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SUITE 1000
1899 L STREET, N.W.
WASHINGTON, D. C. 20036

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**FLEXIBLE HYDROGEN PLANT UTILIZING MULTIPLE
REFINERY HYDROCARBON STREAMS**

by

Kimberly A. Kramer
Principle Process Engineer

and

Nitin M. Patel
Engineering Associate
Air Products and Chemicals Inc.
Allentown, Pennsylvania

Sanjeev Sekhri
Supervising Process Engineer
Kinetics Technology International Corporation
San Dimas, California

and

Mark G. Brown
Staff Process Engineer
Shell Oil Products Company
Martinez, California

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Flexible Hydrogen Plant Utilizing Multiple Refinery Hydrocarbon Streams

K.A. Kramer and N.M. Patel
Air Products and Chemicals Inc., Allentown, PA
S. Sekhri
Kinetics Technology International Corp., San Dimas, CA
M. G. Brown
Shell Oil Products Company, Martinez, CA

INTRODUCTION

Two trends are currently creating major changes in the worldwide refining industry. The first is a long-term trend (with periods of fluctuations) shifting to lower quality crudes from 1980 to the present. The second trend is increasingly stringent environmental regulations on transportation fuels evidenced recently with the implementation of the federal Clean Air Act Amendments (CAAA) (1993-2000) and California Air Resource Board (CARB) Phase II (1996) regulations. The result is a need for additional upgrading capability and operational flexibility to handle a variety of crudes, while still producing a constant stream of clean, light transportation fuels (in effect a double edged sword). The impact of California's environmental legislation has been heard around the world as evidenced by the US EPA adopting the 0.05% low sulfur diesel specifications in 1993; Europe in 1996; and several countries in Asia (Japan, Australia, S. Korea, Thailand) in the 1996-2000 time frame. (See Figure 1: "Road to Clean Motor Fuels" graph for details).

The US refiners have been challenged with investing tens of billions of dollars to reconfigure their refineries to produce cleaner and lighter transportation fuels. Shell Oil at its Martinez Manufacturing Complex has invested over \$1 billion itself for its Clean Fuels Project, currently in progress. Refiners face a multitude of potential technology solutions and integration options into their existing operations. Additional hydrogen processing is a key ingredient in meeting more strict environmental legislation (e.g. CARB Phase II regulations) which continues the trend of more stringent gasoline specifications from three environmentally related specifications to eight.

As a result, there has been a significant growth in new and supplemental hydrogen requirements for refineries worldwide. The first wave of hydrogen plants was concentrated in California and the US Gulf coast (primarily as a result of the two driving forces already mentioned). The second wave is underway as demonstrated by hydrogen capacity being selectively added in other regions of the US and Europe. Additional hydrogen capacity will also be required in Asia, where the high growth economies in this region require increased conversion to meet the rapid growth in the

demand for transportation fuels. Refiners and industrial gas companies, such as Air Products, have already invested over \$1 billion worldwide to meet this new hydrogen production demand and we expect these trends to continue well into the next century.

In 1992, recognizing the environmental legislation impacts and crude oil trends, Air Products formed an alliance with Kinetics Technology International Corporation (KTI), a leading supplier of hydrogen plants to the refining industry. The hydrogen plant combines the operating, engineering and construction experience of Air Products and KTI. In addition to the Shell Martinez "over the fence" (onsite) hydrogen supply arrangement, several other major refiners have awarded Air Products and KTI the opportunity to supply hydrogen facilities in California and the US Gulf Coast for their expanded refinery hydrogen requirements. Air Products extensive hydrogen experience includes the operation of 33 steam methane reforming plants, 16 offgas hydrogen facilities, and 3 major hydrogen pipeline systems worldwide, with a combined capacity of 850 MMSCFD by the end of 1996. KTI's experience in the design of more than 160 steam methane reforming (SMR) plants contributes significantly to the joint experience base.

Numerous processes are available to produce hydrogen, however, steam reforming is still the dominant and currently preferred process because it can economically process a variety of refinery feedstocks into hydrogen. This paper will discuss the Air Products 88 MMSCFD hydrogen plant built by KTI, adjacent to Shell's Martinez refinery, which utilizes up to eight separate refinery hydrocarbon streams as feed and fuel for the production of hydrogen in the steam reforming unit. The integration of refinery hydrocarbon purge streams, normally sent to fuel, allows greater flexibility in refinery operations and increases the overall refinery fuel efficiencies.

The hydrogen plant also incorporates a number of process control design features to enhance reliability, such as "two out of three" voting systems, in-line sparing, and reduced bed PSA operation. The final section of the paper describes the environmental features of the plant required for operation in the Bay Area Air Quality Management District (BAAQMD). Air Products and KTI designed BACT features into the hydrogen plant to minimize emissions from the facility.

