

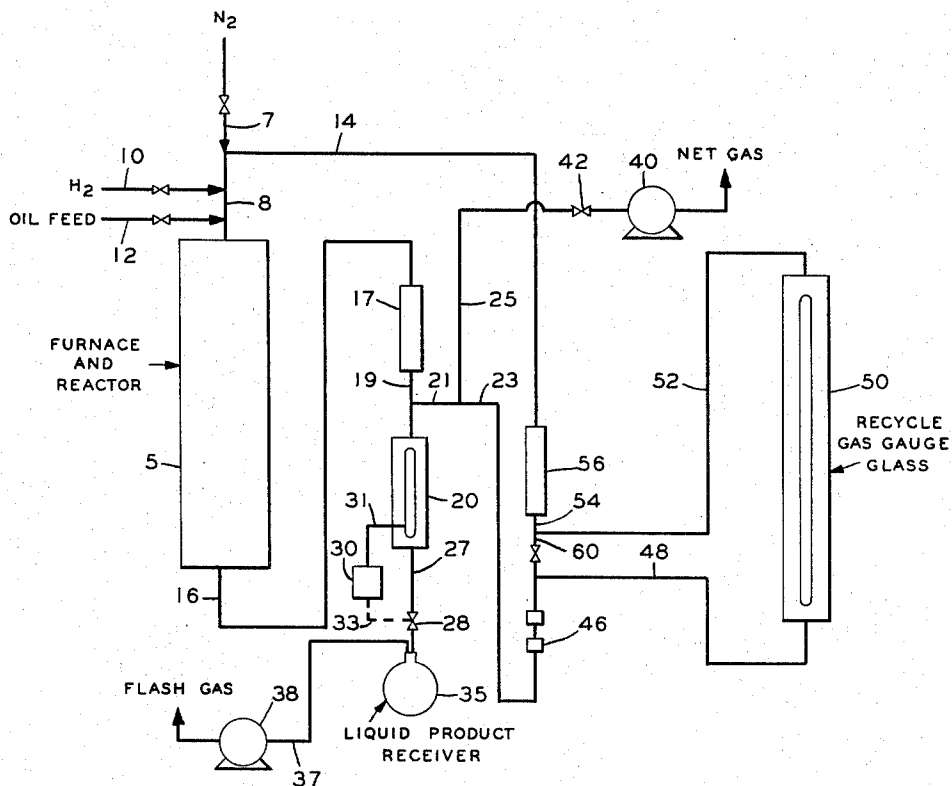
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W. P. BURTON ET AL

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METHOD OF STARTING UP A REFORMING PROCESS

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2,902,434

METHOD OF STARTING UP A REFORMING PROCESS

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This invention relates to an improved method for starting up a reforming operation for treatment of light hydrocarbon oils, and more particularly, it pertains to an improved method for starting up a reforming operation by means of an inert gas.

It has been the practice heretofore to employ hydrogen for starting up a reforming operation. As a result, it has been necessary to provide storage facilities for hydrogen or to undertake the expense of manufacturing large quantities of hydrogen for starting up the reforming operation. In any event, there is a need for a method of starting up whereby the cost is substantially reduced over the operation involving hydrogen as the material to be used for this purpose. Therefore, it is proposed hereunder to provide a method of starting up a reforming operation by the use of an inert gas thus eliminating the need for the use of hydrogen in this connection.

It is an object of this invention to provide an improved method of starting up a reforming operation by means of an inert gas.

Another object of this invention is to provide an improved method for starting up a reforming operation by means of nitrogen.

Still another object of this invention is to provide an improved method of starting up a reforming operation whereby a loss in product quality and yield is not significant in view of the economical saving incurred by reason of the inert gas.

Other objects and advantages will become apparent from the following description and explanation thereof.

By means of the present invention, it is proposed starting up a reforming operation by passing inert gas to a contacting zone containing a reforming catalyst, maintaining the temperature of the catalyst in the reforming zone at a level where the predominant reaction which can take place is dehydrogenation, passing a light hydrocarbon oil containing naphthenes in contact with the catalyst thereby effecting the dehydrogenation of the naphthenes to produce hydrogen, withdrawing from said contacting zone gaseous material containing hydrogen and inert gas until such time as the hydrogen concentration thereof is sufficiently high for use in a reforming operation, continuing the withdrawal of gaseous material from the contacting zone and adjusting the temperature and pressure conditions within the contacting zone for the desired reforming operation.

