



Catalytic polyforming of gas oil at increased mol ratios  
by Robert D Lewallen

A THESIS Submitted to the Graduate Faculty in partial fulfillment of the requirements for the degree of Master of Science in Chemical Engineering at Montana State College  
Montana State University  
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Abstract:

This investigation was made primarily to study the effect of an increase in mol ratio on gasoline yields in catalytic polyforming. The secondary objective, was to study the effect of pressure upon catalytic polyforming at high mol ratios.

The investigation was made using Houdry synthetic aluminum silicate fixed-bed catalyst with virgin gas oil as the charging stock, and a mixture of hydrocarbon gases as the outside gas. The mol ratios of gas to oil investigated varied from 2.0 to 15.8. Temperatures were varied from 564 to 537° in obtaining maximum gasoline yields for each mol ratio. Liquid space velocities were held constant at 4-6 hr<sup>-1</sup>. Pressures varying from atmospheric to 2000 psig were investigated at mol ratios ranging from 9-11.5. During a typical run, approximately 490 grams of feed were passed over the 1000 ml of catalyst.

Results of the investigation of mixed-gas catalytic polyforming at increased mol ratios are as follows: (1) gasoline yields in excess of those obtained from catalytic cracking can be obtained, (2) gasoline yields increase steadily with each increase in mol ratio up to a mol ratio range 9-11.5, after which gasoline yields remain constant through a mol ratio of 15.8, (3) pressure increases from atmospheric to 300 psig had greatest effect upon gasoline yields, (4) increases in pressure from 300 psig to 2000 psig had no noticeable effect upon gasoline yields, (5) an increase in mol ratio tends to increase ultimate yield and decrease carbon laydown.

It was found that a mol ratio of 9.45 at a conversion of 61.9 per cent gave a maximum gasoline yield of 49.2 per cent.

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AT

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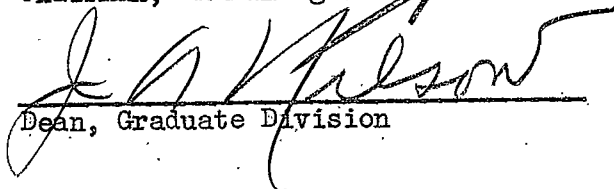
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TABLE OF CONTENTS

	Page
ABSTRACT.....	3
I. Introduction.....	4
II. Equipment, Methods and Materials.....	
A. Equipment.....	8
B. Methods.....	13
C. Materials.....	17
III. Sample Calculations.....	18
IV. Results.....	24
V. Summary.....	29
VI. Acknowledgment.....	30
VII. Literature Cited.....	31
VIII. Appendix.....	32

### ABSTRACT

This investigation was made primarily to study the effect of an increase in mol ratio on gasoline yields in catalytic polyforming. The secondary objective was to study the effect of pressure upon catalytic polyforming at high mol ratios.

The investigation was made using Houdry synthetic aluminum silicate fixed-bed catalyst with virgin gas oil as the charging stock, and a mixture of hydrocarbon gases as the outside gas. The mol ratios of gas to oil investigated varied from 2.0 to 15.8. Temperatures were varied from 364 to 537°C in obtaining maximum gasoline yields for each mol ratio. Liquid space velocities were held constant at 4-6 hr<sup>-1</sup>. Pressures varying from atmospheric to 2000 psig were investigated at mol ratios ranging from 9-11.5. During a typical run, approximately 490 grams of feed were passed over the 1000 ml of catalyst.

Results of the investigation of mixed-gas catalytic polyforming at increased mol ratios are as follows: (1) gasoline yields in excess of those obtained from catalytic cracking can be obtained, (2) gasoline yields increase steadily with each increase in mol ratio up to a mol ratio range 9-11.5, after which gasoline yields remain constant through a mol ratio of 15.8, (3) pressure increases from atmospheric to 300 psig had greatest effect upon gasoline yields, (4) increases in pressure from 300 psig to 2000 psig had no noticeable effect upon gasoline yields, (5) an increase in mol ratio tends to increase ultimate yield and decrease carbon laydown.

It was found that a mol ratio of 9.45 at a conversion of 61.9 per cent gave a maximum gasoline yield of 49.2 per cent.

## INTRODUCTION

Catalytic cracking and thermal polyforming are two processes in Petroleum Technology which were developed to a greater or lesser degree during the war period. The catalytic cracking process is in widespread use, but the polyforming process is still being developed.

The polyform process has been described by Offut et al (1) as an efficient means for producing high quality motor gasoline by the thermal conversion of naphthas or gas oils at high temperatures and high pressures in the presence of recirculated hydrocarbon gases. These light hydrocarbons in the charge make possible a substantially greater degree of severity of cracking and higher gasoline octane level than it is possible to achieve in most thermal cracking processes. These gases also minimize tar and coke formation by interaction with many of the olefins produced, some of which would otherwise undergo polymerization to form tar and coke. At the same time, the conditions of temperature and pressure are suitable for conversion of C<sub>3</sub> and C<sub>4</sub> hydrocarbons to gasoline. The hydrocarbon gases used in the polyform process may be those obtained from cracking or they may be from some other source.

To incorporate the advantages of the thermal polyforming and catalytic cracking process, Dev (2) investigated the process of gas oil polyforming under catalytic conditions using propane as the outside gas. This process was called catalytic polyforming. The yields of gasoline from the catalytic cracking of virgin gas oil over Houdry catalyst with

and without propane at various pressures were compared. This study showed that an increased yield of gasoline could be obtained at higher pressures in the catalytic polyform process using propane as the outside gas. The increased gasoline yield was obtained at temperatures lower than those used for thermal polyforming.

Mayfield (3) investigated catalytic polyforming using isobutylene as the outside gas and found that gasoline yields were approximately the same whether obtained at space velocities of 4-6 hr<sup>-1</sup> or 0.5-1 hr<sup>-1</sup>. The lower space velocity is in the range normally used in catalytic cracking. This ability of catalytic polyforming to operate efficiently at space velocities slightly higher than the space velocity used in catalytic cracking is one of the advantages of catalytic polyforming over catalytic cracking.

Polich (4) evaluated several catalysts at 900 psig using normal butane as the outside gas. The space velocity was held constant in the range of 4-6 hr<sup>-1</sup>. Of the catalysts studied Houdry aluminum silicate catalyst was found to be best suited to catalytic polyforming.

Ennenga (5) investigated catalytic polyforming under pressures of 0, 300, 600, 900, and 1200 psig using iso-butane as the outside gas. Space velocity was held relatively constant at 4-6 hr<sup>-1</sup>. It was found that gasoline yields increased through 900 psig. The greatest increase in gasoline yield as pressure was varied, occurred between 0 and 300 psig. When pressures of 1200 psig were applied a slight decrease in gasoline production was noted.

From a study of catalytic polyforming using a mixed hydrocarbon

gas and pressures ranging up to 2000 psig, Hamilton (6) found definite increases in gasoline yields and octane numbers over those obtainable from conventional atmospheric catalytic cracking. At a pressure of 1500 psig, Hamilton obtained a gasoline yield of 37.7 per cent gasoline based on oil charged. Octane numbers obtained at this pressure were 79.1 by motor method and 84.4 by the research method. Catalytic cracking gave a gasoline yield of 33.6 per cent and octane numbers of 72.8 by motor method and 79.3 by the research method. These increased yields and high octane numbers obtained from catalytic polyforming are evidence of the advantages of catalytic polyforming over the conventional atmospheric catalytic cracking process.

The mixed gas used by Hamilton contained the following constituents expressed in weight per cents: 30 per cent n-butane, 10 per cent iso-butane, 20 per cent iso-butylene, 20 per cent propane, and 20 per cent propylene. This gas is representative of the effluent gases of most refineries and was found more effective than any single gas in procuring high yields of gasoline with relatively high octane ratings.

In these previous investigations, mol ratio of gas to oil was held constant at approximately 3.0 with the result that little was known about mol ratio as a process variable in catalytic polyforming. In view of the uncertainty of the effect of this variable upon catalytic polyforming, an investigation was undertaken to determine the effect of mol ratio on gasoline yield in catalytic polyforming.

The octane numbers and gasoline yields obtained from catalytic polyforming at mol ratios ranging from 2 to 15.8 were compared. The

ultimate yields and carbon formations obtained from each mol ratio were also compared. At each mol ratio, the maximum yield was found by varying the temperature at which the reaction took place. The data thus obtained gave a curve, when plotted, of yield versus conversion for each mol ratio; and in this way the maximum gasoline yield for a given mol ratio was determined. To evaluate the results obtained from catalytic polyforming, catalytic cracking data obtained in the same reaction unit were used as a basis for comparison.

## EQUIPMENT, METHODS, MATERIALS

### A. Equipment

The equipment used in this investigation consisted of a 0-2500 psig reaction system, shown in Figure 1. This reaction system may be broken down into four major sections according to function. They are as follows: (1) feeding section, (2) reactor section, (3) condensing and receiving section, and (4) gas section.

#### Feeding Section

The feeding section consisted of a feed cylinder, a Jerguson gage, and a pump. The body of the feed cylinder was constructed from a 12-inch section of extra strong three-inch pipe and two extra strong three-inch steel caps. The pipe was threaded and the caps were screwed on each end of the pipe and welded securely in place. Both caps were drilled and tapped for half-inch pipe, and close nipples of extra strong steel pipe were screwed into the caps and also welded in place. One-half inch Ideal needle valves of 1000 psig rating were fitted to each nipple. The valves were fitted with brass fittings so that 1/4-inch copper tubing could be connected to one end and 1/2-inch copper tubing could be connected to the other end. The large tubing, together with the pressure equalization line shown in Figure 1 was designed to insure free flow of feed to the Jerguson. The bottom of the Jerguson was connected to a stainless steel Kerotest globe valve by 1/4-inch copper tubing. By means of the valve, the feed rate was adjusted until the pressure in the reactor became high

enough to equal the pressure in the feed cylinder. The valve was, in turn, connected by two lengths of 1/4-inch tubing to each side of a double acting pump. The pump used was a Hills-McCanna, Type UM-2F, single unit, two feed, 1/4 horse-power pump. The pump was connected to the top of the reactor by 1/8-inch stainless steel tubing and Weatherhead Ermeto pressure couplings and fittings.

### Reactor Section

The reactor was constructed from a three-foot piece of 18-8-C, Type 347, 2-1/2-inch extra strong I. P. S. pipe. A 2-1/2-inch welding cap, fitted with a 1/2-inch extra strong pipe nipple, was welded to the lower end of the stainless steel pipe. An extra heavy tee was fitted to the 1/2-inch pipe. The lower opening held the 4-foot long, 1/4-inch standard pipe thermowell which extended through the reactor along the central axis. To the side opening of the tee a 1/2-inch pipe was fitted and allowed to protrude through the reactor case. An extra strong ell was attached to the short length of pipe and was fitted with a six-inch nipple leading to an extra strong tee. To the side opening of the tee was connected a 0-5000 psig pressure gage. The run was fitted with a 25,000 pound stainless steel Aminco super-pressure valve.

To the top of the reactor a 2-1/2-inch extra strong welding neck flange was welded and a 2-1/2-inch extra strong blind flange bolted to the welding neck flange. For insurance against loss due to leakage, a stainless steel gasket was inserted between the flanges in the final assembly. A short nipple was welded to the flange and was, in

turn, fitted with an extra strong cross. The top opening of this cross was connected to a high pressure Hoke needle valve. An Emeto tee was screwed into the valve and the run openings were connected to the feed lines from the pump. One side opening of the extra strong cross was connected to the nitrogen cylinder so that the reactor might be purged. The other opening was connected by a short nipple to an extra strong tee. The off-stream side of the tee was fitted with a Black, Sivalls, and Bryson frangible safety disc which was equipped with a 3100 psig Monel diaphragm discharging to the exterior of the building by means of a blow-down line. The run of the tee was connected to another Hoke valve to permit passage of air into the reactor for the catalyst burn-off.

The reactor was originally designed to hold 3000 ml of catalyst because it was believed liquid space velocities of  $0.5-1.0 \text{ hr}^{-1}$  would be optimum. Subsequent research carried out with the 0-900 psig unit indicated that the process operated efficiently at space velocities of  $4-6 \text{ hr}^{-1}$ . Therefore, the catalyst volume was reduced to 1000 ml. This was done by inserting a number of cylindrical steel blocks which were drilled to allow sufficient space for the passage of the feed between the blocks and the thermowell. These blocks served very efficiently as a preheat section.

Heat was supplied to the reactor by means of five 48-foot lengths of Nichrome wire with resistance of 1.71 ohms per foot. These wires were threaded with fish-spine insulators and wrapped around the reactor over a layer of asbestos tape. Three of the coils furnished the heat for the preheat section and the remaining two covered the

catalyst section. Each of the windings drew six amperes from a 110-volt autotransformer.

Four iron-constantan thermocouples were inserted into the thermowell; one junction was placed in the preheat section and the other three were equally spaced along the catalyst bed. Four-hole porcelain insulators were obtained from the Stupakoff Company and were of such a diameter that two separate lengths of the insulators could be simultaneously inserted into the 1/4-inch thermowell. This arrangement made possible a permanent thermocouple. These thermocouples were connected to a potentiometer.

For safety, the reactor was housed in a steel case which took the shape of an inverted frustrum of a pyramid. Each side of the frustrum was 18 inches by 60 inches by 8 inches by 1/4-inch thick. Three sides were fastened together by fillet welding to 1/8-inch by 1-1/2-inch angle irons placed in the corners of the case. The bottom plate was bolted to the angle iron, as was the front side. The case was supported by two short lengths of angle iron which were welded to the case and to the laboratory rack. Further support was gained by welding three one-inch pipes to the tops of the sides of the reactor case. These pipes extended to the floor in the form of a tripod.

To support the reactor in the case, a piece of 1/4 by 2-inch bar stock 20 inches long was drilled with a one-inch hole and slipped over the one-inch nipple extending from the blind neck flange of the reactor. This bar was held in place by the extra strong cross and the ends were pinned to the sides of the case so that the reactor would

be rigidly held in place. After the reactor had been lowered in place, the space between the reactor and the case was completely filled with diatomaceous earth to minimize heat losses.

#### Condensing and Receiving Section

To the bottom of the pressure regulating valve was attached a short piece of copper tubing which was connected to a Pyrex glass Liebig condenser. A 1000 ml flask with a side opening was used as a receiver. The flask was cooled in an isopropanol-dry-ice bath contained in a one-gallon thermoflask. The outlet of the receiver was connected by rubber tubing to the gas condensing system shown in Figure 1.