DESIGN FEATURES

The hydrogen plant produces 88 MMSCFD of hydrogen at 2000 psig and approximately 250,000 lbs/hr of 650 psig superheated steam for export to Shell. Up to eight different refinery hydrocarbon streams are used as feed and/or fuel to the hydrogen plant. A description of the process design follows which highlights those features which are unique to meeting the requirements of the Shell refinery. For reference, a simplified process flow diagram is included in Figure 2 and a photograph of the facility is included in Figure 3.

Feed Compression

The hydrogen plant is designed to handle a blend of refinery gases as well as a maximum of 2050 barrels per day of Liquefied Petroleum Gas (LPG) as feed. The feed gas is delivered to the hydrogen plant in one line and the blend is totally controlled by the refinery. This blend can vary on a seasonal or even a daily basis, depending on the demands on Shell's refinery. An on-line gas chromatograph continuously monitors the feed composition in order to fine tune critical operating parameters such as the steam to carbon ratio. Table 1 represents the composition of individual refinery hydrocarbon streams that go into the feed gas blend. Table 2 illustrates four typical feed gas blends that Shell expects to provide.

To enhance onstream reliability, two multiservice compressors are operated in parallel. Each compressor combines feed gas and hydrogen product compression duties and is designed for a capacity of 75% on both circuits. The Distributed Control System (DCS) automatically loads and unloads the compressors to optimize power consumption as plant rates change.

Selection of this multiservice design resulted in lower power consumption and required less plant real estate. Steam methane reforming produces a greater standard volume of product than hydrocarbon feed; therefore, for a given required hydrogen product pressure, overall compression power is reduced as the hydrocarbon feed pressure is maximized. The multiservice compressor arrangement was a good match with these required duties and also resulted in a more compact compression skid as compared to separate services. Figure 4 depicts the compression scheme in more detail.

Blended feed gas from Shell is compressed to above 600 psig. During the time that excess LPG is available from the refinery or when one of the multiservice compressors is down, the LPG is pumped to a steam heated LPG vaporizer and vaporized LPG is blended, under flow control, into the refinery feed gases.

Feed Purification

Feed gas is preheated in a series of exchangers, against low and high temperature shift effluent, before entering a hydrogenator reactor. Several of the refinery streams contain some level of olefins and sulfur. Olefins tend to thermally crack and form carbon in the top portion of the reformer tubes; organic sulfur will poison the reformer catalyst. To prevent this, all of the olefins and organic sulfur species present in the feed gas are hydrogenated over a nickel/molybdenum (NiMo) catalyst in the reactor. NiMo was chosen over the conventional Cobalt Molybdenum catalyst due to its ability to operate over a large temperature range and its higher activity for the olefin hydrogenation reaction. In addition, NiMo is less active for methanation of carbon oxides, which may be present in refinery off gas feeds. The highly exothermic methanation reaction is undesired as it contributes to the exotherm in the hydrogenator reactor, potentially resulting in thermal cracking of the hydrocarbons, or vessel overheating. The hydrogenation of olefins is a moderately exothermic reaction and, as

such, the temperature of the reactor must be carefully controlled as the olefin content of the blended feed gas changes. The temperature at the outlet of the reactor is monitored and controlled with a cascade loop as shown in Figure 5. The hydrogenator operates between 580°F and 680°F inlet temperature depending on the feed gas blend and corresponding amount of olefins. If the outlet temperature on the reactor approaches the design temperature of the vessel, as detected by two out of four temperature transmitters, the feed gas flow to the reactor is shut off.

The hydrogenation reactor is followed by two desulfurizer reactors, containing zinc oxide catalyst, which remove hydrogen sulfide from the feed gas by absorption and reaction. A layer of chloride guard catalyst is also added above the zinc oxide bed to prevent trace amounts of chlorides, that may be present in the refinery gas feeds, from poisoning the primary reforming catalyst. The two desulfurizer reactors cannot be regenerated on-line and therefore are configured in series such that zinc oxide catalyst change out does not require a plant outage.

Steam-Hydrocarbon Reforming

Once purified, the feed gas is mixed with superheated process steam and the mixed feed/steam enters the reformer above 700°F and 550 psig.

Process steam to the reformer is flow controlled to maintain the desired steam to carbon ratio (S/C). A minimum steam to carbon ratio is necessary to avoid carbon formation on the reforming catalyst. An alkali promoted reforming catalyst, loaded into the upper portion of the primary reformer tubes, enables the hydrogen plant to maintain a constant S/C ratio independent of the blend of refinery feed gases being processed. The promoted catalyst and the relatively high concentration of hydrogen in the feed gas enable plant operation at a lower S/C ratio. Designing for this low S/C ratio resulted in capital savings associated with smaller reformer tubes and furnace box. Operationally, the lower S/C ratio also resulted in reduced fuel consumption.