The reforming operation is started up by means of an inert gas which is readily available and inexpensive. An inert gas is one which has little or no effect upon the reforming operation, that is, it does not react with the hydrocarbon oil or poison the catalyst to the extent that its use is undesirable. An inert gas which can be used for this purpose includes, for example, nitrogen; CO₂; normally gaseous hydrocarbons, e.g., methane, ethane, propane, etc.; flue gas resulting from the combustion of carbonaceous material with oxygen containing gas; etc.

The starting up procedure of this invention is applicable to a system in which catalyst has been first regen-

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erated, and then it is intended to resume the reaction phase of the operation or this invention is applicable to situations in which the system is being started up for the first time or after a shut-down. The reforming system for which the present invention is effective involves fixed or moving beds in which the catalyst is of a fluid or non-fluid type. The fixed bed system of the non-fluid type may involve one or more reactors in which the reforming operation is conducted under non-regenerative or regenerative conditions. In the case of a non-regenerative system, the regeneration treatment is usually conducted by the "block-out" technique which involves essentially discontinuing the supply of oil to the system, purging the same of volatile combustible materials and then regenerating the reactors during this period of shut-down. The other type of regeneration is the "swing" regeneration and in this case one or more reactors remain on stream at all times; whereas one or more reactors are being regenerated at all times. In the fixed bed system, the catalyst is generally used in the form of a pellet or a pill having a diameter in the range of about $\frac{1}{32}$ of an inch to about $\frac{1}{4}$ of an inch and a length of about $\frac{1}{4}$ of an inch to 1 inch. Better contact between the hydrocarbon reactant and the catalyst is obtained by the use of small particle sizes.

The catalyst employed in the reforming operation can be one of the known types which possesses dehydrogenation-hydrogenation properties or aromatization properties. The catalyst is selected from several classes of materials including, for example, a compound of a group V or VI metal, e.g., the oxide or sulfide, etc.; a heteropoly acid in which the outer acid forming element is, for example, molybdenum, and the central acid forming element is, for example, phosphorus, e.g., phosphomolybdic acid; platinum or palladium catalysts; etc. The catalytic elements enumerated above are usually supported on a carrier material such as, for example, alumina, zinc spinel, silica-alumina, activated charcoal, pumice, kieselguhr, etc. In the finished catalyst, the catalytic element comprises about .01 to about 30% of the total material. Examples of catalysts which can be used for the reforming operation are molybdenum trioxide-alumina, platinum-alumina, etc.

In the starting up procedure, the catalyst in the reforming zone is maintained at a level where predominantly the dehydrogenation reaction occurs. The temperature is selected on this basis for the reason that it is desirable to produce hydrogen without subjecting the light hydrocarbon oil to reactions which are adversely influenced in the absence of hydrogen or in the presence of only small quantities thereof. The temperature range in which this purpose can be effected is, for example, about 650° to about 850° F., although it is preferred to employ a temperature in the range of about 700° to about 800° F.

After the catalyst is controlled at the desired temperature for the starting up operation and the inert gas is supplied to the reforming zone, the system is ready for the light hydrocarbon oil charge. At this point, there may be differences in the operation between a fixed bed system and a moving bed system. In the fixed bed system, the inert gas can be supplied to the reforming zone in a quantity sufficient to maintain either an atmospheric or super-atmospheric pressure which is lower than the desired operating pressure for the reforming reaction. In this case, the inert gas which is supplied to the system may be sufficient to provide a pressure of, for example, about 1 atmosphere to about 100 p.s.i.g. and the inert gas supply is stopped until other steps in the procedure necessitate recycle. However, it should be understood that higher pressures can be used with less satisfactory results. In a moving bed system, it is necessary to maintain a flow

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of inert gas through the catalyst in order to effect fluidization. Consequently, in this type of a system, the inert gas may be recycled continuously to the reforming zone after it has been vented therefrom. This technique can also be used in the case of a fixed bed system. Further, the pressure at which the reforming zone is maintained by means of the inert gas can also be used in a fluid-moving bed system.