#### Gas Section

The cold traps were followed by a tee, the off side of which was connected to a 200 ml evacuated glass bottle so that a sample of the non-condensable gases could be taken during the run. A Precision Wet Test Meter followed the gas sample bottle; and it, in turn, was connected to a blow-down line to the outside of the building.

Distillation of the product was carried out in a 16-plate Oldershaw column. A distilling head suitable for either high or low temperature fractionation was attached to the column. This head could be filled with dry ice-isopropanol for low boiling constituents and with wet ice for those boiling above 20°C. Two vapor traps, connected in series and immersed in dry ice-isopropanol were used to catch the product of distillation. The distillation flask was heated by a 110-volt 550 watt heater controlled by an autotransformer.

Octane number determinations were done by the Phillips

Petroleum Company in their Bartlesville, Oklahoma laboratories.

### B. Methods

#### 1200 psig runs

The feed cylinder was evacuated with a Cenco Magavac Pump, charged with a predetermined amount of gas oil, weighed on a 20-kg capacity triple beam balance, and then cooled. After the oil had been cooled thoroughly, the cylinder was placed on the balance and connected to a cylinder of mixed gas by means of a short length of Saran tubing. The valve on the feed cylinder and the valve on the gas cylinder were opened and the desired amount of mixed gas was allowed to pass into the feed cylinder. The actual amount of mixed gas in the feed cylinder was determined by re-weighing the cylinder. The charged cylinder was then allowed to warm to room temperature.

The reactor was heated to the desired temperature and purged with nitrogen. Upon completion of the nitrogen purge, all valves were closed with the exception of the feed valve between the pump and the reactor. After the feed cylinder was weighed and connected to the system as shown in Figure 1, the gas sample bottle was evacuated, weighed and connected to the gas section of the apparatus. The condensing and receiving section was set up as shown in Figure 1. The Jerguson was filled with a portion of the charge by opening the lower valve of the feed cylinder.

At one-minute intervals from the start of the run, thermocouple readings were recorded. Timing of the run began when the valve between the pump and the Jerguson gage was opened. The pump was started

when the valve was opened, but the rate of feed was controlled with the aid of the Jerguson gage and a valve until the pressure in the reactor equalled the pressure in the feed cylinder. This initial feed rate was regulated by noting the time for any given drop in liquid level in the gage which had been previously calibrated. After the pressure built up, feed rate was adjusted by regulating the stroke of the pump.

As the reactor pressure regulating valve was closed, the hot expanding gases within the reactor caused the pressure gradually to rise. When the pressure reached the desired level, the pressure regulating valve was opened just enough to maintain this pressure until the end of the feeding period which was indicated by the last of the liquid feed passing out of the Jerguson gage. The valve in the feed line on top of the reactor was immediately closed and the pump shut off. A sample of the non-condensable gas was taken at this time and weighed so that its density could be determined. The reactor was continuously bled to atmospheric pressure at approximately the same rate as it had been pressurized.

Upon completion of the run, the receivers in the condensing train were immediately weighed to determine the weight of condensable product. These liquids were poured into the receiving flask and the total weight recorded.

So that any traces of oil in the reactor might be recovered, the reactor was connected through a series of cold traps to the vacuum pump and evacuated. The reactor was simultaneously purged with nitrogen which aided materially in recovering the last traces of oil. The

recovered material was weighed and added into the weight balance as residue since under these conditions the potential gasoline in it would be negligible.

#### Distillation Procedure

The receiving flask which contained the condensable gases in addition to cracked stock was attached to the Oldershaw column by means of a ground glass taper joint. The distilling head and "10-250°C thermometer were also attached by means of ground glass joints using silicone grease as the sealing compound. A dry ice-isopropanol mixture was used to cool the head in the first part of the distillation. The product take-off entered two vapor traps connected in series and was placed in Dewar flasks which contained dry-ice and isopropanol. The vapors in the pot were allowed to reflux until equilibrium was reached. The autotransformer was set at 34 volts and the distillation allowed to proceed to 7°C. At this point, after a period of total reflux, the vapor traps containing the lights were weighed and replaced by a weighed gasoline trap. When the temperature approached 20° C, the isopropanol was removed from the distilling head and wet ice inserted in its place. The distillation was then allowed to proceed to 204°C. The gasoline so obtained was weighed, and the residue was weighed when the column reached room temperature.

#### Catalyst Regeneration

During the run, a deposit of carbonaceous material was laid down upon the catalyst reducing its activity. In order to restore catalyst

activity and establish a weight balance, this carbonaceous deposit had to be burned off. This was accomplished by passing air through a gas meter and into the reactor. The reactor was previously heated to a temperature of approximately  $450^{\circ}\text{C}$  by means of the heating elements. When the burning had started, the heating coils were shut off and the temperature controlled by adjusting the rate of air passing over the catalyst. The effluent gas was analyzed at regular intervals by means of an Orsat apparatus to determine the per cent of carbon dioxide, carbon monoxide, and oxygen. These per cents were plotted and the carbon lay-down was calculated as shown in the sample calculations.

#### Determination of Feed Remaining in Feed System

A certain amount of gas remains in the feed cylinder at the end of each run. To determine the amount of gas left in the cylinder, the method used by Mayfield was adopted. By this method, the valves of the feed cylinder were closed at the end of the run before the feed system was depressurized. The cylinder was then removed from the system and weighed. This weight was then compared with the weight of the evacuated cylinder. It was found that ten grams of gas remained in the cylinder at the end of each run. Since the 10 grams was outside gas uncharged, it was subtracted from the weight of the gas charged to the feed cylinder prior to each run; and calculations were based on the corrected gas weight. To determine the amount of feed remaining in the feed system between the Jerguson gage and the top of the reactor,

the method used by Hamilton was adopted. Hamilton weighed the amount of oil required to fill the feed system between the Jerguson gage and the top of the reactor. The weight was found to be 72 grams.

### C. Materials

The gas oil in this investigation was a Berger, West Texas Virgin Gas oil obtained from Phillips Petroleum Company. Laboratory inspection data are given in Table I.

The outside gas used was a mixture of 30 weight per cent n-butane, 10 per cent iso-butane, 20 per cent iso-butylene, 20 per cent propane, and 20 per cent propylene prepared by the Matheson Company

The catalyst used in the investigation was pelleted Houdry synthetic aluminum silicate.

### SAMPLE CALCULATIONS

The calculations of the liquid space velocity, weight of permanent gases, average temperature of the run, weight of carbon deposited on the catalyst, over-all weight balance, yield of gasoline, conversion, ultimate yield and mol ratio for Run Number 33 are presented as typical of all runs made.

#### A. Calculation of Liquid Space Velocity:

Data:

Volume of catalyst in the reactor	=	1000	cc
Feeding time	=	8.75	min
Weight of charge	=	480	grams
Density of charge	=	0.8	gm/ml
Volume of charge	=	600	ml

$$\text{Space Velocity} = \frac{600 \text{ ml} \times 60 \text{ min/hr}}{8.75 \text{ min} \times 1000 \text{ ml}} = 4.04 \text{ hr}^{-1}$$

B. Calculation of Carbon from Regeneration

Data:

Time (min)	Air (Liters)	Percent Analysis		
		CO <sub>2</sub>	O <sub>2</sub>	CO
6	14	6.0	7.8	1.6
30	65	11.8	0	8.6
90	161	11.0	0	9.2
120	201	10.6	0	9.4
180	290	10.4	0	9.4
240	385	9.0	0	11.6
300	476	10.6	0	6.0
330	546	7.0	0	5.0
340	575	3.6	15.0	0
360	600	4.2	13.2	0
390	630	1.2	17.6	0

Average from Plot (Fig.2)

CO <sub>2</sub>	Percent CO	CO <sub>2</sub> +CO	(Liters) CO+CO <sub>2</sub>
3.8	0.8	4.6	0.64
9.4	5.2	14.6	7.45
12.3	9.5	21.8	20.9
10.8	9.3	20.1	8.05
10.4	9.3	19.7	17.5
9.6	10.9	20.5	17.8
9.8	9.2	19.0	17.3
8.8	5.2	14.0	9.8
5.3	2.7	8.0	2.32
4.0	0	4.0	1.0
2.0	0	2.0	<u>.6</u>
			103.36

$$\begin{aligned} \text{Weight Carbon} &= \frac{103.36 \times 634 \times 273 \times 12}{22.4 \times 760 \times 298} \\ &= 42.5 \text{ grms} \end{aligned}$$

C. Weight of Permanent Gases:

Data:

Volume of noncondensable gases	=	78	l
Volume of gas sample bottle	=	208.3	ml
Barometric pressure	=	634	mm
Room temperature	=	25	°C
Weight of bottle and gas	=	127.686	gm
Weight of evacuated bottle	=	127.479	gm
Weight of gas by difference	=	0.207	gm
Weight of 1 liter of gas at 25°C and 634 mm Hg	=	$\frac{0.207 \times 1000}{208.3}$	0.99 gm
Weight of permanent gases	=	77.6	gm

D. Calculation of Average Temperature:

Data:

Time (min)	Thermocouple Number			
	1	2	3	11
0	533	481	438	438
1	531	488	442	444
2	525	495	472	450
3	512	502	496	455
4	470	492	537	473
5	423	440	528	500
6	418	430	494	520
7	414	425	475	515
8	412	418	464	500
9	404	418	455	489
10	418	423	454	484
11	422	429	456	480
12	430	435	460	477
13	437	437	464	478

Average Temperature = 464°C

E. Calculation of Over-all Weight Balance:

Data:

Weight of Cylinder and Gas Oil	=	12484	gm.
Weight of Cylinder (Evacuated	=	12303	gm.
Weight of Oil in Cylinder	=	181	gm.
Weight of Cylinder, Gas Oil, and Gas	=	12865	gm.
Weight of Gas in cylinder	=	381	gm.
Weight of Gas remaining in Cylinder	=	10	gm.
Weight of Charge not reaching Reactor	=	72	gm.
Gas Oil Charged to Reactor $181(1 - \frac{72}{552})$	=	157	gm.
Outside Gas Charged to Reactor $371(1 - \frac{72}{552})$	=	323	gm.
Total Charge to Reactor	=	480	gm.
Recovered Material			
Condensable Gases	=	201.2	gm.
Permanent Gases	=	77.6	gm.
Product in Receiving Flask	=	155.7	gm.
Recovery	=	3.6	gm.
Carbon from Burn-Off	=	38.4	gm.
Total Weight Recovered	=	476.5	gm.
Weight of Losses by Differences	=	3.5	gm.
Per Cent Losses on Charge	=	0.73	%

F. Calculation of Per Cent Gasoline Yield:

Data:

Weight of oil charged	=	157	gm.
Weight of Gasoline from Distillation	=	58.8	gm.
Per Cent Gasoline on Oil Charged	=	$\frac{58.8}{157} \times 100$	38.4 %

G. Calculation of Per Cent Conversion

Data:

Weight of Oil Charged	=	157	gm.
Weight of Residue from Distillation	=	28.4	gm.
Weight of oil recovered from Catalyst Bed	=	3.6	gm.
Total Weight of unconverted Oil	=	32.0	gm.
Per Cent conversion on oil Charged	=	$100 - \left( \frac{32.0}{157} \times 100 \right)$	80.0 %

H. Calculation of Per Cent Ultimate Yield:

Data:

Per Cent Gasoline on Oil Charged	=	38.4	%
Per Cent Conversion on Oil Charged	=	80.0	%
Per Cent Ultimate Yield on Oil Charged	=	$\frac{38.4}{80.0} \times 100$	48.0 %

I. Calculation of Mol Ratio:

Data:

Weight of Oil Charged	=	157	gm.
Molecular Weight of Oil	=	250	gm/mol
Weight of Gas Charged	=	323	gm.

I. Calculation of Mol Ratio (Cont'd)

Data: (Cont'd)

$$\text{Molecular Weight of Gas} = 51.6 \text{ gm/mol}$$

$$\text{Mol Ratio} = \frac{323}{51.6} \times \frac{250}{157} = 9.97$$

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## RESULTS

Catalytic polyforming was investigated at mol ratios from 2 to 15.8. At each given mol ratio a series of runs was made with temperature as a variable, and the liquid space velocities relatively constant at 4 to 6 hr<sup>-1</sup>. The purpose of varying the temperature was to determine gasoline yield as a function of conversion. Plots were made of gasoline yield versus conversion to determine the conditions at which maximum gasoline yields could be obtained using the materials previously described. Data for all runs are listed in tabular form in the Appendix.

Runs 4, 5, 6, 7, 8, and 30 were made at a mol ratio of 2.3-3.7 using the mixed gas and a variation in temperature from 539° to 423°C. Permanent gas formation at this mol ratio was considerable at the higher temperatures. At 539°C, 161 grams of gas were formed, while only 13.1 grams of gas were formed at 423°C. Permanent gas formation was found to be greatest at the highest temperature for all mol ratios investigated. Carbon formation ranged from 4.84 per cent at 423°C to 18.9 per cent at 539°C. Percentages used in expressing carbon formation are based on the total charge.

At this mol ratio, 2.3-3.7, gasoline yields were found to vary from 37.1 per cent gasoline on oil charged at a conversion of 51.2 per cent on oil charged to 22.4 per cent gasoline at a conversion of 89.1 per cent. The plot of gasoline yield versus conversion for this mol ratio is shown in Figure 3. This plot was not fully determined in

the lower temperature range; but, since cracking decreases at temperatures below 400°C, it was assumed that a maximum had been reached in this case.

Runs 18, 19, 20, 21, 22, and 30 were made in the mol ratio range 4-5. Permanent gas formation and carbon laydown in this series of runs are substantially the same as in the case of the lower mol ratios. A plot of the data obtained from this mol ratio range is shown in Figure 5. There is no noticeable change in gasoline yield over that obtained at mol ratios of 2-3. Figure 6 is a plot of the ultimate yield from this series of runs.

Gasoline yields were found to increase slightly in runs 11, 12, 13, 14, 15, 16, 17, and 29 where mol ratios between 5 and 6 were used. The maximum gasoline production came at substantially the same temperature and conversion at which maximum gasoline was obtained in the case of the lower mol ratio. The plot of the gasoline data from these runs can be seen in Figure 7. Carbon laydown and ultimate yield are plotted in Figure 15 and Figure 8, respectively. It is interesting to note that the change in mol ratio had very little effect upon carbon laydown or the ultimate yield from the gas oil.