The steam-to-carbon control scheme is configured to maintain a preset ratio when the demand on the reformer changes. The feed gas composition is measured by the on-line gas chromatograph enabling the DCS to calculate a carbon content of the feed. The S/C controller then calculates a setpoint for the steam flow controller based on the operator input setpoint for S/C ratio. A lead lag control scheme ensures steam is in excess at all times. That is, if the plant capacity is to increase, the steam flow rate increases first and only after that the hydrocarbon flow is increased. Conversely, if the plant capacity is to reduce, then the feed rate reduces before the steam flow rate is allowed to reduce.

In the event of a significant loss of steam flow as detected by two out of three voting on the S/C ratio controller, the feed gas valve will be tripped closed to protect the reforming catalyst from carbon formation.

Reformed gas exits the reformer tubes at 1575°F and consists of a mixture of hydrogen, carbon oxides, unreacted methane, steam and nitrogen that was present in the feed. The reformer effluent is cooled by generating 700 psig steam in the process gas boiler. An internal bypass in the boiler controls the temperature of the syngas before it enters the high temperature shift reactor.

The high reformer operating pressure coincident with the reformer outlet temperature of 1575°F results in a high severity design of the primary reformer. The reformer is a Kinetics Technology International Corp. (KTI) top-fired, box type containing over 400 tubes in 10 rows and over 100 burners in 11 rows. KTI's reformer program was used to model the radiant section and provide point-by-point information on the reaction in the tubes and heat transfer in the firebox. The reaction kinetics and advanced heat transfer theory incorporated in the program have been extensively tested against operating installations and adjusted to suit actual experience.

Precise predictions of tubewall temperatures, together with the latest improvements in tube metallurgy, result in thinner catalyst tubes. Micro alloy metallurgy with nominal composition of chrome/nickel and other doping materials was selected for the tubes. This selection will result in longer tube life because of the higher creep rupture strength exhibited by this alloy.

Two layers of reformer catalyst were selected due to the wide range of feed stocks. The top layer is alkali promoted, followed by the conventional reforming catalyst. As previously discussed, the dual catalyst layers allow flexibility in feed gas composition and allow operation at a relatively low steam to carbon ratio which improves efficiency.

The reformer convection section is designed to maximize heat recovery and reduce the flue gas temperature to less than 300°F. The heat recovery in the convection section includes steam generation and superheat as well as combustion air preheat.

Shift Conversion

Most of the carbon monoxide in the reformer effluent is reacted with water to produce carbon dioxide and additional hydrogen in the high and low temperature shift reactors (HTS and LTS). When shift catalyst is new, the HTS Reactor operates with an inlet temperature of 600°F and the LTS Reactor operates at an inlet temperature of 400°F. Process gas leaving the LTS Reactor has a residual carbon monoxide concentration of approximately 0.3 vol% (dry).

Correct adjustment of the HTS Reactor inlet temperature is necessary over time because the exothermic shift reaction equilibrium is favored by low temperature, but the catalyst activity is increased by higher temperature. Thus, an optimum intermediate temperature is selected. As the shift catalyst ages, the inlet temperature is raised to restore activity.

The heat generated by the shift reactions is used to preheat the feed gas and boiler feed water for steam generation. After the hydrogen rich gas is cooled to ambient temperature, it enters the Pressure Swing Adsorption (PSA) unit for final purification.

Hydrogen Purification

Pure hydrogen is recovered from the raw hydrogen gas by the use of an Air Products engineered and supplied pressure swing adsorption unit (PSA). The PSA simultaneously and nearly completely removes all constituents of the gas except hydrogen. Carbon monoxide concentration in the hydrogen is controlled to below 1 ppm(v). Variations in the feed gas composition, as Shell changes the feed gas blend to suit the refinery, have an insignificant effect on the performance of the PSA unit.

The PSA unit included in this design consists of 10 adsorber vessels. Each adsorption vessel follows a cycle of adsorption, stepwise depressuring, purging and stepwise repressuring. The high feed gas compressor discharge pressure enables the PSA adsorption step to occur at 450 psig which is higher than typical PSA feed pressure. This high pressure adsorption results in two major performance advantages. The adsorption of the impurities onto the adsorbent is favored at higher pressures so recovery is increased. Also, the high pressure adsorb step enables the cycle to contain multiple pressure equalization steps. The equalization steps effectively utilize the residual hydrogen in an adsorber vessel at the end of its cycle, to repressurize the other vessels and provide hydrogen for purging.

The purge gas, containing unreacted methane from the reformer and unrecovered hydrogen from the PSA, is used as fuel in the reformer. This fuel provides up to 70% of the total firing duty.

Product Compression

Product hydrogen from the PSA unit is at 450 psig and is then compressed in the multiservice compressor to a pressure of 2000 psig.

Hydrogen is delivered to Shell at three pressure levels.

Low Pressure: Twenty-five percent of the product hydrogen can be supplied at the PSA discharge pressure of 450 psig.

Medium Pressure: Up to 16 MMSCFD hydrogen can be supplied at the compressor interstage pressure of 950 psig.

