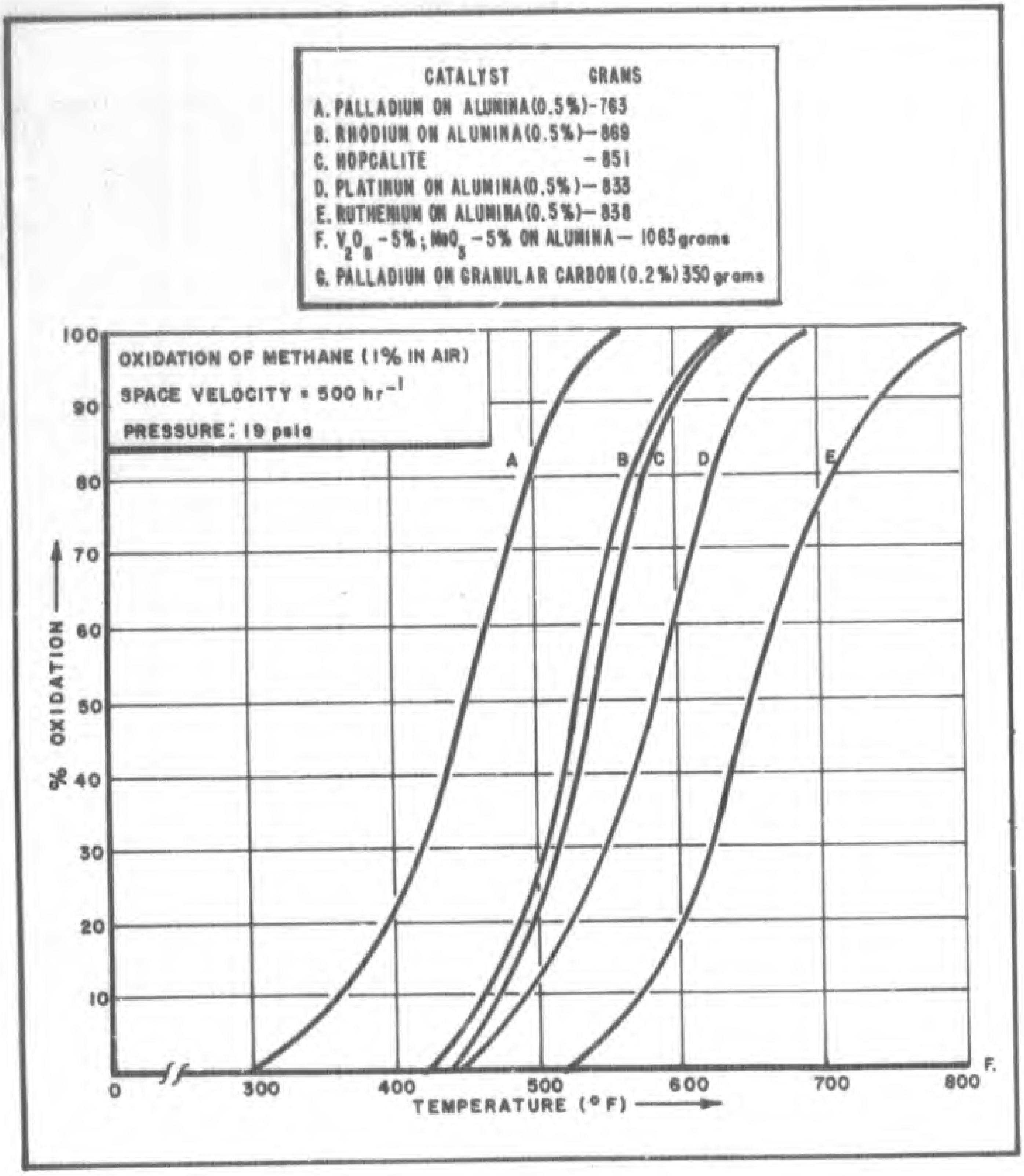

Figure 6. Temperature vs Conversion for all Catalysts