Runs 23, 24, 25, 26, 27, and 31 were made at mol ratios in the range of 6.5-7.5. These runs, as plotted in Figure 9, show no increase in gasoline yield over the yields obtained at a mol ratio range of 5-6. The maximum gasoline production occurred at a conversion near 60 per cent as did the maximum for the lower mol ratio ranges already discussed.

The series of runs, 32 through 43 inclusive, made at a mol ratio of 9-11.5 gave the highest gasoline yields recorded. The gasoline yields plotted against conversion, are shown in Figure 11. A maximum gasoline yield of 49.2 per cent occurred at a conversion of 61.9 per cent. The temperature 440°C, at which this high yield was obtained is in the same range at which maximums were obtained at the other mol ratios previously discussed. Permanent gas formation was relatively low at 32.8 grams, as was carbon laydown at 6.8 per cent. This series of runs was characterized by high yields of gasoline which ranged from 37.4 per cent to 49.2 per cent. The temperatures were varied from 360°C to 469°C with conversions ranging from 80 per cent to 42.4 per cent.

Two runs, 49 and 50, were made using mol ratios between 15 and 16. These runs were made at optimum conditions of conversion as determined by runs made at other mol ratios. These two runs indicate that there is no further advantage to be gained in gasoline yields by increasing the mol ratio above 11.5. The results of the two runs are shown plotted in Figure 13.

Runs 37, 44, 45, 46, 47, and 48 were made to determine the effect of pressure on catalytic polyforming employing high mol ratios. This series of runs was made at conditions of temperature previously determined as optimum. These conditions with the exception of pressure were held relatively constant throughout the pressure investigation. It was found, as shown in Figure 14, that pressure between 300 psig and 2000 psig had little effect upon catalytic polyforming employing

high mol ratios. The run made at atmospheric pressure was characterized by a low yield at a relatively low conversion, while the runs made at pressures ranging from 300 psig to 2000 psig gave high yields of gasoline and showed very little fluctuation in carbon laydown, ultimate yield, and permanent gas formation.

Mol ratio had very little influence on percentage of carbon laydown on the catalyst. There was a decrease in the percentage of carbon laydown at the higher mol ratios; this trend, however, is very slight. Carbon laydown was plotted against conversion and is shown in Figures 15 and 16.

Ultimate yield plotted against conversion is shown in Figures 4, 6, 8, 10 and 12. A comparison of the plots indicates that increases in mol ratio tend to increase the ultimate yield. In general, it was found, that ultimate yield increased and carbon laydown decreased with increases in mol ratio. Permanent gas formation remained largely a function of conversion.

Catalytic Polyforming data may be evaluated by comparing it with catalytic cracking data obtained in the same reaction unit using identical charge stock. Liquid space velocities used in catalytic polyforming were held constant at 4-6  $\text{hr}^{-1}$  while liquid space velocities in catalytic cracking were held constant at 0.7  $\text{hr}^{-1}$ . Catalytic polyforming space velocities calculated on gas oil charged were approximately 1.7  $\text{hr}^{-1}$  at a mol ratio of 10.0 and at lower mol ratios the space velocity increased accordingly. The gasoline fraction of the product was taken between 7 and 204°C in both processes.

Gasoline yields obtained in catalytic cracking ranged from 31.8 to 33.6 per cent on oil charged at conversions ranging from 52.4 to 71.6 per cent based on oil charge. These data are plotted in Figure 17. Catalytic Polyforming gave gasoline yields ranging from 45-49.2 per cent based on oil charged at identical conversion. Space velocities, based on oil charge, used in these catalytic polyforming runs were approximately 2.5 times those used in catalytic cracking.

SUMMARY

The results obtained in this investigation of catalytic polyforming may be summarized as follows:

- (1) Gasoline yields well in excess of those obtainable by conventional catalytic cracking may be obtained by catalytic polyforming using increased mol ratios.
- (2) Gasoline yields increase with each increase in mol ratio up to 9-11.5 when further increases of mol ratio up to 15.8 have no apparent effect upon gasoline yields.
- (3) Increases in mol ratio tend to decrease percentage of carbon laydown and increase ultimate yield.
- (4) Using mol ratios 9-11.5, greatest increases in gasoline yields occur between pressures from 0-300 psig.
- (5) Pressure increases between 300 and 2000 psig have no apparent effect upon gasoline yields at high mol ratios.

ACKNOWLEDGMENT

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APPENDIX

	Page
Table I - Gas Oil Inspection Data.....	34
Table II - Catalytic Polyforming Data, Runs 4, 5, 6, 7, 9, 10, 11, 12.....	35
Table III - Catalytic Polyforming Data, Runs 13, 14, 16, 17, 18, 19, 20, 21.....	36
Table IV - Catalytic Polyforming Data, Runs 22, 23, 24, 25, 26, 27, 29, 30.....	37
Table V - Catalytic Polyforming Data, Runs 31, 32, 33, 34, 35, 36, 37, 38.....	38
Table VI - Catalytic Polyforming Data, Runs 39, 40, 41, 42, 43, 44, 45, 46.....	39
Table VII - Catalytic Polyforming Data, Runs 47, 48, 49, 50.....	40
Figure 1 - Schematic Diagram of Catalytic Polyforming Unit.....	41
Figure 2 - Composition of Effluent Gas During Catalyst Burn-off.....	42
Figure 3 - Effect of Conversion on Gasoline Yield at 2.3- 3.7 Mol Ratio.....	43
Figure 4 - Effect of Conversion on Ultimate Yield at 2.3- 3.7 Mol Ratio.....	44
Figure 5 - Effect of Conversion on Gasoline Yield at 4.- 5. Mol Ratio.....	45
Figure 6 - Effect of Conversion on Ultimate Yield at 4.- 5. Mol Ratio.....	46
Figure 7 - Effect of Conversion on Gasoline Yield at 5.- 6. Mol Ratio.....	47
Figure 8 - Effect of Conversion on Ultimate Yield at 5.- 6. Mol Ratio.....	48
Figure 9 - Effect of Conversion on Gasoline Yield at 6.5- 7.5 Mol Ratio.....	49

APPENDIX (Cont'd.)

	Page
Figure 10 - Effect of Conversion on Ultimate Yield at 6.5- 7.5 Mol Ratio.....	50
Figure 11 - Effect of Conversion on Gasoline Yield at 9.- 11.5 Mol Ratio.....	51
Figure 12 - Effect of Conversion on Ultimate Yield at 9.- 11.5 Mol Ratio.....	52
Figure 13 - Effect of Conversion on Gasoline Yield at 15.- 15.8 Mol Ratio.....	53
Figure 14 - Effect of Conversion on Gasoline Yield at 9.- 11.5 Mol Ratio and Pressure from 0- 2000 psig.....	54
Figure 15 - Carbon Laydown on Catalyst Versus Conversion for 2.3- 6. Mol Ratio.....	55
Figure 16 - Carbon Laydown on Catalyst Versus Conversion for 6.5- 11.5 Mol Ratio...	56
Figure 17 - Effect of Conversion on Gasoline for conventional catalytic cracking.....	57

TABLE I

GAS OIL INSPECTION DATA  
(Borger, Texas, Virgin Gas Oil)

A. S. T. M. Dist. °F

First Drop	527
5% cond. 760 mm	555
10%	565
20%	577
30%	591
40%	605
50%	622
60%	639
70%	662
80%	693
90%	725
95%	740
End Point	742
Recovery	98 %
Residue and Loss	2.0 %
Gravity A. P. I.	36.0
Viscosity, SSU/100°F	53.6
Weight Per Cent Sulphur	0.31

TABLE II

## CATALYTIC POLYFORMING DATA

Run No.	4	5	6	7	9	10	11	12
Charge Stock				Gas Oil				
Type of Catalyst				Houdry				
No. of Runs on Catalyst	14	15	16	17	4	5	6	7
Volume of Catalyst				1000 cc				
Ave. Reactor Temp. °C.	423	494	480	426	473	539	507	488
Ave. Operating Pressure				1500 psig				
Space Velocity Hr. <sup>-1</sup>	4.28	3.24	4.82	4.12	5.04	5.03	4.50	4.00
Mol Ratio	3.67	2.39	2.41	2.34	2.61	2.50	5.77	5.40
Material Charged, gm.								
Outside Gas		Isobutane				Mixed Gas		
Outside Gas	265	173	158	155	182	163	272	253
Charge Stock	349	350	317	320	337	316	228	227
Total Charge	614	523	475	475	519	479	500	480
Hydrocarbon Liquid Product	294.3	179.4	147.3	242.4	147.9	85.5	127.6	119.9
Condensable Gases	198	173.7	153.4	168.7	199.2	110.5	182.7	180.2
Permanent Gases	13.1	93.7	88.9	19.2	107.4	161.4	114.7	111.8
Oil from Catalyst	15.6	10.3	29.4	7.6	8.9	4.3	7.8	6.3
Carbon by Burn-off	26.2	67.4	51.2	32.7	51.9	90.5	64.9	70.5
Losses by Difference, gm	5.2	1.5	-6.8	-4.6	10.24	26.8	2.3	8.7
% Losses on Charge	0.96	0.29	1.43	0.97	0.72	5.60	0.46	1.81
Distillation Data, gm.								
(7-240)°C. Gasoline	114.1	98.4	94.2	106.1	101.2	70.7	76.1	75.3
Residue	139.6	75.9	84.1	132.9	70.9	30.9	37.2	40.1
Condensable Gases	221.4	122.3	106.3	145.5	143.6	57.2	164.3	141.5
% (7-240) Gasoline	37.1	28.1	29.6	33.2	30.1	22.4	33.4	33.15
% Conversion	51.2	76.8	67.6	43.3	78.4	89.1	81.1	81.0
% Ultimate Yield (7-204)	72.4	36.6	43.8	76.7	38.4	25.1	41.2	40.9
% Carbon Laydown	4.84	12.88	10.8	6.9	20.95	18.9	12.94	14.7

TABLE III

## CATALYTIC POLYFORMING DATA

Run No.	13	14	16	17	18	19	20	21	
Charge Stock				Gas Oil					
Type of Catalyst				Houdry					
No. of Runs on Catalyst	8	9	11	12	13	14	15	17	
Volume of Catalyst				1000 cc					
Ave. Reactor Temp. °C	446	407	429	508	418	447	449	501	
Ave. Operating Pressure				1500 psig					
Space Velocity Hr. <sup>-1</sup>	4.03	2.82	2.90	4.67	5.53	4.87	5.20	5.20	
Mol Ratio	5.86	5.88	5.61	5.53	4.04	4.16	4.15	4.02	
Material Charged, gm.									
Outside Gas				Mixed Gas					
Outside Gas	285	271	267	262.0	219.0	224.0	226.0	216	
Charge Stock	235	223	230	230.0	263.0	261.0	264.0	261	
Total Charge	520	494	497	492.0	482.0	485.0	490.0	477	
Hydrocarbon Liquid Product	206.5	213.5	213.7	77.8	181.6	181.9	165.9	137.3	
Condensable Gases	227.4	214.8	205.7	181.9	219.9	206.2	225.4	168.8	
Permanent Gases	42.3	23.6	36.8	148.3	26.2	41.6	41.3	102.7	
Oil from Catalyst	5.9	10.7	8.9	11.6	17.8	18.8	16.5	4.4	
Carbon by Burn-off	44.1	30.2		67.2	27.1	36.1	38.8	55.1	
Losses by Difference	6	-1.2	3.3	5.2	9.4	0.4	2.1	8.7	
% Losses on Charge	1.15	0.24	0.67	1.05	1.95	0.08	0.45	1.82	
Distillation Data, gm.									
(7-204) °C, Gasoline	95.4	93.4	85	61.7	96.4	83.4	88.0	59.8	
Residue	64.4	90.4	67.7	31.0	96.0	85.5	79.3	47.0	
Condensable Gases	214.9	196.8	204.1	112.8	143.9	171.5	175.5	153.5	
% (7-204) Gasoline	40.5	41.8	37.0	26.8	36.6	32.0	33.33	22.9	
% Conversion	71.6	55.5	67.7	82.1	58.3	60.2	64.9	81.9	
% Ultimate Yield (7-204)	56.5	75.5	54.6	32.6	62.9	53.1	51.4	28.3	
% Carbon Laydown	8.50	6.13	5.75	13.65	5.61	7.45	7.92	11.5	

TABLE IV

## CATALYTIC POLYFORMING DATA

Run No.	22	23	24	25	26	27	29	30	
Charge Stock				Gas Oil					
Type of Catalyst				Houdry					
No. of Runs on Catalyst	18	19	20	21	22	23	25	26	
Volume of catalyst				1000 cc					
Ave. Reactor Temp. °C.	481	500	456	423	412	500	384	397	
Ave. Operating Pressure				1500 psig					
Space Velocity Hr. <sup>-1</sup>	5.07	3.82	4.64	4.88	4.65	4.51	4.62	4.97	
Mol Ratio	4.00	7.00	7.24	6.90	6.50	7.27	5.46	3.86	
Material Charged, gm.									
Outside Gas				Mixed Gas					
Outside gas	215.5	293	304	279	251	282	248	194	
Charge Stock	261.5	203	204	196	191	188	220	244	
Total Charge	477	496	508	475	444	470	468	438	
Hydrocarbon Liquid Product	141.7	72.9	190.9	204.0	177.0	100.0	249.1	222.8	
Condensable Gases	195.6	170.5	218.2	204.4	184.6	182	170.3	155.7	
Permanent Gases	80.5	168.6	42.4	14.4	21.2	120	6.7	9.85	
Oil from Catalyst	8.5	6.3	18.3	13.6	21.7	15.9	12.6	10.6	
Carbon by Burn-off	42.4	68.7	36.4	30.7	19.5	42.2	25.8	17.3	
Losses by Difference, gr.	8.3	9.0	1.8	7.9	10.0	9.9	3.5	11.8	
% Losses on Charge	1.74	1.81	0.36	1.66	2.31	2.10	0.75	2.70	
Distillation Data, gm.									
(7-204)°C. Gasoline	70.5	57.9	79.3	63.0	62.7	65.0	67.2	73.0	
Residue	61.2	32.7	68.5	91.6	86.8	34.0	140.2	121.0	
Condensable Gases	161.3	117.7	195.0	188.3	156.2	137.1	140.2	132.8	
% (7-204) Gasoline	27.0	28.5	38.9	32.2	32.8	34.6	30.6	29.6	
% Conversion	75.1	82.3	58.2	46.6	47.0	79.6	35.8	46.7	
% Ultimate Yield (7-204)	35.9	34.6	66.9	69.0	70.0	43.5	85.5	63.5	
% Carbon Laydown	8.88	13.84	7.17	6.47	3.95	8.95	5.5	3.82	