High Pressure: The full 88 MMSCFD hydrogen product can be supplied at 2000 psig.

Steam Generation

A single steam drum operating at 700 psig serves all the steam generation sources. The plant has a total of 470,000 lbs/hr steam production capacity. Steam is generated by heat recovery from the reformer product stream in the process gas boiler and from the flue gas stream in the convection section. The convection section boilers operate on forced circulation; the process gas boiler is a natural circulation boiler.

Approximately 75% of the boiler feed water requirements are met by importing R.O. quality make-up water from Shell. The remaining 25% is recycled process condensate that is degassed before entering the steam system.

A portion of steam produced is used as process steam in the reforming reaction or as deaerator stripping steam with the remainder exported to Shell. The process, as well as export steam, is superheated against hot flue gases in the convection section of the reformer. The steam drum is designed to generate steam containing less than 0.1 ppm total solids and less than 20 ppb sodium.

Fuel System

An important feature that is built into the design of the fuel system is the utilization of a low BTU gas. The plant can operate with or without the 120 BTU/SCF low btu gas which provides as much as 15% of the total firing requirement. The low BTU fuel is flow controlled and combines with the PSA purge gas. This feature enables Shell to consistently realize fuel value from a stream that would otherwise sometimes be flared.

Most of the remaining fuel requirement is satisfied by combustion of purge gas from the PSA purification unit. The PSA purge gas contains all of the impurities in the gas fed to the PSA unit along with unrecovered hydrogen and makes up about 70% of the firing load. The balance of the heat requirement is made up by firing Shell's refinery fuel gas. This refinery fuel gas is a collection of refinery unit purge streams and can vary in BTU content by nearly 50% in a short period of time. Using advanced control logic, the DCS compensates for the changing BTU content of this fuel and maintains a constant reformer outlet temperature.

Table 3 lists the composition of the low BTU fuel gas and refinery fuel gas.

RELIABILITY FEATURES

The hydrogen and steam products from the plant supplement Shell's own producing units, including a catalytic reformer and existing steam methane reformers. The Air Products plant, however, supplies a large percentage of the refinery's total hydrogen and steam requirement. Therefore, a high level of reliability is designed into the facility.

Voting systems have been used on all shutdown circuits in order to reduce outages due to transmitter failures. In these cases, multiple transmitters are in the same service and in order for an event (such as a trip) to be initiated, two or more of the transmitters must agree. Averaging and deviation alarms are used as appropriate in concert with multiple transmitters.

Two, 75% multiservice compressors are used to compress the feed and hydrogen product. In the event of a compressor failure, the plant controls will automatically fully load the second compressor. LPG feed can then be vaporized and combined with the feed to make up the additional 25% of plant production. In this mode, 25% of the hydrogen product must be delivered to Shell directly off the PSA into the low pressure hydrogen product header within Shell's refinery. By configuring the compression this way, a compressor failure will not curtail hydrogen or steam supply to the refinery. In addition, compressor maintenance can be performed without interrupting delivery to Shell.

One of the most common causes of product curtailments and/or interruptions in a hydrogen plant utilizing PSA purification is switch valve failure. The PSA skids contain very large valves that cycle every few minutes. The DCS monitors every valve throughout the cycle, to ensure that it is in the correct position. When the DCS detects a valve failure, the affected vessel is isolated and the unit transitions into a 9-bed mode of operation. The beds are sized such that once cycle times are adjusted, 100% production can be achieved in 9-bed mode. Solenoid or valve maintenance can be performed by the operator before the 10th vessel is placed back into production. The unit can also be manually switched from 9-bed or 10-bed to 5-bed mode for instances when in-line work is required.

The multiple fuel design offers additional reliability. The refinery fuel gas header normally provides 15% of the reformer firing duty; however, it is designed to provide up to 50% of the firing duty. Should the purge gas/low btu gas header need to be taken out of service, the plant can be operated at 35 MMSCFD hydrogen production rate utilizing refinery fuel gas only. As a result, the loss of the fuel header that normally provides 85% of the reformer firing does not result in a total product interruption.

In the event that one of Shell's units which is providing feed gas to the hydrogen plant trips off-line and that feed is instantaneously lost, the system is designed for natural gas to pressure control the feed gas header. The control system is tuned such that in this event, natural gas makes up for the lost feed stock to the compressors at a rate that does not upset plant operations.

ENVIRONMENTAL FEATURES

The Bay Area Air Quality Management District (BAAQMD) required Best Available Control Technology (BACT) to be used in the hydrogen plant.

The dual fuel burners feature low NOx emissions designs. NOx refers to a mixture of nitrogen oxides formed from the combustion of fuel gas and is considered a pollutant. The burners utilize staged-fuel combustion and flue gas recirculation to minimize NOx production. Staged-fuel combustion involves introducing the fuels at different places to create two combustion zones. NOx is reduced in the first zone due to the low adiabatic flame temperatures resulting from the lean combustion conditions; NOx is reduced in the second zone due to the quenching effect of the inert combustion products from the first zone. Flue gas recirculation reduces NOx by introducing inert combustion products into the combustion zone which reduces flame temperature. The burners were shop tested prior to installation to ensure that CO and NOx emissions guarantees would be met.