The next step in the starting up operation involves charging hydrocarbon feed to the reforming zone. The hydrocarbon feed can be charged at the rate that it is normally used in the reforming operation or it can be at a rate which comprises at least 50% of the desired reforming rate and up to about 100%. At the conditions existing in the reforming zone, the naphthenes contained in the light hydrocarbon oil are dehydrogenated to produce hydrogen. Consequently, a reaction product is discharged from the reforming zone comprised of normally liquid material and a gaseous material containing hydrogen and inert gas. If the recycle gas system in a fixed bed operation is not in use at this point, it is now convenient to commence with this phase of the operation in order that the hydrogen partial pressure in the reforming zone can be steadily increased as hydrogen is produced from the dehydrogenation of naphthenes. Further, a portion of the gaseous material which is discharged from the reforming zone is continuously vented from the system at a rate which is approximately equal to the rate at which hydrogen is produced in the reforming zone through the dehydrogenation of naphthenes. The venting of gaseous material from the reforming system is continued until the hydrogen concentration in the gaseous product is sufficient for use in the desired reforming operation. The hydrogen concentration may be below the optimum value for this purpose, consequently, it can be at least 35 to 40% by volume in the gaseous material, preferably, it is at least 50% by volume and up to 95% by volume in any case. With the hydrogen concentration in the gaseous effluent at the desired value for use in the reforming operation, the venting of gaseous material is stopped, and thereafter, the system is allowed to increase in pressure to the desired reforming level. After the desired reforming pressure is reached, it is preferred to raise the temperature to the desired reforming level. Alternatively, the temperature and pressure to be used in the reforming operation can be raised simultaneously at this stage of the start up procedure, or the temperature can be increased to operating level.

The reforming operation involves a light hydrocarbon oil, e.g., gasoline, naphtha or kerosene, being contacted with a suitable reforming catalyst. The operation can be effected at a temperature of about 750° to about 1075° F., a pressure of about 25 to about 1000 p.s.i.g., a weight space velocity of about 0.05 to about 15, in the presence of hydrogen in the amount of about 500 to about 15,000 standard cubic feet of hydrogen (60° F. and 760 mm.) per barrel of oil feed (1 barrel equals 42 gallons), abbreviated as s.c.f.b. In a moving bed system, a catalyst to oil ratio, on a weight basis, of about 0.05 to about 10 can be employed. The operation can be either a regenerative or non-regenerative system in which the reaction cycle runs for at least about 1500 hours. The conditions of operation can be selected on the basis of effecting either a net production or a net consumption of hydrogen.

The catalyst becomes contaminated with carbonaceous material, consequently, it is necessary to regenerate by burning the carbonaceous material with an oxygen containing gas, e.g., air or diluted air. The regeneration can be effected at a temperature of about 500° to about 1150° F. and a pressure of about 1 atmosphere to about 1000 p.s.i.g. In the case of platinum catalysts, the initial stage of regeneration is preferably mild in order to effect a removal of carbon under conditions conducive to low water partial pressure and low temperature. Accordingly, the first stage may involve a low oxygen partial pressure

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of about 0.01 to about 3 p.s.i.a., or a regeneration gas containing about 0.1 to about 3% by volume of oxygen. The temperature of this treatment varies from about 500° to 850° F. The final stage of treatment involves severe conditions with an oxygen partial pressure of about 5 to about 200 p.s.i.a. The temperature is about 800° to about 1150° F., and the period of treatment varies from about 0.25 to about 15 hours.

In order to provide a better understanding of the present invention, reference will be had to the accompanying drawing which contains an illustration of a test unit employed for the purpose of evaluating the starting up procedure of this invention.

The reactor 5 is a vertical, cylindrical vessel having an internal diameter of 1 inch and a length of 4 feet. This reactor was filled with platinum catalyst containing about 0.6% by weight of platinum supported on alumina. The catalyst was employed in the form of extruded pellets having a diameter of about 1/16 inch and a length of about 1/8 inch. The catalyst bed weighed approximately 400 grams of which 30 grams was catalyst and 370 grams was 8-10 mesh tubular alumina and occupied about 2 feet of the reactor length. Heat is supplied to the reactor 5 by external means, not shown, and suitable thermocouples in the bottom and top of the reaction bed, not shown, are employed for the purpose of temperature indication. Nitrogen is supplied to the reactor 5 by means of a valved line 7, and thence, it flows through inlet manifold 8 which is connected to the top thereof. Hydrogen is supplied to the manifold 8 by means of valved line 10. Similarly, oil feed is supplied to the manifold 8 by means of a valved line 12. Recycled hydrogen is supplied to the reactor 5 by means of a line 14, which in turn, is connected to the inlet manifold 8.