TABLE V

## CATALYTIC POLYFORMING DATA

Run No.	31	32	33	34	35	36	37	38
Charge Stock				Gas Oil				
Type of Catalyst				Houdry				
No. of Runs on Catalyst	27	28	29	30	31	32	33	34
Volume of Catalyst				1000 cc				
Ave. Reactor Temp. °C.	391	449	464	369	374	440	431	419
Ave. Operating Pressure				1500 psig				
Space Velocity Hr. <sup>-1</sup>	4.63	4.20	4.04	3.8	4.95	4.5	4.84	4.67
Mol Ratio	7.12	9.45	9.97	9.27	9.89	9.45	10.10	10.20
Material Charged, gm.				Mixed Gas				
Outside Gas				Mixed Gas				
Outside Gas	282	304	323	322	328	311	331	324
Charge Stock	192	156	157	169	161	156	159	154
Total Charge	474	460	480	491	489	467	490	478
Hydrocarbon Liquid Product	248.5	221.7	155.7	193.0	177.4	201.1	235.9	206.1
Condensable Gases	173.7	178.8	201.2	232.4	238.8	198.9	183.7	216.7
Permanent Gases	16.3	25.8	77.6	24.6	21.2	32.8	27.0	13.1
Oil from Catalyst	13.6	2.7	3.6	23.1	25.0	1.9	.9	5.5
Carbon by Burn-off	15.8	42.0	38.4	18.6	22.8	32.0	20.2	21.0
Losses by Difference, gm.	6.1	11.6	3.5	-0.3	3.8	0.3	14.3	15.6
% Losses on Charge	1.28	2.52	0.73	0.06	0.77	0.06	2.91	3.26
Distillation Data, gm.								
(7-204)°C. Gasoline	72.2	75.0	58.8	65.2	60.1	76.8	73.7	72.8
Residue	113.8	56.0	28.4	80.5	66.6	58.2	53.9	69.7
Condensable Gases	173.1	216.9	229.3	235.2	240.6	226.4	247.8	193.6
% (7-204) Gasoline	37.8	48.2	38.4	38.6	37.4	49.2	46.3	48.5
% Conversion	43.5	63.0	80.0	42.4	44.5	61.9	61.2	52.4
% Ultimate Yield (7-204)	87.0	76.7	48.0	91.0	84.2	79.5	75.6	92.5
% Carbon Laydown	3.35	9.1	8.0	3.8	4.7	6.8	4.12	4.4

TABLE VI

## CATALYTIC POLYFORMING DATA

Run No.	39	40	41	42	43	44	45	46
Charge Stock				Gas Oil				
Type of Catalyst				Houdry				
No. of Runs on Catalyst	35	36	37	38	39	40	41	42
Volume of Catalyst				1000 cc				
Ave. Reactor Temp. °C.	421	461	397	469	378	467	411	436
Ave. Operating Pressure			1500 psig			900	0	1200
Space Velocity Hr. <sup>-1</sup>	4.6	4.2	4.57	4.25	4.6	4.33	4.39	4.39
Mol Ratio	10.47	9.98	10.07	9.86	11.15	10.12	9.84	10.27
Material Charged								
Outside Gas				Mixed Gases				
Outside Gas	341	321	316	319	352	315	353	322
Charge Stock	158	156	152	157	153	151	174	152
Total Charge	499	477	468	476	505	466	527	474
Hydrocarbon Liquid Product	262.9	179.6	231.3	202.4	274.1	188	149.8	181.3
Condensable Gases	186.3	220.4	200.2	200.7	196.8	220.3	338.7	218.3
Permanent Gases	13.9	4.9	9.85	4.7	6.5	25.5	12.6	33.8
Oil from Catalyst	5.5	2.2	9.4	4.3	12.5	1.8	5.0	1.7
Carbon by Burn-off	21.2	33.5	21.8	32.5	20.0	31.6	15.9	30.3
Losses by Difference, gm.	9.0	-7.0	-4.2	-10.9	-4.0	-1.2	5.0	9.4
% Losses on Charge	1.8	1.5	.9	2.31	10.97	-0.2	.95	1.92
Distillation Data, gm.								
(7-204) Gasoline	73.8	74.9	67.5	72.8	66.8	72.7	44.3	74.2
Residue	63.8	33.4	70.8	35.1	80.7	40.6	107.2	45.7
Condensable Gases	255.1	250.7	152.6	261.3	299.5	267	290.5	260.2
% (7-204) Gasoline	46.6	47.8	44.5	46.3	43.5	48.0	25.5	48.8
% Conversion	58.3	78.4	50.5	77.2	47.3	72.6	35.5	69.2
% Ultimate Yield (7-204)	80.0	61.0	88.8	60.0	91.8	66.3	71.8	70.6
% Carbon Laydown	4.25	7.0	4.65	8.9	3.96	6.8	3.02	6.4

TABLE VII

## CATALYTIC POLYFORMING DATA

Run No.	47	48	49	50
Charge Stock		Gas Oil		
Type of Catalyst		Houdry		
No. of Runs on Catalyst	43	44	45	46
Volume of Catalyst		1000 cc		
Ave. Reactor Temp. °C.	455	428	440	421
Ave. Operating Pressure	300	2000	1500	1500
Space Velocity Hr. <sup>-1</sup>	4.85	4.43	4.17	3.96
Mol Ratio	9.85	9.73	15.12	15.77
Material Charged, gm.				
Outside Gas		Mixed Gas		
Outside Gas	321	311	415	432
Charge Stock	158	155	133	132
Total Charge	479	466	548	574
Hydrocarbon Liquid Product	166.3	228.3	138.0	221.6
Condensable Gases	248.7	164.8	339.8	252.8
Permanent gases	21.5	26.0	34.2	48.0
Oil from Catalyst	4.0	2.2	1.0	2.2
Carbon by Burn-off	23.0	25.0	27.9	29.4
Losses by Difference, gm.	10.8	18.7	7.1	10.0
% Losses on Charge	2.3	4.0	1.29	1.72
Distillation Data, Gm.				
(7-204)°C. Gasoline	70.2	72.1	59.3	62.2
Residue	48.0	55.0	49.3	51.3
Condensable Gases	274.1	230.9	335.6	313.8
% (7-204) Gasoline	44.4	46.6	44.6	47.1
% Conversion	68.6	63.5	62.2	59.5
% Ultimate Yield	64.6	73.5	71.8	79.3
% Carbon Laydown	4.8	5.37	5.10	5.21

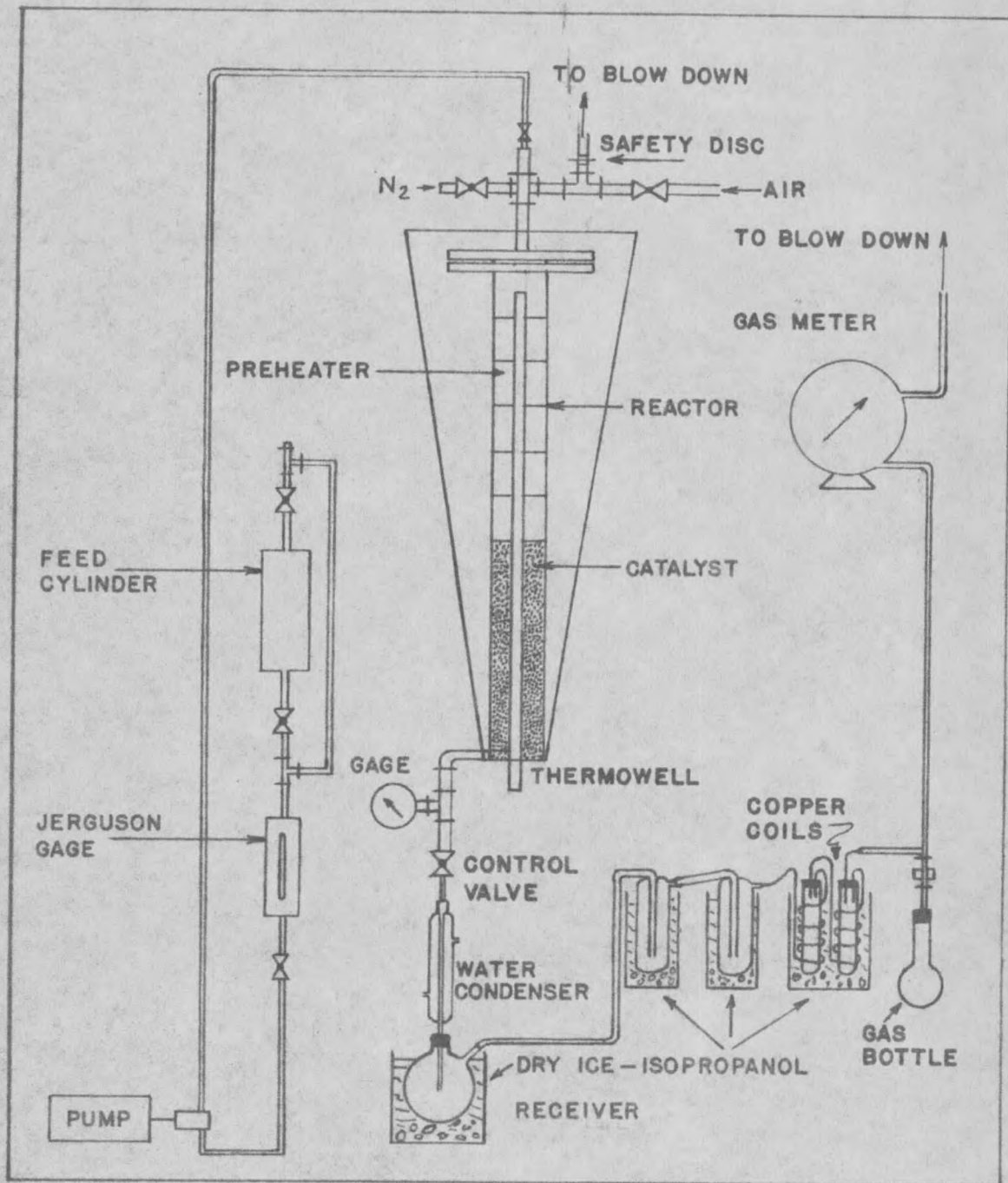


Figure 1 - Schematic Diagram of Catalytic Polyforming Unit

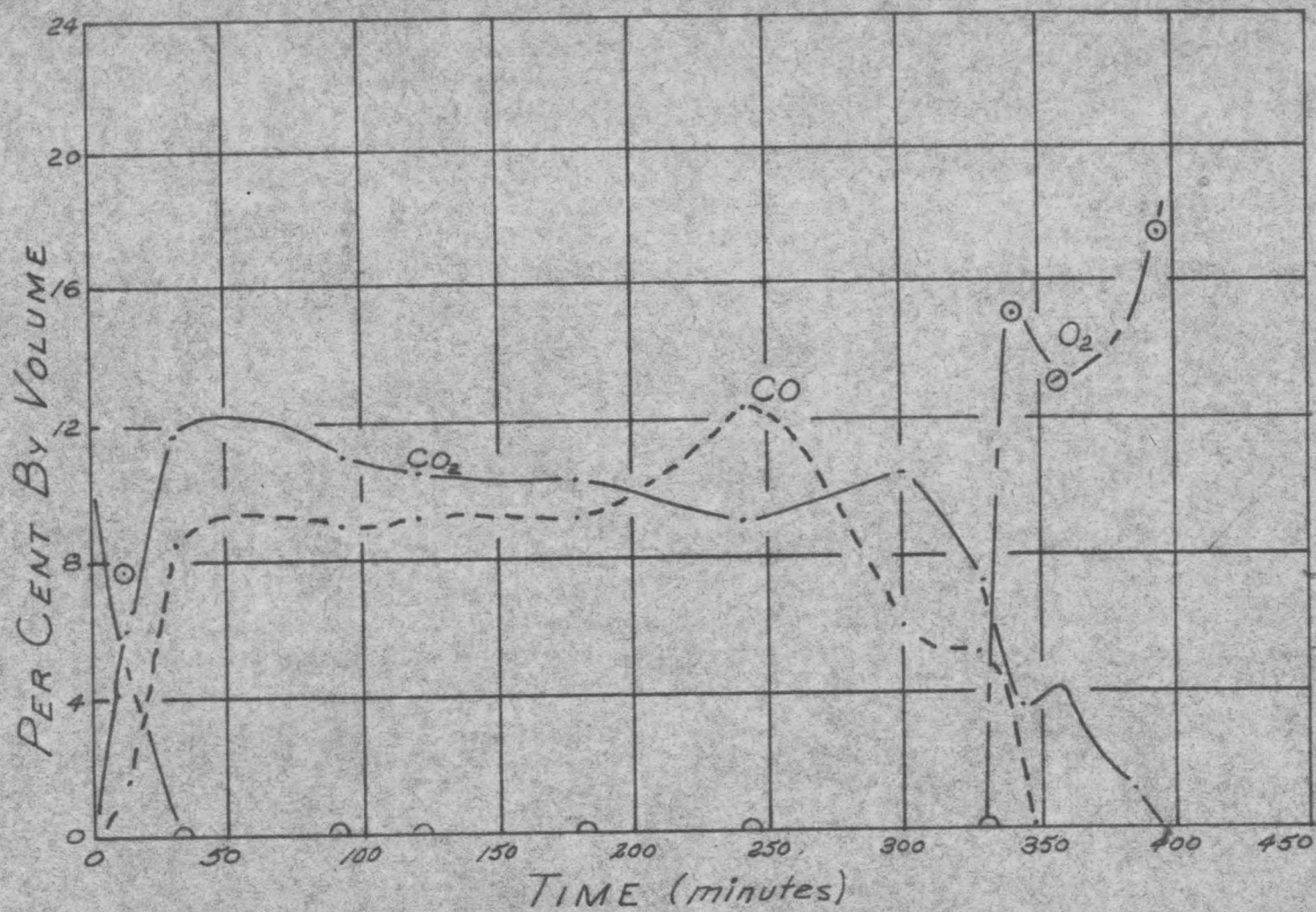


Figure 2 - Composition of Effluent Gas During Catalyst Burn-off.