Low NOx burner technology has not progressed to the point of being able to meet the strict BAAQMD requirements alone and therefore a SCR (Selective Catalytic Reduction) unit was installed in the reformer convection section. With an SCR, ammonia is vaporized and injected into the flue gas, which then passes over a honeycomb-shaped vanadium pentoxide / titanium dioxide catalyst. The ammonia reacts with the NOx in the presence of oxygen to form nitrogen and water. By varying the ammonia injection rate, NOx is controlled to the desired level.

The hydrogen plant flue gas stack contains a Continuous Emissions Monitoring System (CEMS) to continuously monitor the stack emissions. CEMS electronically generates emissions reports that are required by BAAQMD.

CONCLUSIONS

In conclusion, eight different hydrocarbon streams from Shell's Martinez refinery are sent to Air Product's hydrogen plant as either feed or fuel. Air Products and KTI have tailored the process design of the hydrogen plant to accommodate these multiple feed and fuel streams to produce valuable hydrogen and steam products for Shell. The hydrogen plant control system is designed to minimize the effects of varying feed and fuel composition on hydrogen and steam product quality. The refinery has high reliability and strict environmental goals, which have been addressed through unique design features incorporated into the hydrogen plant.

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Table 1: Refinery Hydrocarbon Feed Streams
(Mole % Dry Basis)

	Feed Gas A	Feed Gas B	Feed Gas C	Feed Gas D	LPG	Natural Gas (NG)
N ₂	3.5	1.3	3.5	2.4	0	1.2
O ₂	0	0.1	0	0	0	0
C ₅ H ₁₂	1.5	1.0	0.1	2.4	0	0
C ₄ H ₁₀	3.0	4.0	0.4	33.4	0.1	0
C ₃ H ₈	6.0	6.0	1.6	27.0	99.8	0
C ₂ H ₆	7.0	15.0	15.4	0.6	0.1	1.9
CH ₄	19.0	36.0	49.3	8.3	0	96.1
CO ₂	0	0.1	0.1	0	0	0
CO	0	0.1	1.3	0	0	0.8
H ₂	60.0	35.0	21.2	25.9	0	0
C ₂ H ₄	0	1.0	4.8	0	0	0
C ₃ H ₆	0	0.3	1.8	0	0	0
C ₄ H ₈	0	0.1	0.5	0	0	0
Total	100.0	100.0	100.0	100.0	100.0	100.0

Table 2: Design Cases

	Feed Gas A (MMSCFD)	Feed Gas B (MMSCFD)	Feed Gas C (MMSCFD)	Feed Gas D (MMSCFD)	LPG (BPD)	Natural Gas (MMSCFD)
Case 1	5.0	16.8	7.1	3.0	0	0
Case 2	5.0	15.3	0	3.0	2050	0
Case 3	16.9	10.8	4.6	3.0	0	0
Case 4	5.0	0	16.3	3.0	0	7.4

Table 3: Fuel Gases

(Mole %)

	Refinery Fuel Gas	Low BTU Fuel Gas
N ₂	7.9	53.47
O ₂ /Ar	0.2	2.11
C ₆ H ₁₄	0.1	0
C ₄ H ₁₀	1.5	0
C ₃ H ₈	3.4	0
C ₂ H ₆	8.9	0.05
CH ₄	46.7	1.30
CO ₂	0.1	6.28
CO	0.8	21.36
H ₂	22.1	14.42
C ₂ H ₄	4.4	0.01
C ₃ H ₆	3.2	0
C ₄ H ₈	0.3	0
H ₂ O	0.4	1.00
Total	100.0	100.00