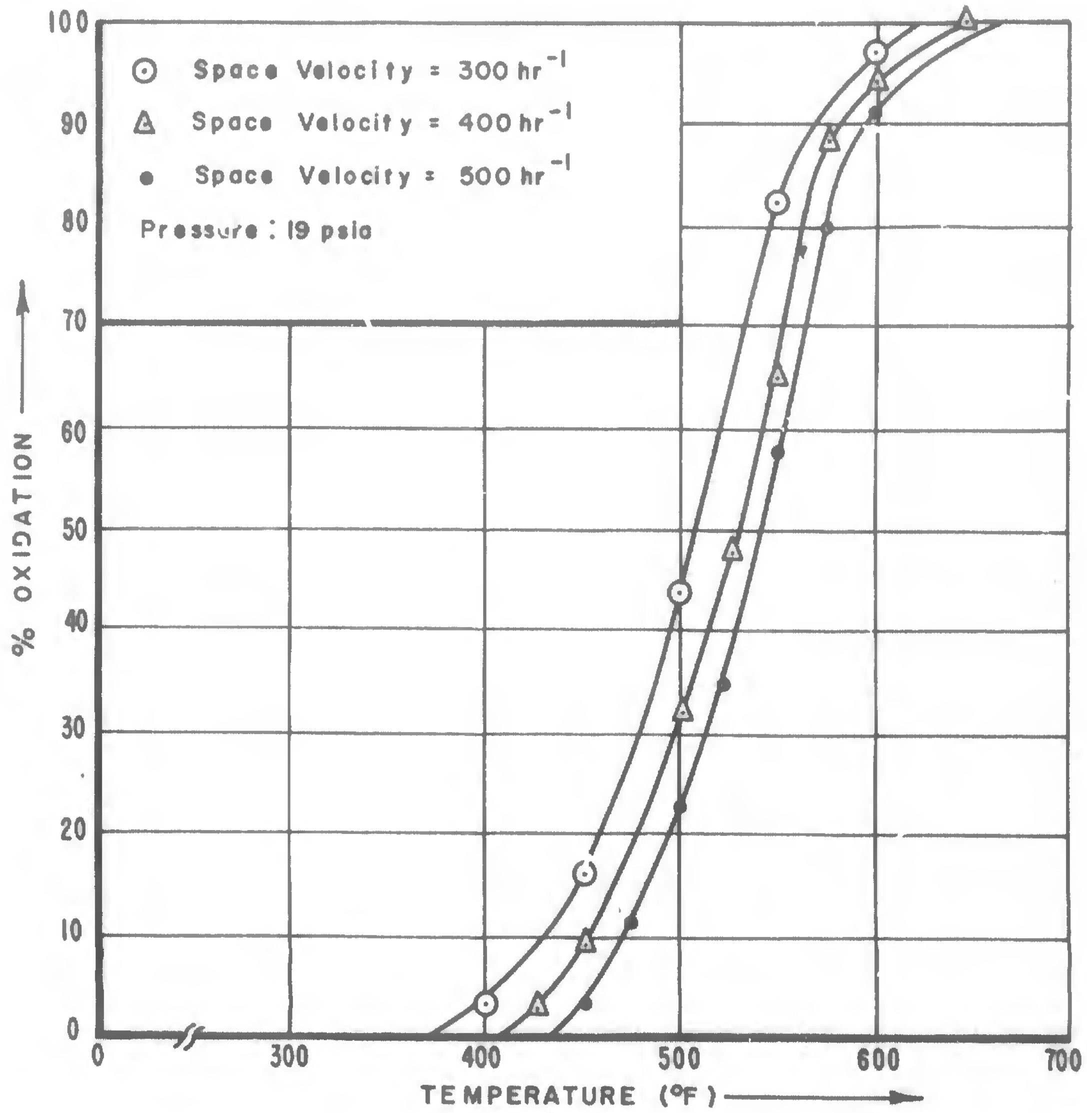

Figure 7. Effect of Space Velocity on Performance of Hopcalite Catalyst (851 grams)