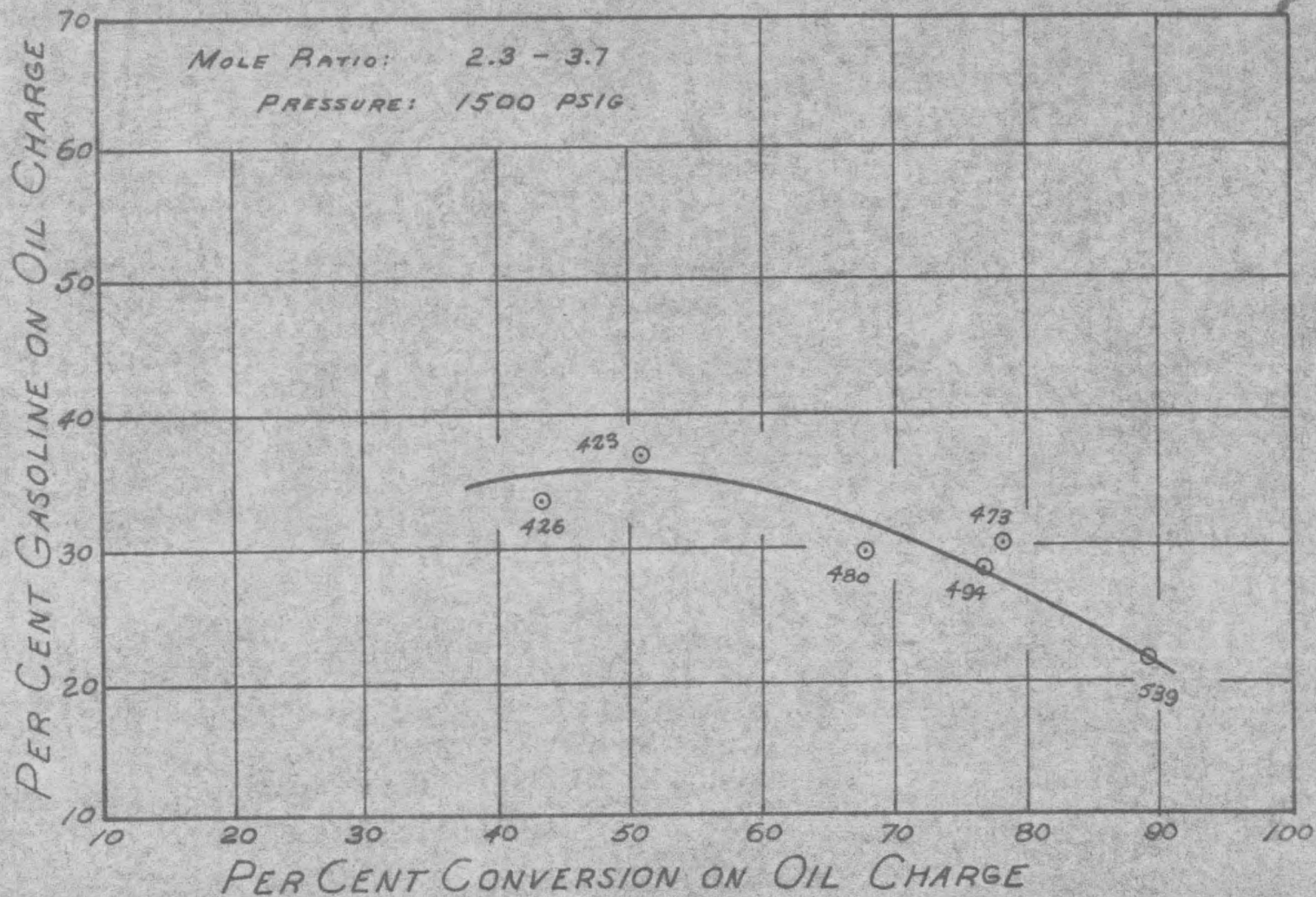


Figure 3 - Effect of Conversion on Gasoline Yield at 2.3-3.7 Mol Ratio.

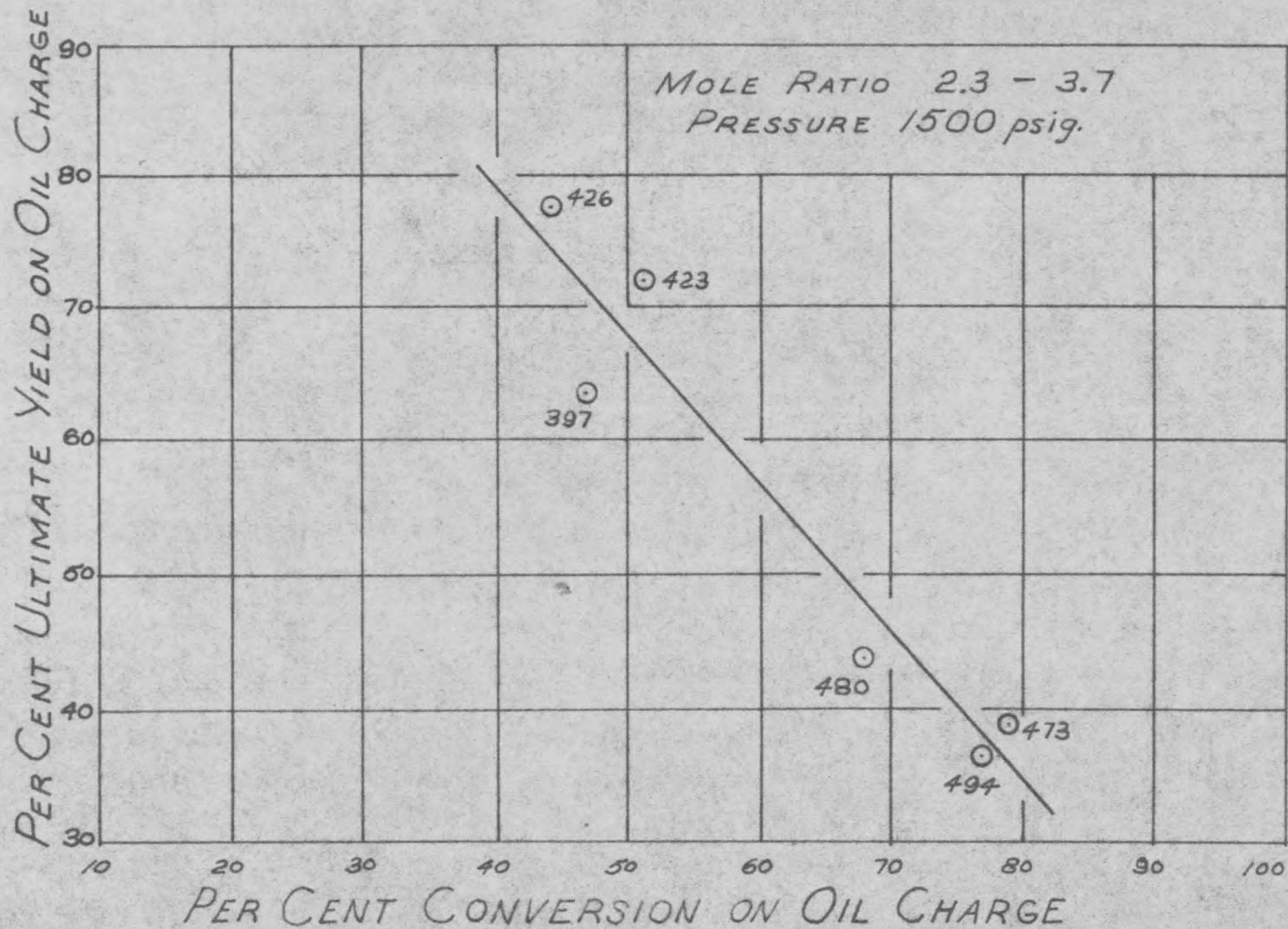


Figure 4 - Effect of Conversion on Ultimate Yield at 2.3-3.7 Mol Ratio.

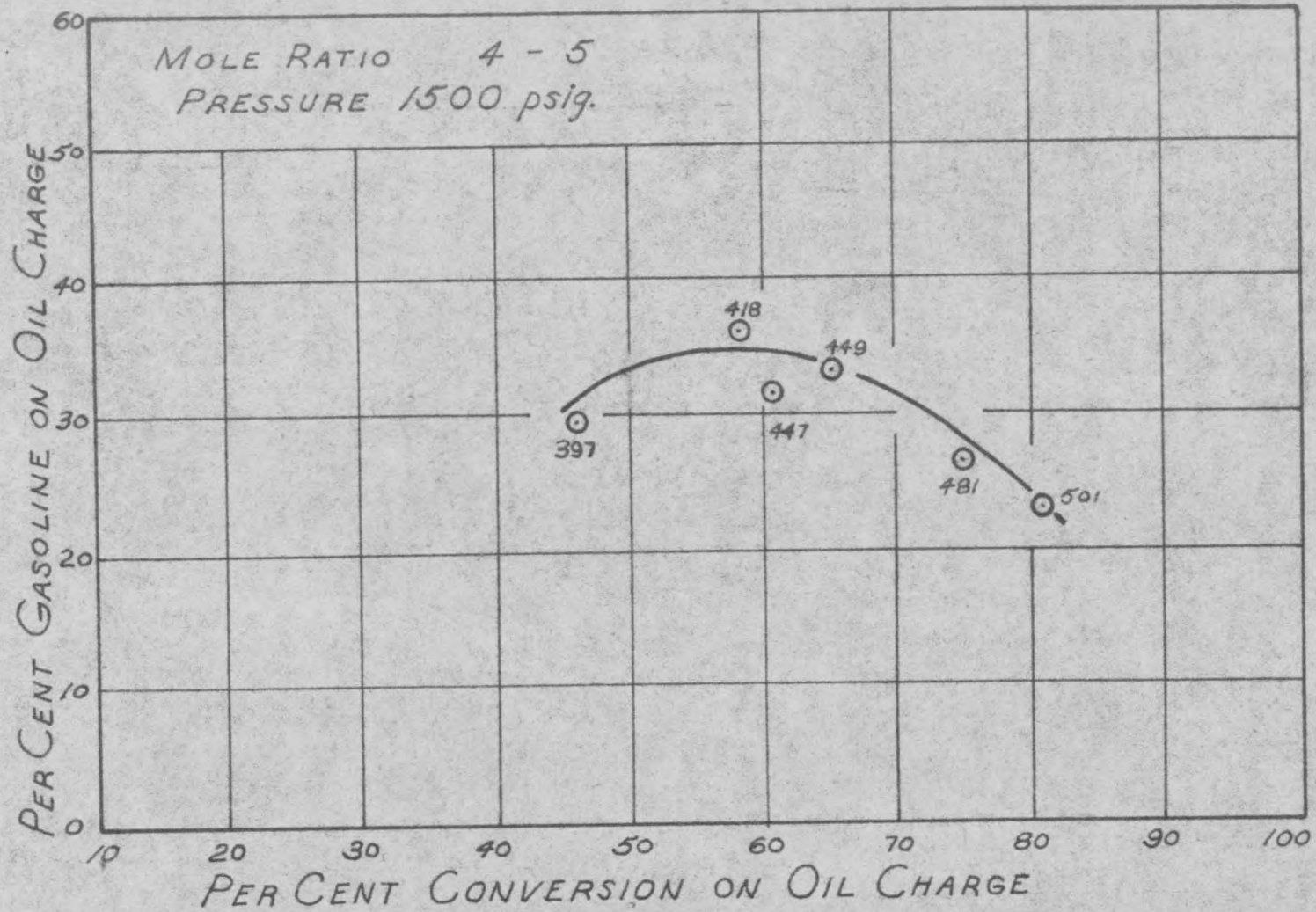


Figure 5 - Effect of Conversion on Gasoline Yield at 4.-5. Mol Ratio.