Figure 1: THE ROAD TO CLEAN MOTOR FUELS

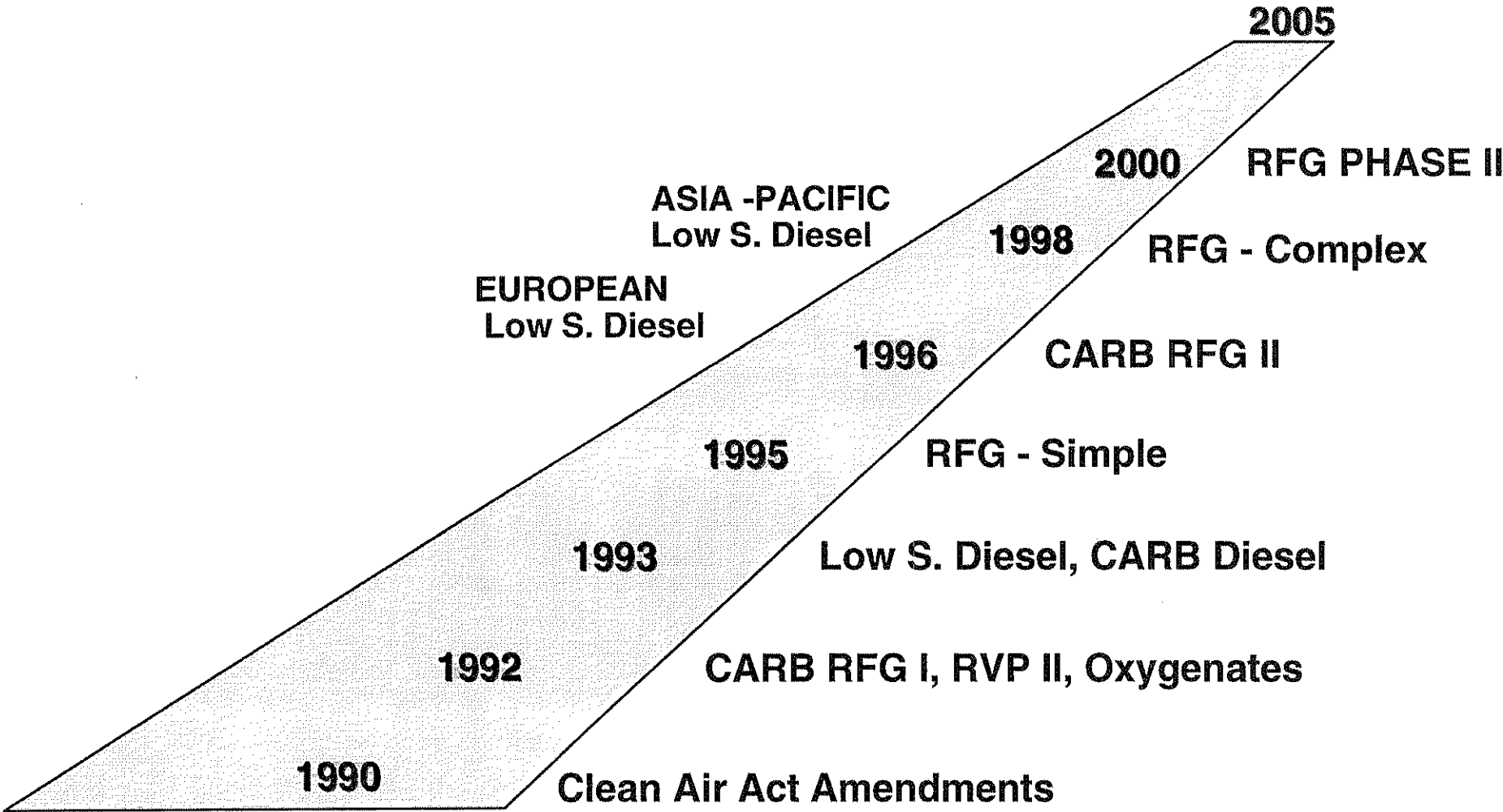


Figure 2: PROCESS FLOW DIAGRAM