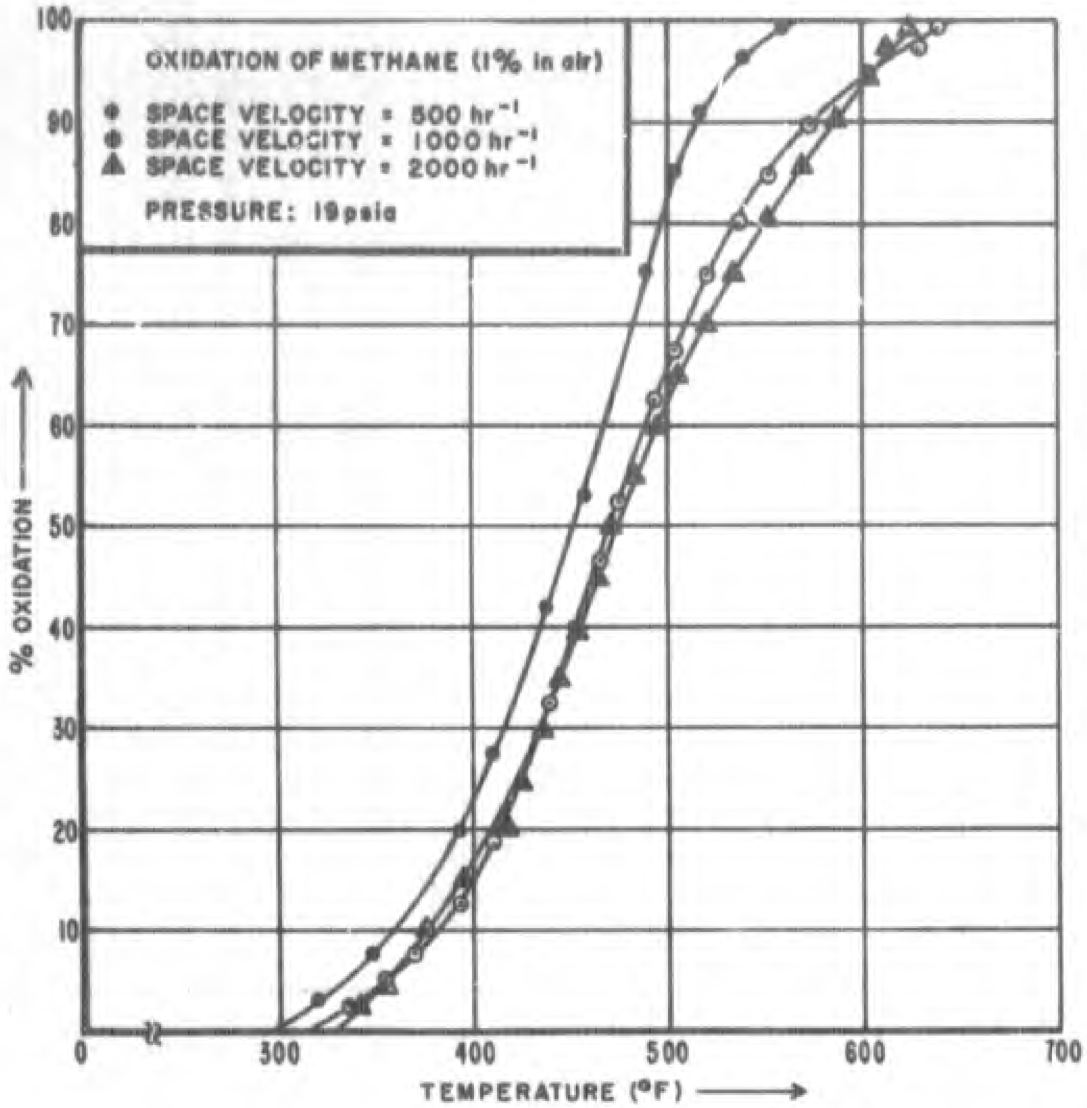

Figure 8. Effect of Space Velocity on Performance of Palladium Catalyst (763 grams)

SECTION IV

DISCUSSION OF RESULTS

PERFORMANCE OF CATALYSTS

A schematic of the reactor is presented in Figure 2. Although two of the catalysts evaluated produced no methane oxidation, the other five catalyzed the reaction to completion at some temperature between 560° and 800°F. Palladium on alumina promoted the reaction to 100 percent efficiency at a temperature of 560°F. Using a space velocity of 17,000 hr^{-1}, a 0.1 percent methane in air mixture and a 0.5 percent palladium on an alumina catalyst, Johns, Garwig and Markels* obtained an instantaneous conversion of 80 percent at 340°F, but the catalyst was quickly poisoned. Later investigations revealed that at a "threshold" temperature of 425°F the conversion was only 8 percent. From Figure 6 it is evident that, in this study, a conversion of 35 percent was encountered at 425°F using an identical type catalyst, but at a much lower space velocity. Catalyst G, palladium on granular carbon, when heated to 600°F, began to oxidize itself rather than the methane. An inspection of the catalyst immediately after cooling revealed that the carbon support was burning. One major difference was noted between Hopcalite and the palladium on alumina catalysts. While Hopcalite caused the conversion to increase slowly and steadily as the temperature rose, the palladium often produced sudden significant changes in the oxidation rate.

TEMPERATURE DISTRIBUTION

Large temperature gradients were present in the catalyst bed due to both reactor geometry and the method of heating. Satisfactory heat transfer from the nichrome wire on the outside of the reactor to the preheater in the center, necessitated rapid conduction by the catalyst bed (see Figure 2). Since both Hopcalite and alumina conduct heat poorly, the preheater was handicapped in its function and did not perform satisfactorily. Large axial temperature gradients were encountered repeatedly and although radial gradients were not measured there can be no doubt that they were present. To combat this, the temperature was elevated slowly and it was assumed that the reaction cite was at or near the hottest thermocouple. As both the temperature and the conversion efficiency increased, however, the hottest zone in the reactor shifted from thermocouple 4 to thermocouple 3. This means, of course, that at the flow rates under consideration only a fraction of the catalyst bed was being utilized and this reaction zone moved from the center of the reactor toward the entrance end as the temperature rose. An isothermal reactor system would be needed to determine the actual kinetics of the reaction. The study of kinetics or reaction rates is not meaningful in a non-isothermal reactor.