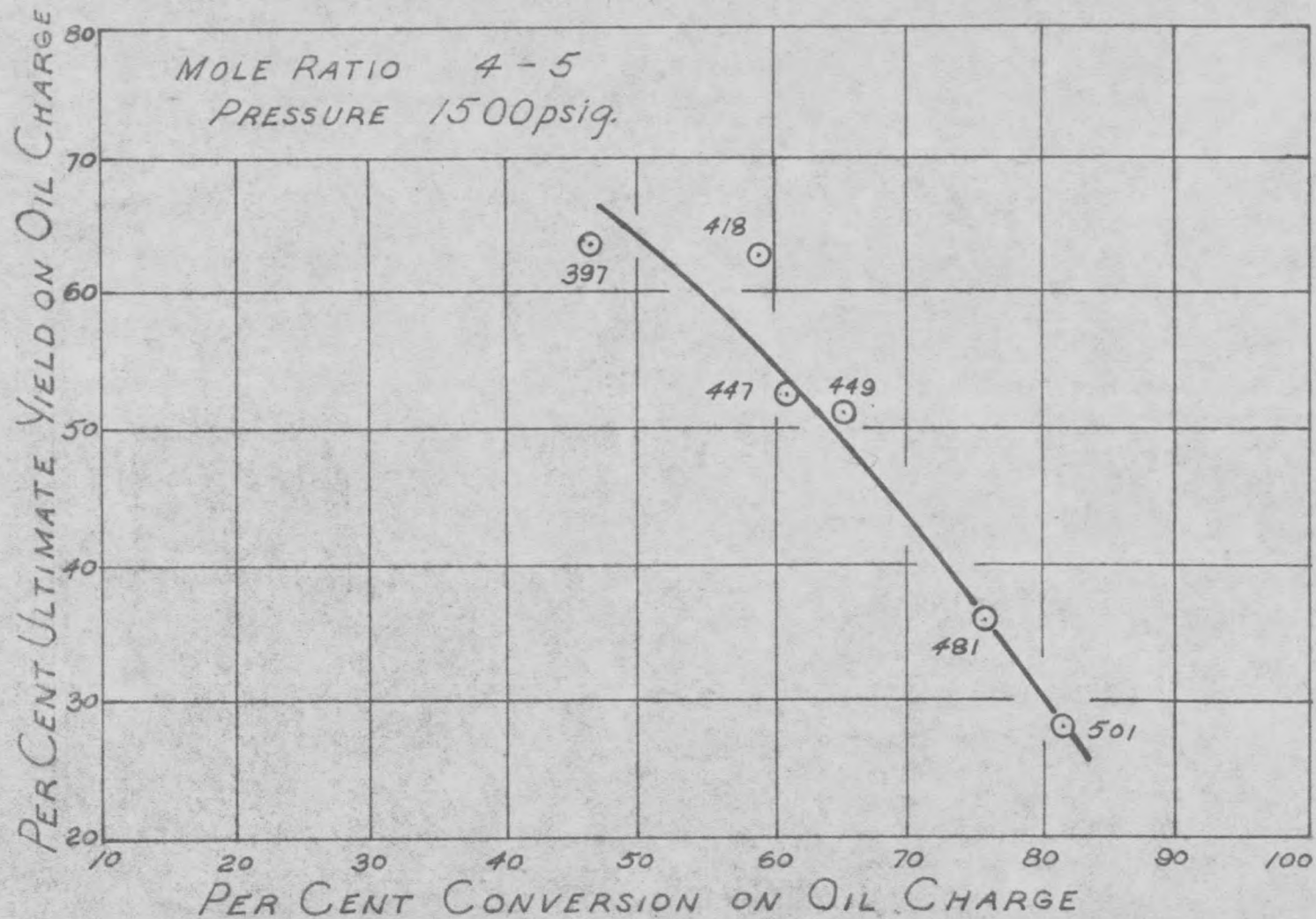


Figure 6 - Effect of Conversion on Ultimate Yield at 4.-5. Mol Ratio

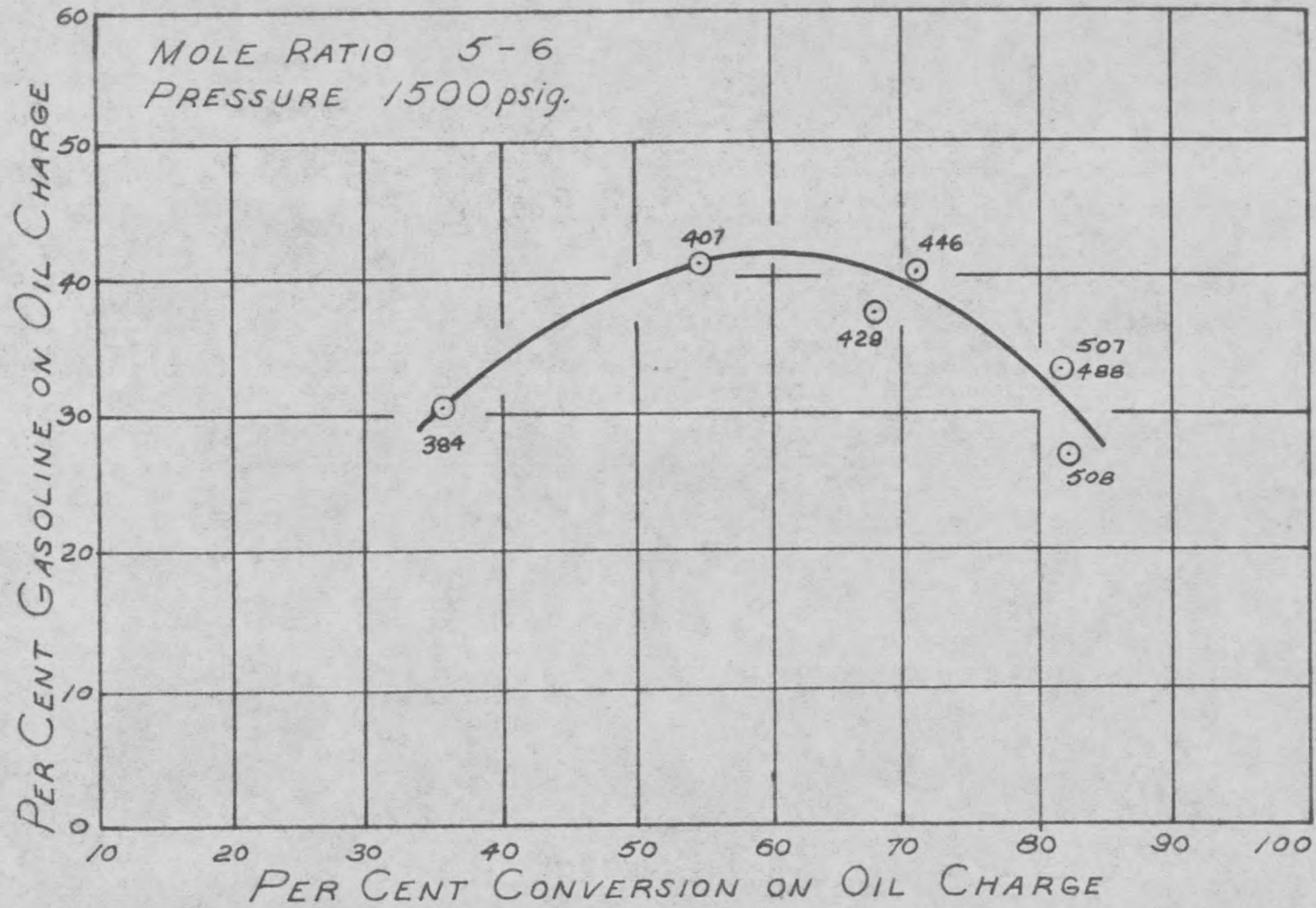


Figure 7 - Effect of Conversion on Gasoline Yield at 5.6. Mol Ratio.

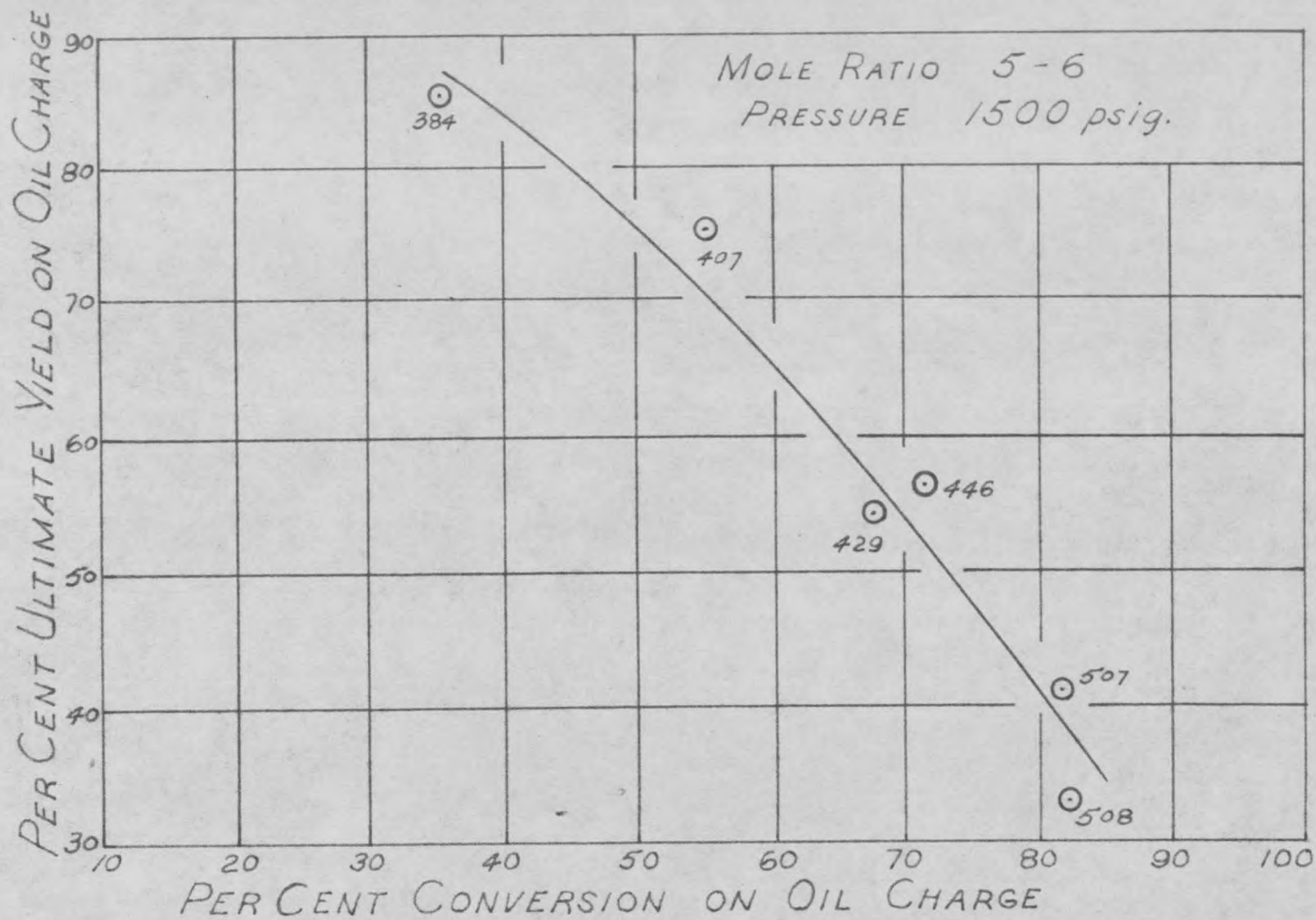


Figure 8 - Effect of Conversion on Ultimate Yield at 5.-6. Mol Ratio.

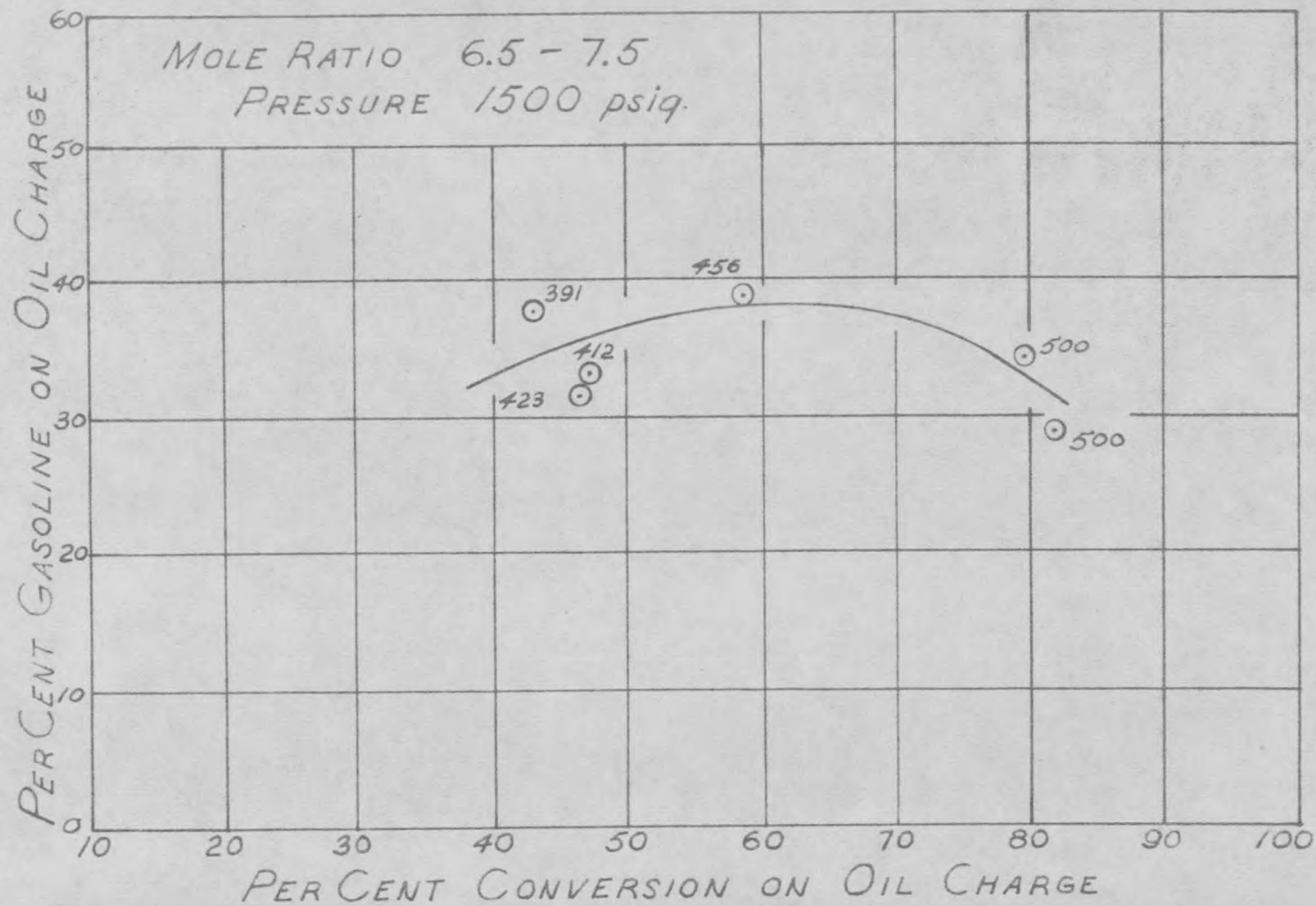
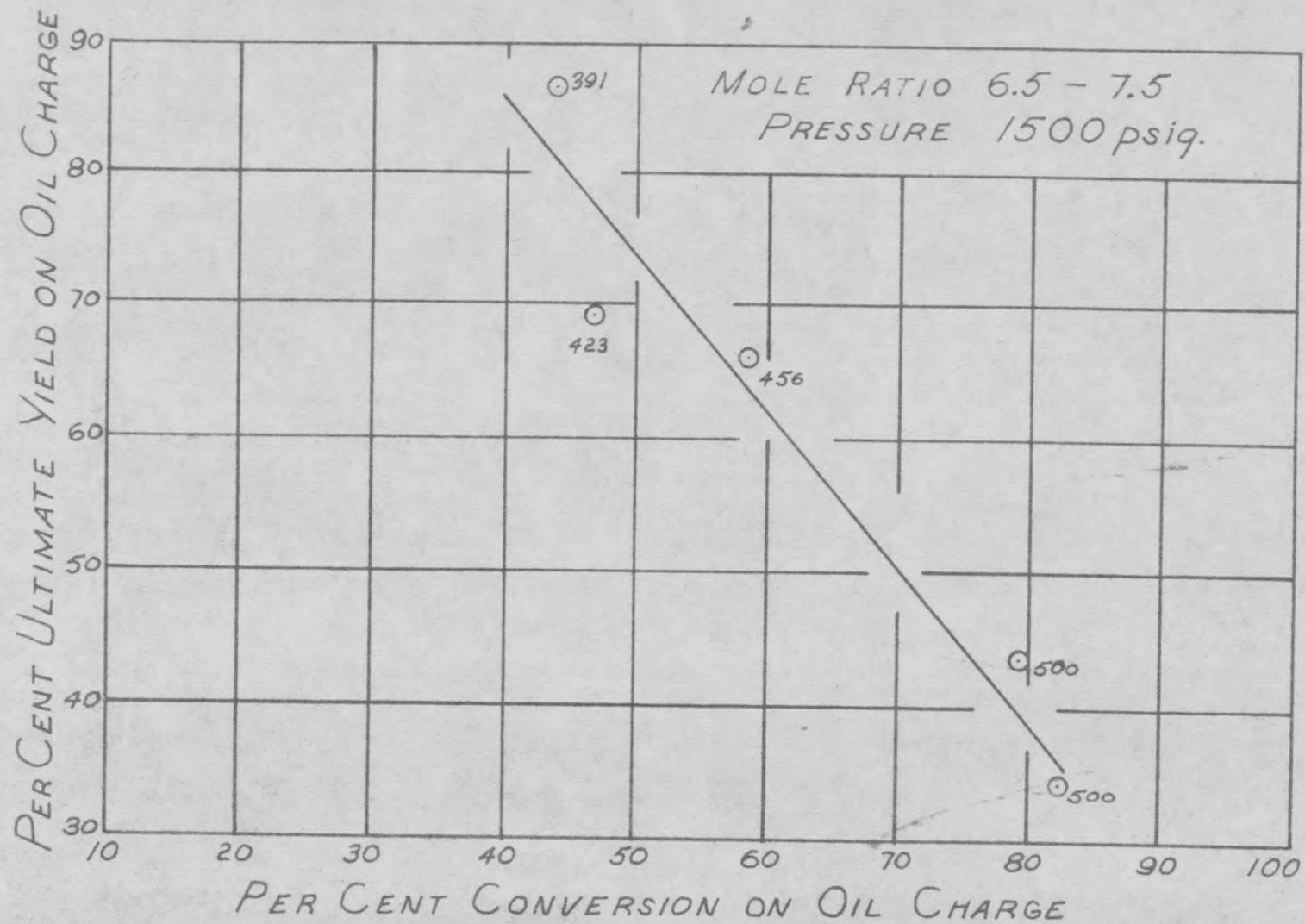


Figure 9 - Effect of Conversion on Gasoline Yield at 6.5-7.5 Mol Ratio.