The gaseous effluent from the reactor 5 is discharged from the bottom thereof by means of a line 16. This gaseous effluent is first allowed to pass through the top of a condenser 17, and then it is discharged from the bottom thereof by means of a line 19, which is connected to the top of high pressure separator 20. The normally gaseous product material is withdrawn from line 19 and it passes into a line 21 for further division into a recycle gas stream for passage through line 23 and the net yield of this product for flow through line 25. The liquid product in high pressure separator 20 is discharged from the bottom thereof through line 27, and thence it passes through a pressure control valve installed therein. A liquid level controller 30 is employed which has one connection, shown as line 31, installed within the bottom part of high pressure separator 20 and the second connection associated with pressure control valve 28. A constant level of product is maintained within high pressure separator 20 by venting the excess product material through line 27 and valve 28 into a liquid product receiver 35. By virtue of the reduction in pressure from super-atmospheric pressure to atmospheric pressure, gaseous product material is flashed therefrom, consequently, line 37 which is connected to the top of the product receiver 35 provides for the discharge of the flashed gaseous material. The quantity of flashed gaseous material is measured by means of a meter installed in line 37. The net production of gaseous material from the reforming operation is yielded through line 25. The quantity of this gaseous product material is measured by means of a meter 40 installed in this line. Prior to being measured in meter 40, the gaseous product material is depressurized to essentially atmospheric pressure by means of a valve 42 installed in line 25. The recycled gas stream flowing through line 23 is passed through a compressor 46 in which the pressure is raised sufficiently to overcome the pressure drop in the system. The compressed recycled gas stream flows from compressor 46 to line 48 and thence to a recycle gas gauge glass 50 when measurement of the rate is desired. Compressed gas is discharged from the top of gauge 50 by means of line 52 as the gas in line 48

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displaces a liquid in the bottom of gauge 50, and thence it flows through line 54 which is connected to the bottom of dryer 56. The dryer contains granular material for the purpose of absorbing moisture from the recycle gas stream. The dried recycle gas stream is then discharged from the dryer through line 14, previously discussed. The recycle gauge glass is used only intermittently in operation of the unit, the normally compressed recycle gas stream by-passes this gauge and passes through a valved line 60 directly through line 54, which is in turn, connected to the bottom of dryer 56.

For the purpose of evaluating the present invention, three procedures were employed which, for the purpose of convenience, will be designated as starting up procedures A, B and C. The details of these procedures are discussed below.

STARTING UP PROCEDURE A

In this starting up procedure, following regeneration, the unit was pressurized with nitrogen at an ambient temperature to 100 p.s.i.g. The reactor temperature was then increased to 700° F. while the recycle gas system was operated by recycling nitrogen which is discharged from the unit at a pressure of 100 p.s.i.g. and for a period of 7 hours. It should be noted that no fresh supply of hydrogen was supplied following the built up pressure of 100 p.s.i.g., but that the recycle gas system functioned merely for the recycle of the nitrogen. The oil feed was then charged to the system and as a result of the temperature being maintained at 700° F., dehydrogenation of the naphthenic compounds produced sufficient hydrogen to allow the pressure to increase to 200 p.s.i.g. While the pressure was increasing by virtue of the net production of hydrogen in the reaction system, the gaseous effluent from the reactor was continuously recycled for a period of about 2 hours. After the pressure of the reactor reached 200 p.s.i.g., gaseous material comprising a mixture of hydrogen and nitrogen was discharged from the system at a rate substantially equal to the amount of hydrogen which was produced by virtue of the dehydrogenation of naphthenes. During this purging or discharge of gas from the system, the temperature was maintained at 700° F. and the pressure was held at 200 p.s.i.g. This procedure was continued for a period of about 2 hours and thereafter the pressure in the reactor was allowed to increase to 500 p.s.i.g. and the temperature raised to 890° F. in a period of 4 hours. At this point, the unit was on stream.

STARTING UP PROCEDURE B

Following regeneration, the test unit was pressurized with hydrogen at the ambient temperature to 60 p.s.i.g. The reactor temperature was increased to 800° F. at a rate of 75° F. per hour while recycling hydrogen at a pressure of 60 p.s.i.g. The naphtha feed was charged to the unit and the pressure of the system was allowed to increase to 500 p.s.i.g. by reason of the hydrogen which was produced from the dehydrogenation of the naphthenes in the feed stock. While the pressure was increasing, the temperature was maintained at 800° F. with continuous recycle of hydrogen to the reactor. Thereafter, the temperature was increased to 890° F. at the rate of 40° F. per hour.

STARTING UP PROCEDURE C

Following regeneration, hydrogen gas was passed through the reactor at atmospheric pressure and ambient temperature in a once-through operation while the temperature of the reactor was being raised to 900° F. Using the hydrogen once-through technique, the catalyst was pretreated for 12 hours at 900° F. and atmospheric pressure. Following the pretreatment of the catalyst, the temperature was lowered to 890° F. and the pressure raised to 500 p.s.i.g. Thence, the naphtha feed was charged to the unit and it was considered to be on stream.