FLOW RATE CONSIDERATIONS

From the Reynolds number calculation in Section II it is evident that all flow rates used were in the transition flow region. All catalysts were evaluated in this flow region for comparison purposes and the results are shown in Figure 6. The Hopcalite catalyst produced a greater degree of conversion at a space velocity of 300 fills per hour than at 500 fills per hour (Figure 7), but due to the effect of the larger flow rate the total amount of methane oxidized actually increased. This would tend to justify using a smaller reactor operating at a much higher space velocity and in turbulent flow. Similar runs using the palladium catalyst at higher flow rates gave only marginally satisfactory results (Figure 8) but the trends are the same as the Hopcalite, i.e., at a given temperature, a lower flow rate produces a higher percent conversion but less actual material is oxidized in a given time interval.

Table III shows this trend for the Hopcalite catalyst. It is suspected that with a lower flow rate only a fraction of the catalyst bed is promoting the reaction and that this,

---

*Ibid., p. 1

along with the method of heating the reactor, may be the cause of hot spots in the catalyst bed. Therefore, from a catalyst utilization standpoint the flow should be at least large enough to "load" the entire bed. The space velocity required for this condition could be determined by using a small reactor and a wide range of flow rates at a constant temperature. Once the bed is at a loaded condition the reaction rate will be controlled by the adsorption-reaction-desorption cycle rather than mass transfer to the catalyst surface. The Reynolds number was found to play no significant role other than its effect on the space velocity. This is to be expected since in a packed bed, the transition from laminar to turbulent flow is a gradual one.

INSTRUMENTATION

A process gas chromatograph with thermal conductivity detection was used for gas analysis during the first part of the work but was replaced by a Perkin-Elmer Hydrogen Flame Detector because of the latter's sensitivity and rapidity in detecting methane. The flame detector required approximately 30 minutes for warm-up and only a moderate account of drift was encountered. The flowmeters were of the tapered tube variety and were extremely sensitive to changes in back pressure from the gas mixer. For this reason a Wallace and Tiernan pressure gauge was periodically installed between the heat exchanger and flame detector. This pressure read approximately one pound over normal atmospheric pressure and varied only with changes in the flow rate. A multipoint recorder plotted the temperature of each thermocouple junction as the reactor heated. Although an automatic controller was not used, excellent temperature control was maintained by a manually operated variable resistor. The stainless steel reactors were designed and fabricated in-house.

SYSTEM DESIGN CONSIDERATION

Operating the catalytic oxidation reactor at the lowest possible temperatures would reduce the start-up and operating power requirement but could increase the risks of producing undesirable gases inside the bed. In either case, a minimum temperature may have to be maintained to avoid certain types of catalyst poisoning, an example of which is irreversible adsorption. Although low flow rates are desirable insofar as requiring less blower power is concerned, the higher space velocities are preferred since they greatly increase catalyst efficiency. A higher efficiency will enable the designer to minimize the size, weight and power requirement of a catalytic burner.

SECTION V

CONCLUSIONS

1. One percent methane can be satisfactorily removed from an airstream by catalytic oxidation and no discontinuity in the results occurs when operating in the transition flow region.

2. The best catalyst tested was the palladium on alumina which produced complete combustion at 560°F.

3. Although at identical temperatures an increase in flow rate means a decrease in conversion efficiency, the higher flow, i.e., space velocity, increases the catalyst efficiency and the total moles of methane oxidized is greater.

4. A carbon support for catalyst materials does not appear feasible since the catalyst support is subject to oxidation.

APPENDIX

RAW DATA ON PERFORMANCE OF ALL CATALYSTS

TEST DATA SHEET

ITEM CATALYST A — PALLADIUM ON ALUMINA	WEIGHT = 763 g; SV = 500 hr^{-1}	
MANUFACTURER	MFR DWG NR	
TEST NR	SPEC NR	TYPE NR
PRIORITY NR	PROJECT NR	QUANTITY SUBMITTED
TEST BY WESS & MARSHALL	DATE 28 MAY 1965	PAGE OF PAGES

THERMOCOUPLES
TEMPERATURE - °F

TIME	1	2	3	4	5	%	% Oxidized
1023	170	245	263	292	210	100	0
1026	185	265	290	322	230	97	3
1029	204	285	317	350	252	92.5	7.5
1032	225	302	340	372	275	87.5	12.5
1037	250	320	361	393	293	80.5	19.5
1041	270	332	381	410	310	75	25
1043	280	345	393	421	320	70.5	29.5
1046	290	352	404	430	328	65	35
1048	300	362	415	442	336	58	42
1050	312	372	425	450	345	52.5	47.5
1052	320	380	435	458	350	47	53
1055	331	390	450	471	360	40	60
1057	340	400	460	480	365	32.5	67.5
1059	347	406	471	490	372	25	75
1101	352	415	481	497	375	20	80
1103	362	423	493	504	384	15	85
1105	372	431	505	513	390	10	90
1108	388	444	525	525	398	5	95
1111	402	458	541	535	405	2.5	97.5
1114	416	470	550	545	414	1.5	98.5
1121	465	515	565	560	429	1	99
1136	517	640	600	600	463	0.5	99.5
1145	543	668	625	625	485	0.5	99.5