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Figure 10 - Effect of Conversion on Ultimate Yield at 6.5-7.5 Mol Ratio

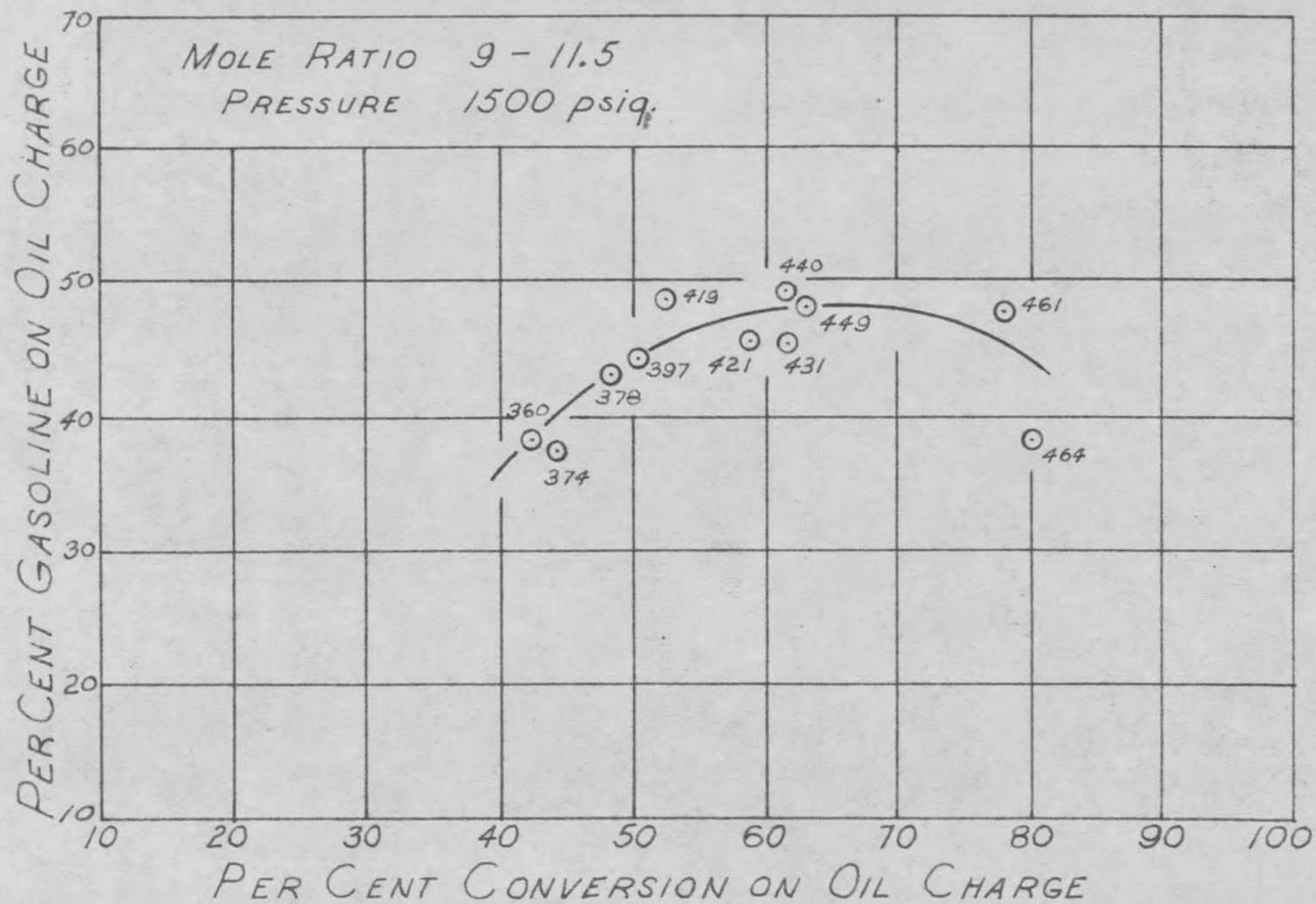


Figure 11 - Effect of Conversion on Gasoline Yield at 9.-11.5 Mol Ratio.

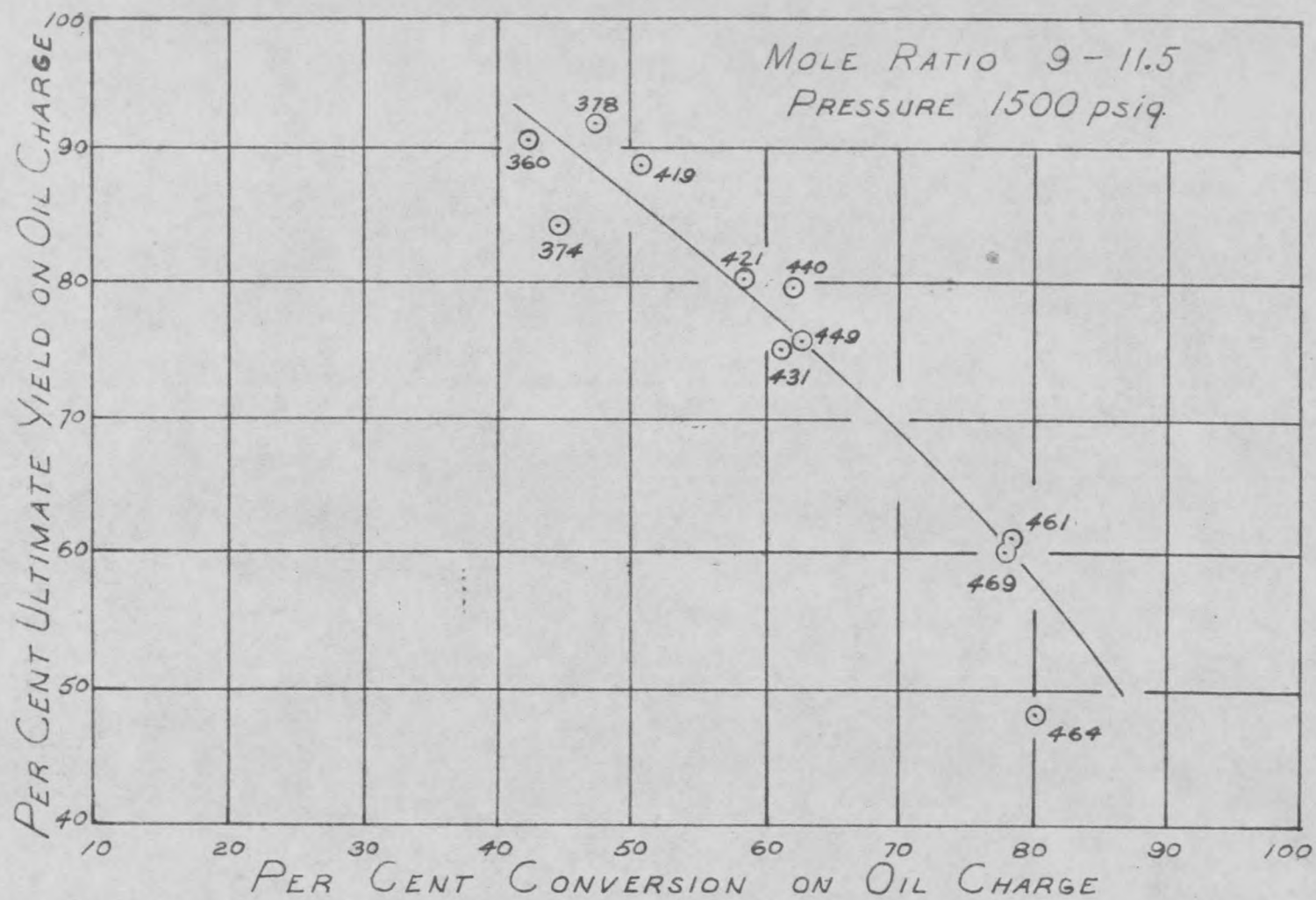


Figure 12 - Effect of Conversion on Ultimate Yield at 9.-11.5 Mol Ratio.

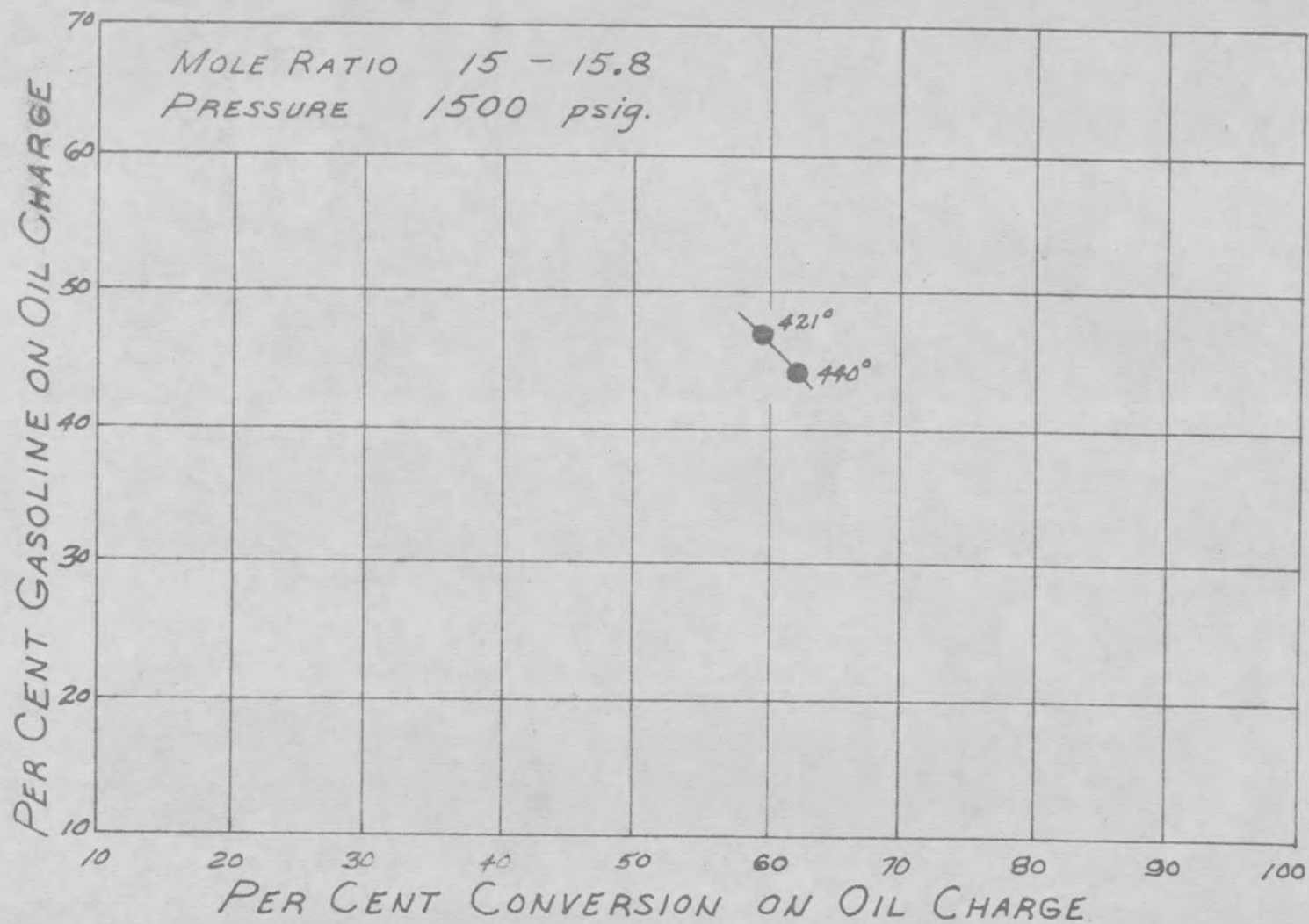


Figure 13 - Effect of Conversion on Gasoline Yield at 15.-15.8 Mol Ratio.