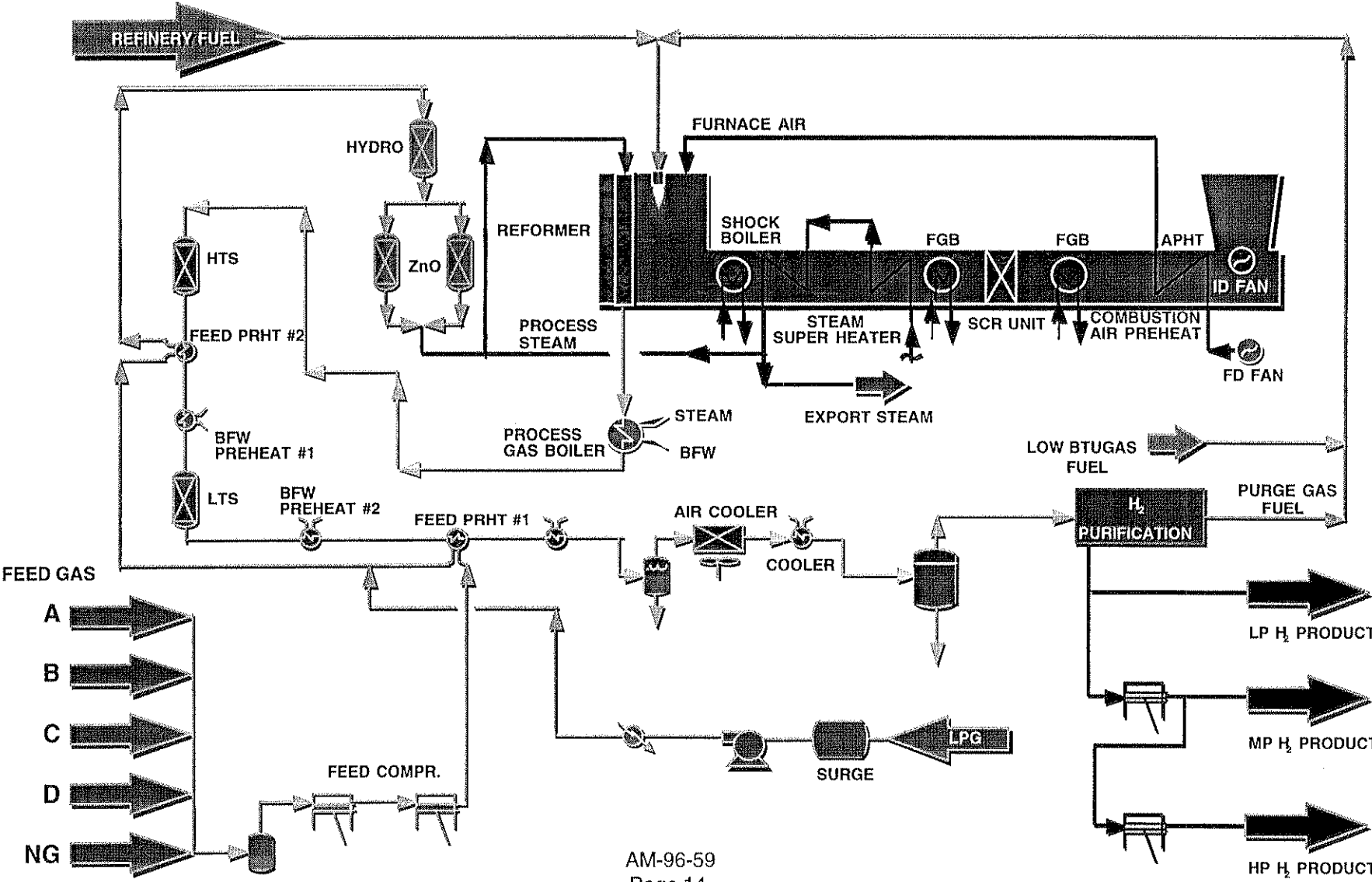


Figure 3: 88 MMSCFD HYDROGEN FACILITY

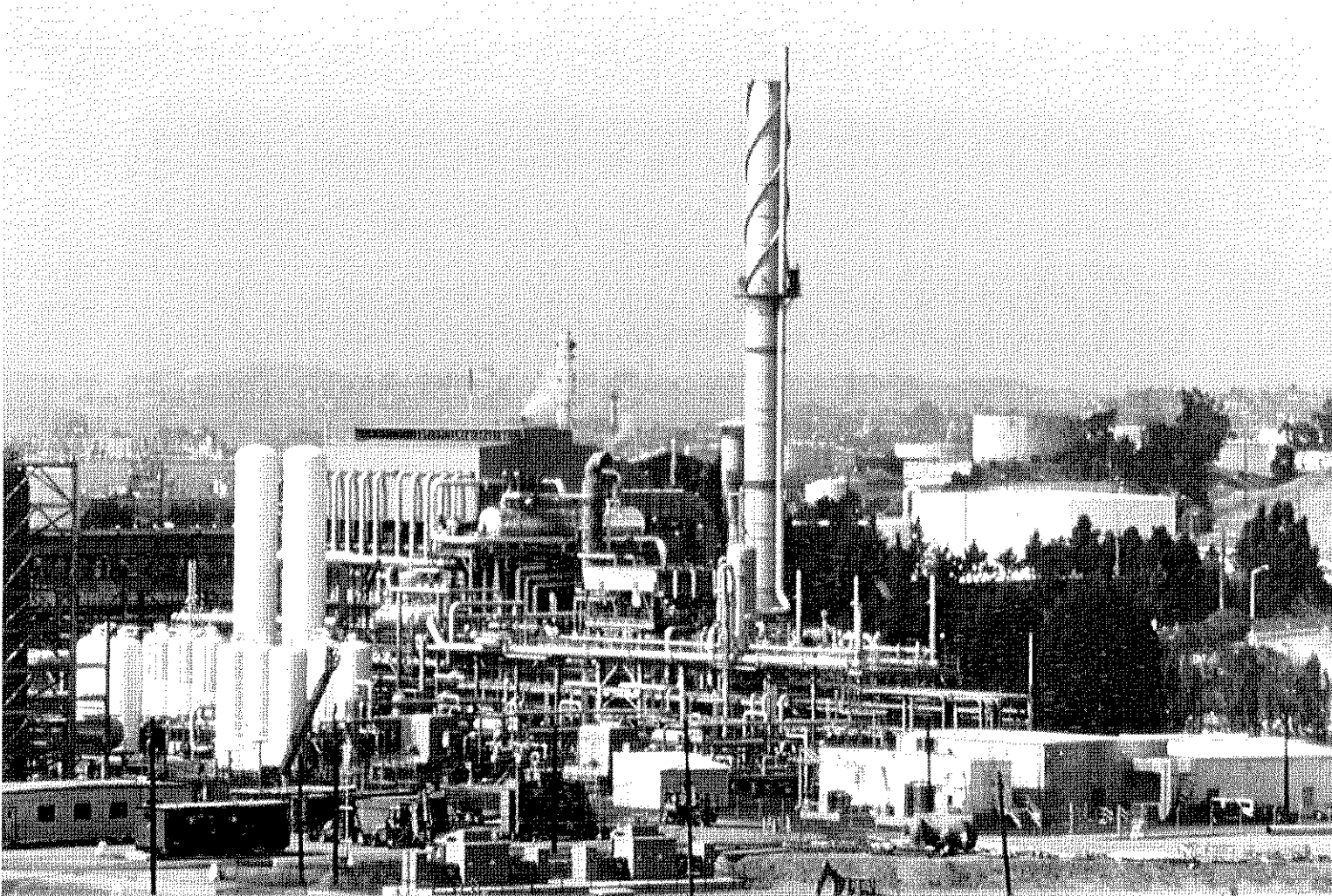