TEST DATA SHEET

ITEM: CATAL.ST A — PALLADIUM ON ALUMINA

WEIGHT = 763 g; SV = 1000 hr^{-1}

MANUFACTURER:

MFR DWG NR:

TEST NR:

SPEC NR:

TYPE NR:

PRIORITY NR:

PROJECT NR:

QUANTITY SUBMITTED:

TEST BY: MARSHALL - WESS

DATE: 5 AUGUST 1965

PAGE OF PAGES

DESCRIPTION OF TEST *

THERMOCOUPLES
TEMPERATURE - °F

TIME		1		2		3		4		5		%	% Oxidized		Press
1229		140		120		125		125		123		100	0		15.19
1241		222		193		200		218		192		100	0		15.1
1248		269		235		255		283		245		100	0		15.20
1254		304		280		300		335		290		97.5	2.5		15.22
1256		321		285		320		355		305		95	5.		15.21
1258		333		295		335		370		322		92.5	7.5		15.22
1300		350		310		354		394		340		87.5	12.5		15.23
1302		365		323		372		412		355		80	20		"
1305		385		345		395		440		378		67.5	32.5		"
1307		395		350		410		454		390		60.	40		"
1308		403		360		420		465		400		53.5	46.5		"
1309		410		366		431		475		405		47.5	52.5		"
1311		425		380		454		495		421		37.5	62.5		"
1312		435		390		465		505		430		32.5	67.5		"
1315		450		403		495		523		445		25	75		15.26
1316		465		415		518		540		455		20	80		15.27
1318		480		430		545		554		470		15	85		"
1320		500		445		575		570		485		10	90		"
1322		523		466		605		590		500		5	95		15.25
1325		554		495		630		607		516		2.5	97.5		15.28
1328		600		545		641		630		538		1.0	99.0		

* Installed Low Pressure Regulator Between Reactor and Detector.

TEST DATA SHEET

ITEM: CATALYST A — PALLADIA ON ALUMINA

WEIGHT = 763 g; SV = 2,000 hr^{-1}

MANUFACTURER:

MFR DWG NR:

TEST NR:

SPEC NR:

TYPE NR:

PRIORITY NR:

PROJECT NR:

QUANTITY SUBMITTED:

TEST BY: MARSHALL - WESS

DATE: 6 AUGUST 1965

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THERMOCOUPLES
TEMPERATURE - °F

TIME	1	2	3	4	5	%	% Oxidized	Press
1030	246	218	255	294	287	100	0	15.72
1040	275	238	275	321	320	100	0	15.71
1045	285	250	286	340	340	97.5	2.5	"
1048	300	257	296	355	355	95.	5.	"
1052	315	270	315	375	375	90	10.	"
1056	323	280	325	395	391	85	15	"
1100	340	293	345	418	415	80	20	"
1102	345	297	350	425	423	75	25	15.75
1104	350	302	355	435	431	70	30	"
1105	355	305	360	445	439	65	35	"
1106	360	310	365	455	445	60	40	"
1108	363	313	370	465	450	55	45	"
1109	365	317	375	473	458	50	50	"
1110	371	320	380	484	465	45	55	"
1111	375	325	385	496	471	40	60	"
1112	379	326	393	505	476	35	65	"
1114	385	335	400	522	485	30	70	"
1115	390	340	410	535	494	25	75	"
1117	398	345	421	551	500	20	80	"
1119	405	352	442	570	510	15	85	"
1121	418	361	475	586	520	10	90	"
1124	430	374	521	604	530	5	95	"
1125	440	380	560	611	535	2.5	97.5	"
1127	451	393	625	620	545	0	100	"

TEST DATA SHEET

ITEM CATALYST B — RHODIUM ON ALUMINA

WEIGHT = 869 g; SV = 500 hr^{-1}

MANUFACTURER

MFR DWG NR

TEST NR

SPEC NR

TYPE NR

PRIORITY NR

PROJECT NR

QUANTITY SUBMITTED

TEST BY WESS & MARSHALL

DATE 26 MAY 1965

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THERMOCOUPLES
TEMPERATURE - °F

TIME	1	2	3	4	5	%	%Oxidized
1221	200	270	295	325	238	100	0
1228	232	304	347	360	280	100	0
1232	258	325	378	410	307	99	1
1236	280	345	403	435	330	96	4
1243	318	377	440	471	365	87.5	12.5
1246	335	390	452	483	375	81.5	18.5
1249	346	400	465	493	384	76.	24
1252	358	410	475	500	393	70	30
1255	365	420	483	510	400	65	35
1257	375	428	492	517	406	57.5	42.5
1300	381	440	504	528	415	47.5	52.5
1303	393	450	513	540	423	40	60.
1306	400	457	525	550	430	32.5	67.5
1309	410	460	542	560	440	25.	75
1313	422	482	562	575	450	17.	83
1316	430	493	580	585	455	12.5	87.5
1319	440	500	595	595	460	8.	92
1322	450	510	608	604	465	5.5	94.5
1325	460	520	622	614	470	3.5	96.5
1328	475	530	630	620	475	2.	98
1330	490	545	638	625	480	1.	99
1334	510	563	642	635	485	0.5	99.5
1339	545	610	650	645	495	0	100
1345	562	645	660	655	503	0	100
1349	575	660	665	665	510	0	100
1400	600	695	680	680	525	0	100
1405	610	708	685	685	535	0	100