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The feed stock employed in the various starting up procedures is given below in Table I.

Table I

50	° API gravity -----	51.3
	ASTM distillation, ° F.:	
	I.B.P. -----	238
	10 -----	270
	50 -----	301
10	90 -----	342
	95 -----	355
	E.P. -----	400
	Aniline point, ° F. -----	138.0
	Octane No., CFRR -----	33.6
15	Aromatics, vol. percent -----	8.5
	Sulfur, weight percent -----	.002
	Watson factor -----	11.97
	Molecular weight -----	128

20 The results obtained by virtue of the various starting up procedures are given in Table II below.

Table II

Run No.	1	2	3
25			
Start up procedure -----	A	B	C
Temperature, ° F. -----	890	890	890
Pressure, p.s.i.g. -----	500	500	500
Space velocity, W _g /hr/W _c -----	3	3	9
H ₂ rate, mols/mol feed -----	6.0	6.0	6.0
30	Reaction period, hours -----	17	37
		8	8
	Liquid product, Octane No., CFRR -----	85.8	87.5
			84.9

It is to be noted that Starting Up Procedure A, involving the use of nitrogen as the inert gas, resulted in a liquid product octane number of 85.7. Starting Up Procedure B, involving the use of hydrogen, resulted in an octane number of 87.5 for the liquid product. In the case of Starting Up Procedure C, involving a hydrogen pretreatment prior to the process cycle, the octane number of the liquid product was 84.9. It can be seen, therefore, that the use of nitrogen as an inert gas for the starting up procedure is not detrimental to the reforming operation in the sense that the catalyst becomes poisoned in any way, thus the use of an inert gas for the starting up procedure is commercially attractive. While it is true that the use of hydrogen in the starting up procedure resulted in a higher octane number for the liquid product for a relatively longer period of operation, nevertheless, the saving which is effected by the use of nitrogen for this purpose completely offsets any minor advantage resulting from the use of hydrogen for this purpose. Further, it should be noted that the system under consideration is being operated under non-regenerative conditions, consequently, a short period of 17 or 37 hours is insignificant in comparison to the total number of hours which is generally expected from this type of operation, namely, at least 2000 hours of operation. It is apparent, therefore, that the use of nitrogen in the starting up procedure of this invention is commercially attractive by virtue of the low cost involved in this procedure and the slightly less attractive results to be obtained in the brief period following the starting up of the processing cycle.

Having thus provided a description of our invention along with specific examples thereof, it should be understood that no undue limitations or restrictions are to be imposed by reason thereof, but that the scope of the above invention is defined by the appended claims.

We claim:

1. A process which comprises passing an inert gas over a reforming catalyst in a contact zone, maintaining the temperature of the catalyst at a level where the predominant reaction which can take place is dehydrogenation, passing a light hydrocarbon oil containing naphthenes in contact with said catalyst while continuing the flow of inert gas, thereby effecting the dehydrogenation

of the naphthenes to produce hydrogen, withdrawing from said contacting zone gaseous material containing hydrogen and inert gas, continuing to introduce inert gas into said contacting zone until such time that the hydrogen concentration of the gaseous effluent is sufficiently high for use in a reforming operation, thereafter substantially decreasing the flow of inert gas into said contacting zone and during the above operation adjusting the pressure and temperature conditions for a reforming operation to produce a higher octane product than in the predominantly dehydrogenation reaction.

2. A process which comprises passing inert gas over a reforming catalyst in a contacting zone, maintaining the temperature of the catalyst at a level where the predominant reaction which can take place is dehydrogenation, passing a light hydrocarbon oil containing naphthenes in contact with said catalyst while continuing the flow of inert gas, thereby effecting the dehydrogenation of naphthenes to produce hydrogen, withdrawing from said contact zone gaseous material containing hydrogen and inert gas, decreasing the flow of inert gas into said contact zone in proportion to the increase in hydrogen concentration in said contacting zone until a substantial portion of the inert gas has been purged from the system and thereafter adjusting the pressure and temperature conditions for a reforming operation to produce a higher octane product than during said dehydrogenation reaction.