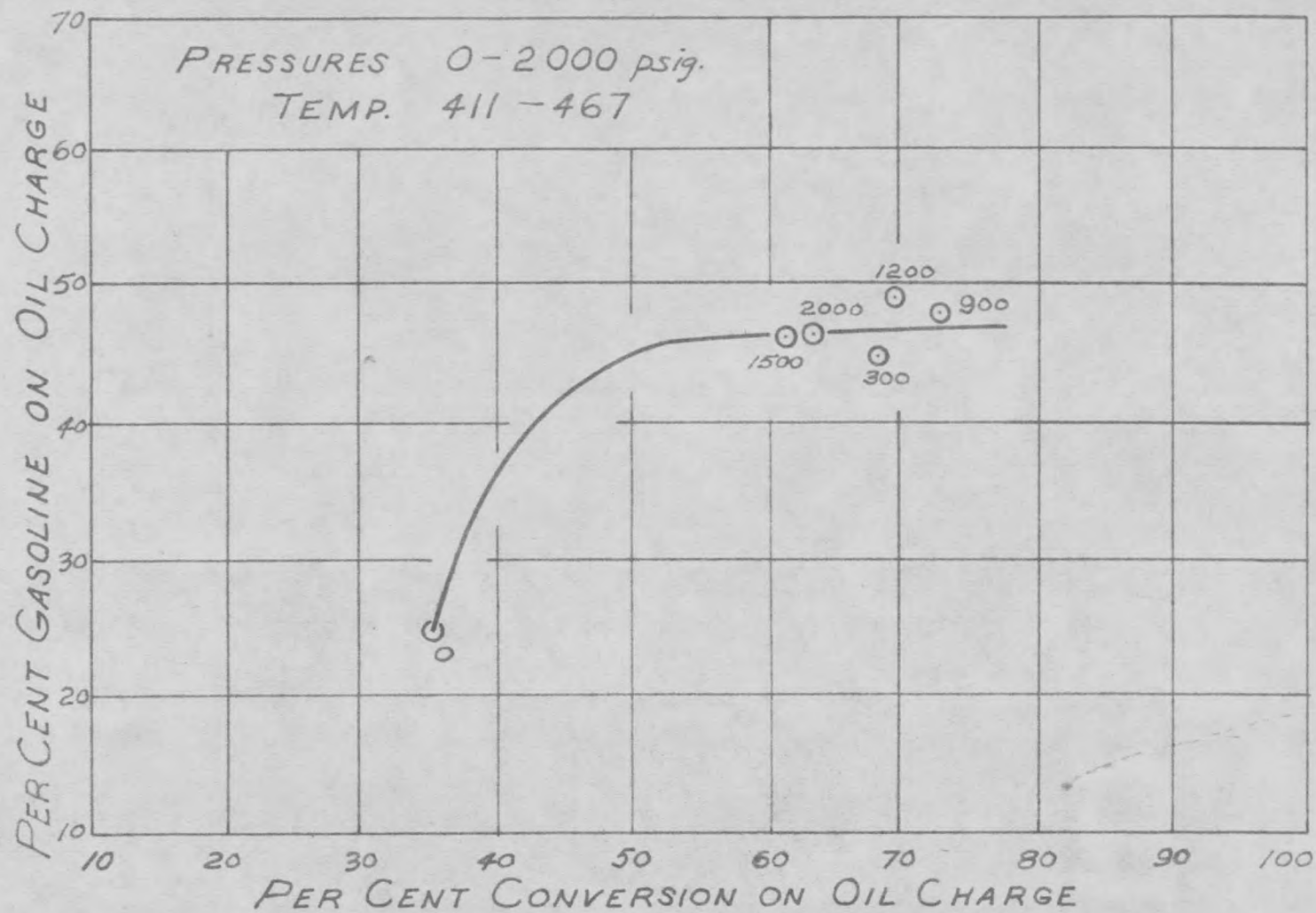


Figure 14 - Effect of Conversion on Gasoline Yield at 9.-11.5 Mol Ratio and Pressure from 0-2000 psig.

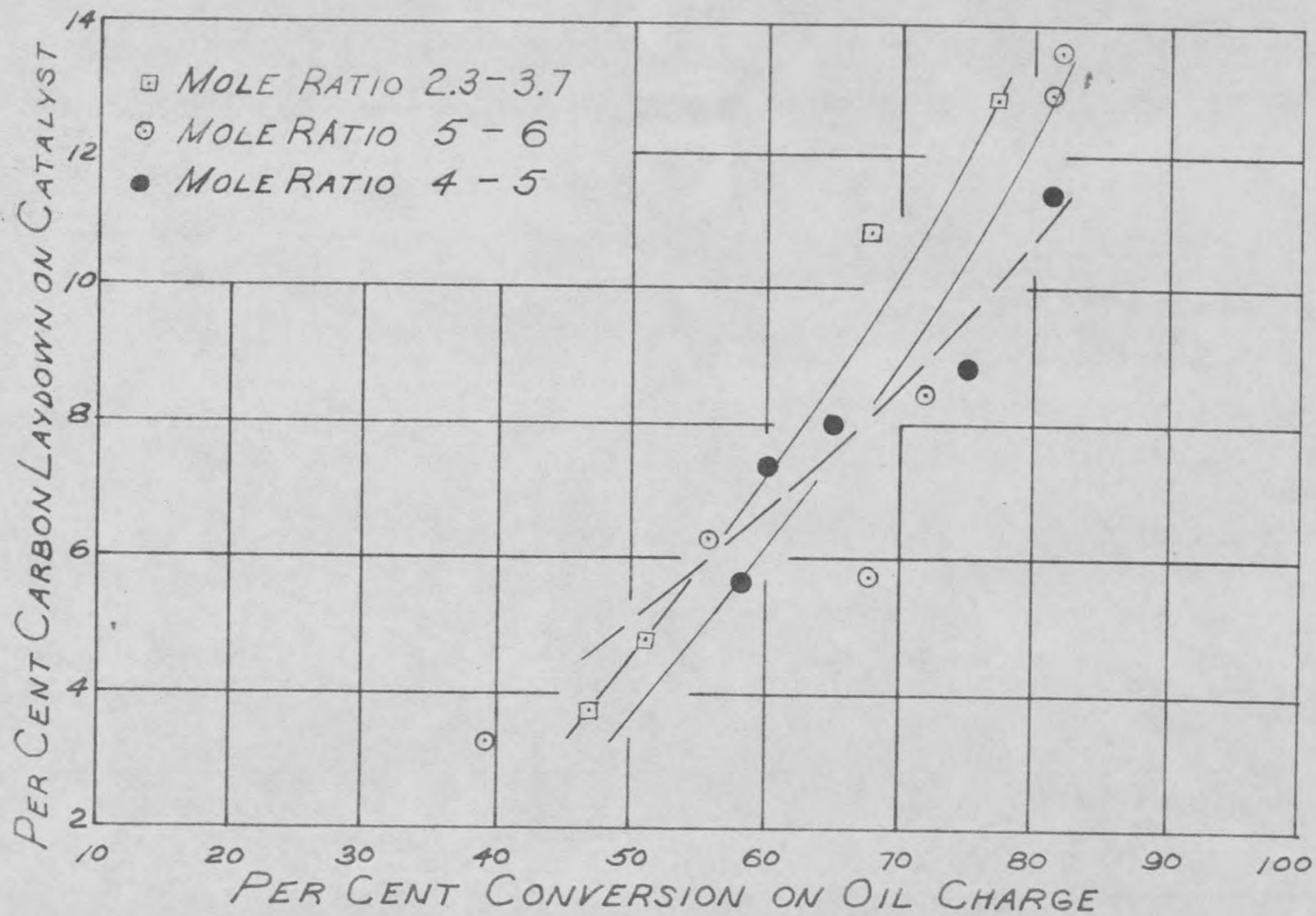


Figure 15 - Carbon Laydown on Catalyst Versus Conversion for 2.3-6. Mol Ratio

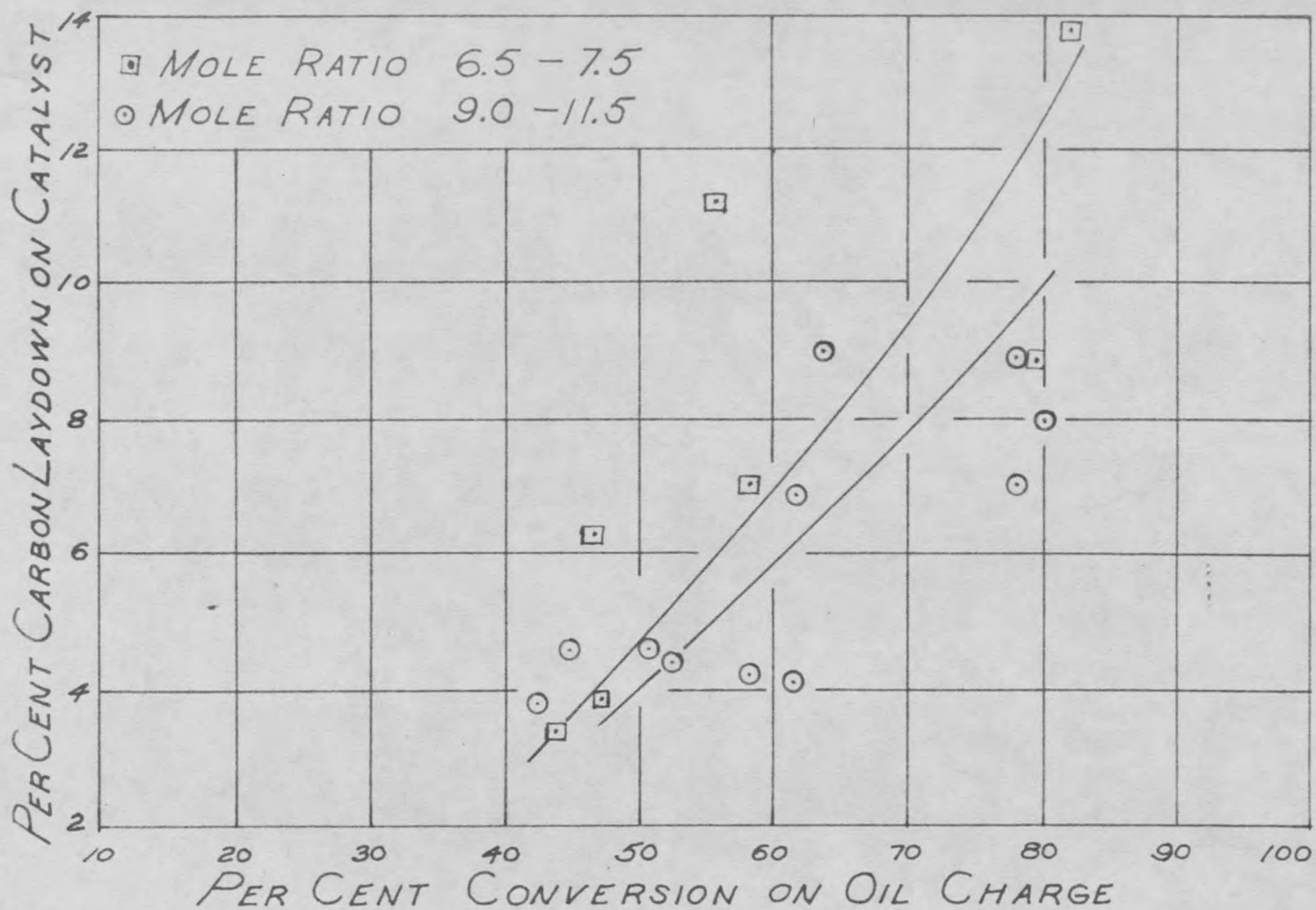


Figure 16 - Carbon Laydown on Catalyst Versus Conversion for 6.5-11.5 Mol Ratio.

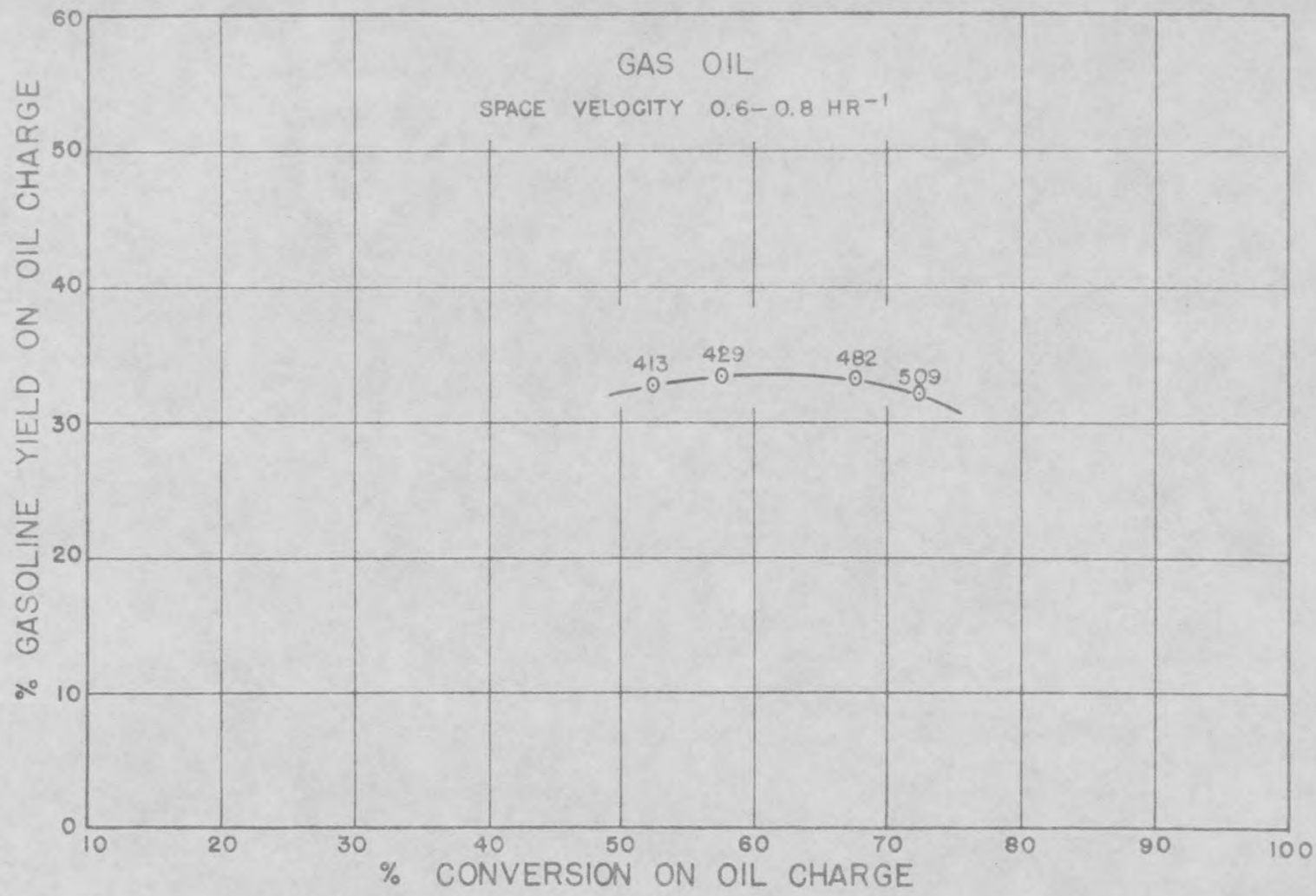


Figure 17 - Effect of Conversion on Gasoline for Conventional Catalytic Cracking.

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