TEST DATA SHEET

ITEM: CATALYST C — HOPCALITE

WEIGHT 851 g; SV = 300 hr^{-1}

MANUFACTURER:

MFR DWG NR:

TEST NR:

SPEC NR:

TYPE NR:

PRIORITY NR:

PROJECT NR:

QUANTITY SUBMITTED:

TEST BY: MARSHALL & CIVETZ

DATE: 28 JANUARY 1965

PAGE OF PAGES

THERMOCOUPLES
TEMPERATURE - °F

1	2	3	4	5	%	% Oxidized
320	315	380	395	328	100	0
310	320	373	385	315	100	0
340	350	399	400	320	97	3
359	350	399	395	327	97	3
400	398	449	449	361	85	15
384	390	448	449	360	85	15
381	391	448	450	359	85	15.
440	435	500	491	395	57	43
430	432	500	500	399	55	45
423	431	500	499	395	57	43
410	460	541	558	424	28	72
462	470	555	550	433	21	79
492	509	610	615	465	3	97

TEST DATA SHEET

ITEM: CATALYST C — HOPCALITE

WEIGHT 851 g; SV = 400 hr^{-1}

MANUFACTURER

MFR DWG NR

TEST NR

SPEC NR

TYPE NR

PRIORITY NR

PROJECT NR

QUANTITY SUBMITTED

TEST BY: MARSHALL & CIVETZ

DATE: 2 FEBRUARY 1965

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THERMOCOUPLES
TEMPERATURE - °F

1	2	3	4	5	%	% Oxidized
235	319	389	420	300	100	0
295	315	387	402	320	100	0
351	370	435	449	355	91	9
355	375	437	450	355	91	9
390	413	481	492	385	70	30
400	419	489	498	385	68	32
402	430	495	503	385	66	34
415	430	515	520	403	52	48
415	450	533	555	425	38	62
429	449	539	549	429	37	63
438	452	542	549	427	35	65
490	496	576	560	443	11	89
486	500	600	588	460	6	94
489	507	597	581	455	6	94
520	556	645	635	492	0	100

TEST DATA SHEET

ITEM: CATALYST C — HOPCALITE

WEIGHT 851 g; SV = 500 hr^{-1}

MANUFACTURER:

MFR DWG NR:

TEST NR:

SPEC NR:

TYPE NR:

PRIORITY NR:

PROJECT NR:

QUANTITY SUBMITTED:

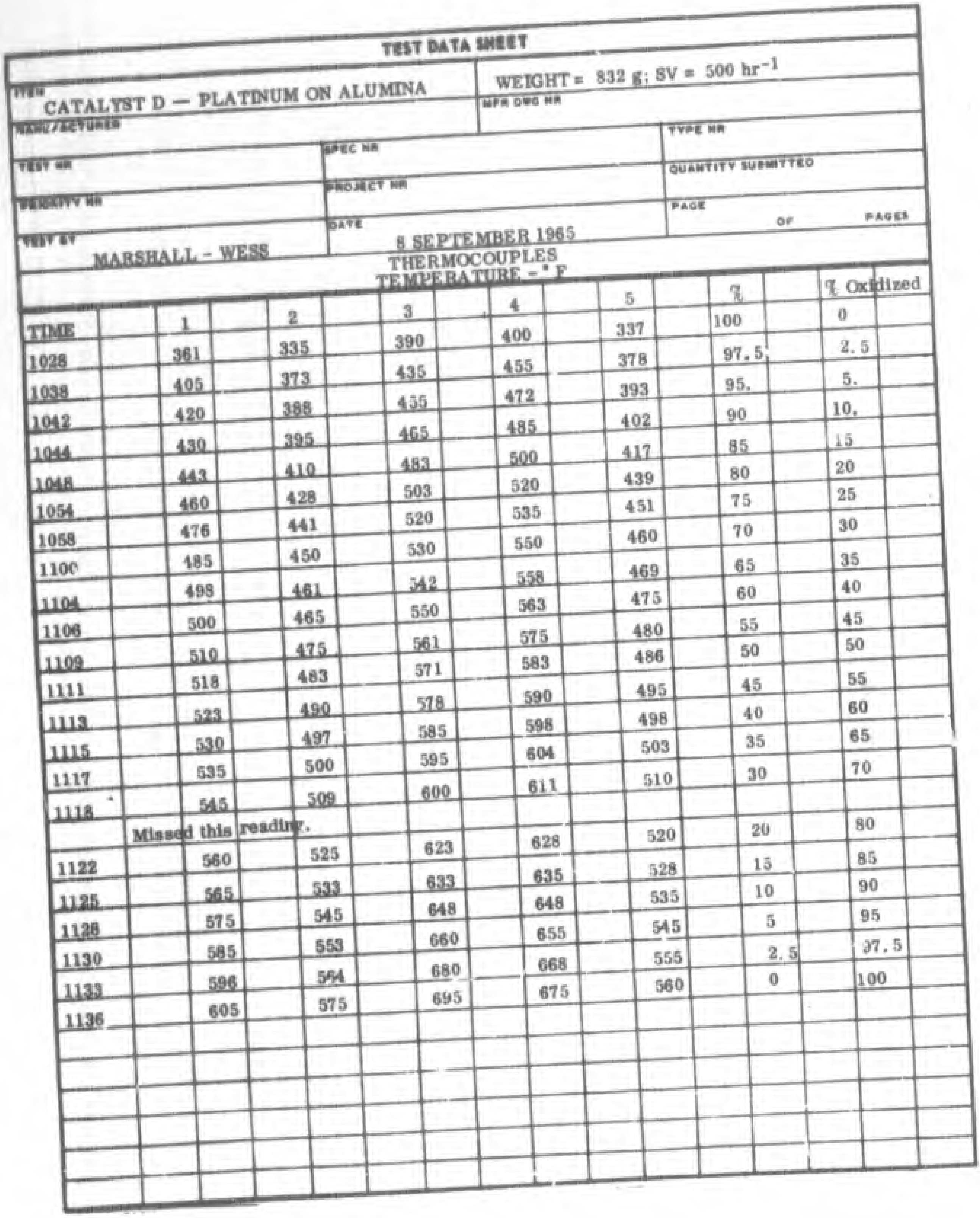
TEST BY: MARSHALL & CIVETZ

DATE: 3 FEBRUARY 1965

PAGE OF PAGES

THERMOCOUPLES
TEMPERATURE - ° F

1	2	3	4	5	%	% Oxidized
319	356	430	450	356	97	3
326	361	425	446	355	97	3
340	382	451	475	372	88	12
350	389	455	475	377	88	12
348	388	456	471	373	89	11
363	406	480	500	395	77	23
365	410	480	500	392	77	23
368	406	480	495	392	78	22
350	407	491	525	400	67	33
387	410	499	525	405	65	35
380	412	499	522	409	65	35
398	432	522	550	423	46	54
400	440	531	556	429	42	58
400	438	530	550	429	43	57
416	455	554	575	440	26	74
425	458	556	575	440	23	77
430	460	565	576	445	20	80
442	479	590	595	456	10	90
450	483	600	595	462	8	92
461	490	602	598	462	7	93
475	510	622	609	475	3	97
480	530	629	619	480	2	98
490	530	620	610	475	2	98
508	570	651	648	501	0	100
511	579	648	640	502	0	100



TEST DATA SHEET

ITEM: CATALYST D — PLATINUM ON ALUMINA

WEIGHT = 832 g; SV = 500 hr^{-1}

MFR DWG NR

MANUFACTURER

TYPE NR

TEST NR

SPEC NR

QUANTITY SUBMITTED

WARRANTY NR

PROJECT NR

PAGE OF PAGES

TEST BY: MARSHALL - WESS

DATE: 8 SEPTEMBER 1965

	THERMOCOUPLES TEMPERATURE - °F						
TIME	1	2	3	4	5	%	% Oxidized
1028	361	335	390	400	337	100	0
1038	405	373	435	455	378	97.5	2.5
1042	420	388	455	472	393	95.	5.
1044	430	395	465	485	402	90	10.
1048	443	410	483	500	417	85	15
1054	460	428	503	520	439	80	20
1058	476	441	520	535	451	75	25
1100	485	450	530	550	460	70	30
1104	498	461	542	558	469	65	35
1106	500	465	550	563	475	60	40
1109	510	475	561	575	480	55	45
1111	518	483	571	583	486	50	50
1113	523	490	578	590	495	45	55
1115	530	497	585	598	498	40	60
1117	535	500	595	604	503	35	65
1118	545	509	600	611	510	30	70
	Missed this reading.						
1122	560	525	623	628	520	20	80
1125	565	533	633	635	528	15	85
1128	575	545	648	648	535	10	90
1130	585	553	660	655	545	5	95
1133	596	564	680	668	555	2.5	97.5
1136	605	575	695	675	560	0	100