3. An improved method for starting up a reforming process which comprises passing an inert gas through a reforming catalyst in a contacting zone, adjusting the reaction conditions for predominately the dehydrogenation of naphthenes, passing a light hydrocarbon containing naphthenes while continuing the flow of inert gas in contact with said catalyst to dehydrogenate naphthenes and produce hydrogen, withdrawing product effluent containing inert gas and hydrogen from said contact zone and recycling the same to said contact zone, discontinuing the flow of inert gas to said contact zone, by separating nitrogen from said recycle gas in proportion to the increase in hydrogen concentration in the effluent gas until the hydrogen concentration is sufficiently high for a reforming operation, increasing the temperature and pressure to reforming conditions and continuing the process to produce a high octane product.

4. An improved method for starting up a reforming process which comprises dehydrogenating a light hydrocarbon oil containing naphthenes in a reaction zone in the presence of an inert gas to produce hydrogen, recovering inert gas and hydrogen from said reaction zone and recycling the same, continuously reducing the concentration of inert gas passed to said reaction zone in proportion to the increase in hydrogen concentration in the effluent until the hydrogen concentration is sufficiently high for use in a reforming operation, increasing the temperature and pressure to reforming conditions and continuing the process to produce a higher octane product than obtainable during said dehydrogenation step.

5. An improved process for initiating a reforming process, which comprises establishing dehydrogenation temperature and pressure conditions in a bed of reforming catalyst with an inert gas, passing a light hydrocarbon containing naphthenes with said inert gas through said catalyst at predominately dehydrogenating conditions to produce a dehydrogenation product effluent gas containing hydrogen and inert gas, separating hydrogen and inert gas from said dehydrogenation effluent and recycling the same until sufficient hydrogen is produced to replace the concentration of inert gas passed to the process and to permit raising the temperature and pressure to reforming conditions, thereafter adjusting the temperature and pressure to reforming conditions and continuing the process to produce a higher octane product than obtainable during said dehydrogenation reaction.

6. An improved method for starting up a reforming process which comprises passing an inert gas in contact with a reforming catalyst in a contact zone to raise the pressure and temperature conditions of the catalyst to predominately dehydrogenating conditions, recycling effluent gas recovered from said contact zone, passing a light hydrocarbon oil containing naphthenes with said inert gas to said contact zone to effect predominately dehydrogenation of said naphthenes to produce hydrogen, continuously venting from the system a portion of the recycle effluent gases at a rate approximately equal to the rate at which hydrogen is produced until the hydrogen concentration in the contact zone is sufficient for use in a reforming operation during the above operation, increasing the temperature and pressure of the contact zone to reforming conditions and continuing the process to produce a higher octane product than obtainable during said dehydrogenation reaction.

7. A method for starting up a reforming process which comprises passing an inert gas through a reaction zone containing a reforming catalyst, maintaining the temperature of the reaction zone at a level where the predominant reaction which can take place is dehydrogenation, passing a light hydrocarbon oil containing naphthenes in contact with said catalyst while continuing the flow of inert gas to produce hydrogen by dehydrogenation of said naphthenes, recovering effluent gases from said reaction zone and recycling effluent gases to said reaction zone, discontinuing the addition of inert gas to the process and continuously removing a portion of the effluent gases from the process in proportion to the increase in hydrogen concentration, and during the above operation adjusting the pressure and temperature conditions for a reforming process to produce a higher octane product than during said dehydrogenation reaction.

8. A method for starting up a reforming process which comprises passing an inert gas in contact with a reforming catalyst in a contact zone, maintaining the temperature of the catalyst at about 650° F. to about 850° F. and a pressure of about one atmosphere to about 100 p.s.i.g., recycling effluent gases recovered from said contact zone, passing a light hydrocarbon oil containing naphthenes with said inert gas in contact with said catalyst to produce hydrogen by dehydrogenation of said naphthenes, continuously removing from the process a portion of the recycle effluent gases at a rate approximately equal to the rate at which hydrogen is produced until the hydrogen concentration in the contact zone is sufficient for use in a reforming operation, increasing the temperature and pressure of the contact zone to reforming conditions and continuing the process to produce a higher octane product than obtainable during said dehydrogenation reaction.

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UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 2,902,434

September 1, 1959

William P. Burton et al.

It is hereby certified that error appears in the printed specification of the above numbered patent requiring correction and that the said Letters Patent should read as corrected below.

Column 6, Table II, fourth column thereof, under the heading "3", fourth line, for "9" read -- 3 --.

Signed and sealed this 17th day of May 1960.

(SEAL)

Attest:

KARL H. AXLINE

Attesting Officer

ROBERT C. WATSON

Commissioner of Patents