Figure 4: MULTI-SERVICE COMPRESSION SCHEME

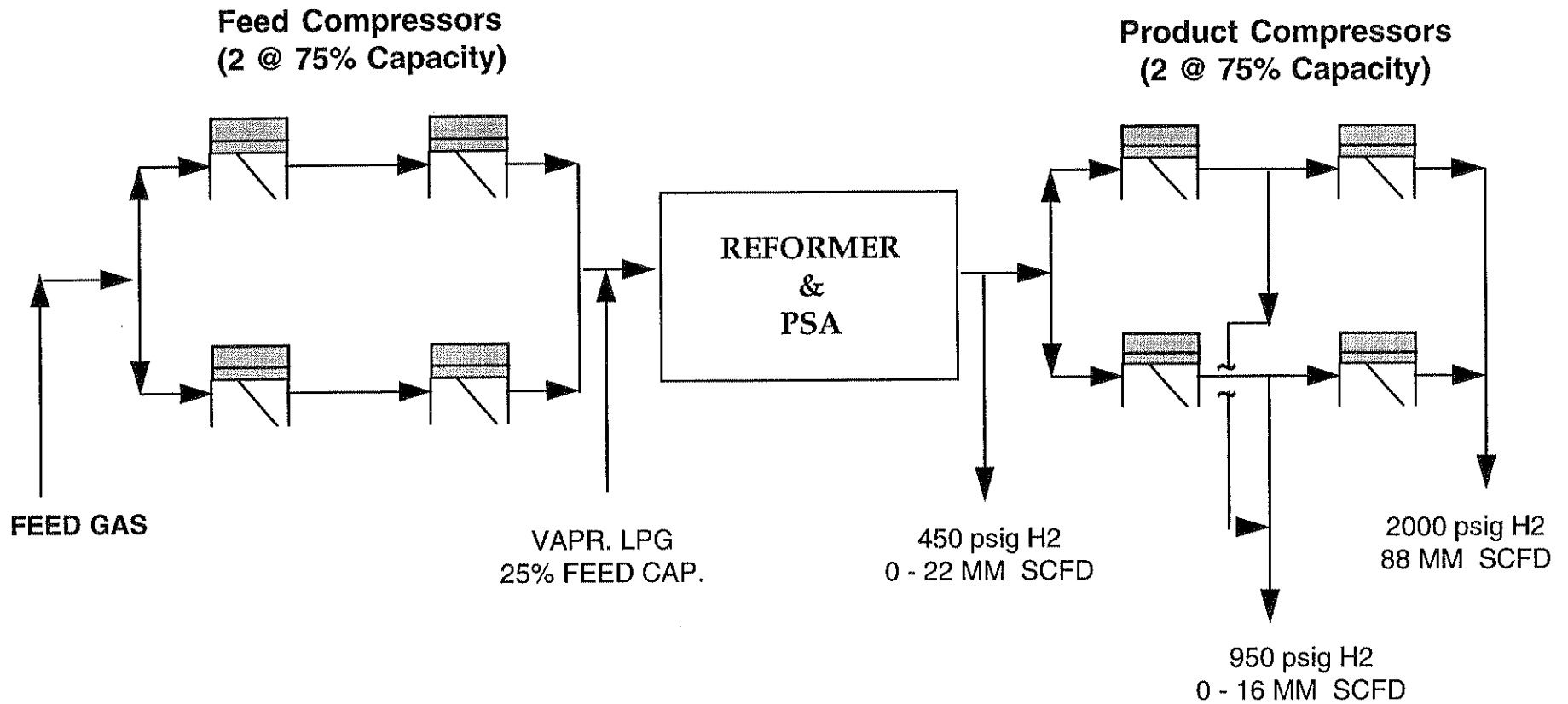


Figure 5: HYDROGENATOR TEMPERATURE CONTROL