TEST DATA SHEET

ITEM CATALYST E — RUTHENIUM ON ALUMINA

WEIGHT = 838 g; SV = 500 hr^{-1}

MANUFACTURER

MFR DWG NR

TEST NR

SPEC NR

TYPE NR

PRIORITY NR

PROJECT NR

QUANTITY SUBMITTED

TEST BY MARSHALL - WESS

DATE 15 SEPTEMBER 1965

PAGE OF PAGES

THERMOCOUPLES
TEMPERATURE - °F

TIME	1	2	3	4	5	%	% Oxidized
1105	485	460	533	535	450	97.5	2.5
1111	495	470	548	550	460	95	5
1124	510	486	568	570	476	90	10
1135	525	500	584	585	490	85	15
1142	535	513	595	595	498	80	20
1148	545	520	605	602	504	75	25
1153	553	526	612	610	510	70	30
1158	560	535	621	617	516	65	35
1202	565	540	630	625	521	60	40
1206	573	549	640	633	529	55	45
1209	579	553	647	640	534	50	50
1213	584	556	655	645	540	45	55
1216	591	563	665	653	545	40	60
1219	596	570	675	660	550	35	65
1223	604	575	684	668	555	30	70
1226	611	584	695	673	560	25	75
1230	620	590	706	684	568	20	80
1234	630	600	721	692	575	15	85
1239	645	613	740	704	586	10	90
1246	663	633	762	720	600	5	95
1254	686	656	788	735	615	2.5	97.5
1306	728	700	798	756	640	1.5	98.5

TEST DATA SHEET

ITEM
CATALYST F — $V_2 0_5$ - $Mo 0_3$ ON ALUMINA

WEIGHT = 1063 g; SV = 500 hr^{-1}

MANUFACTURER

MFR DWG NR

TEST NR

SPEC NR

TYPE NR

PRIORITY NR

PROJECT NR

QUANTITY SUBMITTED

TEST BY
MARSHALL - WESS

DATE
16 JULY 1965

PAGE OF PAGES

THERMOCOUPLES
TEMPERATURE - °F

TIME		1		2		3		4		5		%		% Oxidized	
1000		471		531		625		665		500		100		0	
1019		665		697		815		840		655		100		0	
1050		835		845		963		973		783		100		0	
1057		855		863		985		1000		812		100		0	

TEST DATA SHEET

ITEM CATALYST G — PALLADIUM ON GRANULAR CARBON

WEIGHT = 350 g; SV = 500 hr^{-1}

MANUFACTURER

MFR DWG NR

TEST NR

SPEC NR

TYPE NR

PRIORITY NR

PROJECT NR

QUANTITY SUBMITTED

TEST BY MARSHALL - WESS

DATE 27 JULY 1965

PAGE OF PAGES

THERMOCOUPLES
TEMPERATURE - °F

TIME	1	2	3	4	5	%	% Oxidized
1143	251	275	309	314	251	100	0
1226	745	675	910	750	564	90	10
1230	933	1053	855	763	580	85	15

NOTE: Carbon support began to oxidize between 600° and 700°F and temperature increased rapidly to 1600°F. Passing of pure nitrogen across catalyst terminated oxidation, whereas air supported it. This, along with the condition of the catalyst, confirmed the belief that carbon was being burned preferentially to methane.

UNCLASSIFIED
Security Classification

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Air Force Flight Dynamics Laboratory, Research and Technology Division, Air Force Systems Command, Wright-Patterson Air Force Base, Ohio 45433

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2b. GROUP

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11. SUPPLEMENTARY NOTES

12. SPONSORING MILITARY ACTIVITY
Air Force Flight Dynamics Laboratory, Research and Technology Division, Air Force Systems Command, Wright-Patterson AFB, Ohio

13. ABSTRACT
The feasibility of removing methane from an airstream by catalytic oxidation at low flows was investigated by passing a one percent, by volume, methane in airstream through an in-house designed and fabricated stainless steel reactor. The reactor contained either Hopcalite, 5% V_2O_5 - 5% MoO_3 on alumina, or one of the following materials supported on alumina: 0.5% palladium, 0.5% platinum, 0.5% rhodium, and 0.5% ruthenium. Some discontinuity in the data reported in the literature on methane oxidation at or around the transition flow region prompted a closer investigation in this area. In addition, space velocities around 500 fills per hour were used to insure chemical equilibrium and for comparison with the more common value of approximately 15,000 fills per hour used in most industrial applications. Experiments performed using the above catalysts revealed no deleterious effects associated with operating in the transition flow region, but did point out the advantage of using high space velocities for maximum catalyst utilization. The palladium catalyst proved superior to all others tested and catalyzed the reaction to completion at 560°F. Conversion versus temperature curves are plotted for each catalyst flow rate combination and the effect of space velocity on conversion at constant temperature is shown for the Hopcalite and palladium catalysts. Variables which affected the accuracy of experimentation most are discussed and possible improvements suggested.

DD FORM 1 JAN 64 1473

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14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Catalytic oxidation of methane						
Catalytic oxidation of hydrocarbons						
Precious metal oxidation catalyst						
Trace contamination						
Atmosphere control						
Life support						

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