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LIQUEFIED PETROLEUM GAS

Liquefied petroleum gas (LPG) is a subcategory of a versatile class of petroleum products known as natural gas liquids (NGLs) that are produced along with and extracted from natural gas (see Gas, natural). LPG is also produced from the refining of crude oil (see Petroleum). Although LPG is commercially defined as propane [74-98-6], butane [106-97-8], and butane–propane mixtures, commercial availability is primarily limited to propane (see Hydrocarbons). There are two grades of specification propane, propane HD-5 and special-duty propane. The primary difference in the two grades is that the propylene [115-07-1] content of propane HD-5 is restricted to a maximum of 5 vol %. Propylene (qv) is found only in refinery-produced propane. The principal uses of LPGs are as fuels and feedstocks (qv) for the production of motor gasoline and a wide variety of chemicals (see Fuels, synthetic; Gasoline and other motor fuels).

Other natural gas liquids include natural gasoline [8006-61-9], which is composed of the pentanes and heavier components of the natural gas stream, and ethane [74-84-0]. Most recently ethane has become the principal product of natural gas processing plants.

1. Properties

In general, LPG specifications involve limits for physical properties. Consequently, the composition of the commercial-grade products varies between wide limits. Physical properties of the principal components of LPG are summarized in Table 1 (1).

2. Manufacture and Processing

LPG recovered from natural gas is essentially free of unsaturated hydrocarbons, such as propylene and butylenes (qv). Varying quantities of these olefins may be found in refinery production, and the concentrations are a function of the refinery's process design and operation. Much of the propylene and butylene are removed in the refinery to provide raw materials for plastic and rubber production and to produce high octane gasoline components.

LPG is recovered from natural gas principally by one of four extraction methods: turboexpander, absorption (qv), compression, and adsorption (qv). Selection of the process is dependent on the gas composition and the degree of recovery of ethane and LPG, particularly from large volumes of lean natural gas.

2.1. Turboexpander Process

Ethane has become increasingly desirable as a petrochemical feedstock resulting in the construction of many plants that recover the ethane from natural gas at -73 to $-93^{\circ}\mathrm{C}$. Combinations of external refrigeration and liquid flash-expansion refrigeration with gas turboexpansion cycles are employed to attain the low temperatures

Table 1. Physical Properties of LPG Components[a]

Component	CAS Registry Number	Molecular formula	Boiling point, 101.3 kPa,[b] °C	Vapor pressure, 37.8°C, kPa[b]	Liquid density, g/L[c]
ethane	[74-84-0]	C_2H_6	−88.6		354.9
propane	[74-98-6]	C_3H_8	−42.1	1310	506.0
isobutane	[75-28-5]	C_4H_{10}	−11.8	498	561.5
n-butane	[106-97-8]	C_4H_{10}	−0.5	356	583.0
1-butene	[106-98-9]	C_4H_8	−6.3	435	599.6
cis-2-butene	[590-18-1]	C_4H_8	3.7	314	625.4
trans-2-butene	[624-64-1]	C_4H_8	0.9	343	608.2
n-pentane	[109-66-0]	C_5H_{12}	36.0	107	629.2

[a] Ref. 1.
[b] To convert kPa to psi, multiply by 0.145.
[c] At saturation pressure.

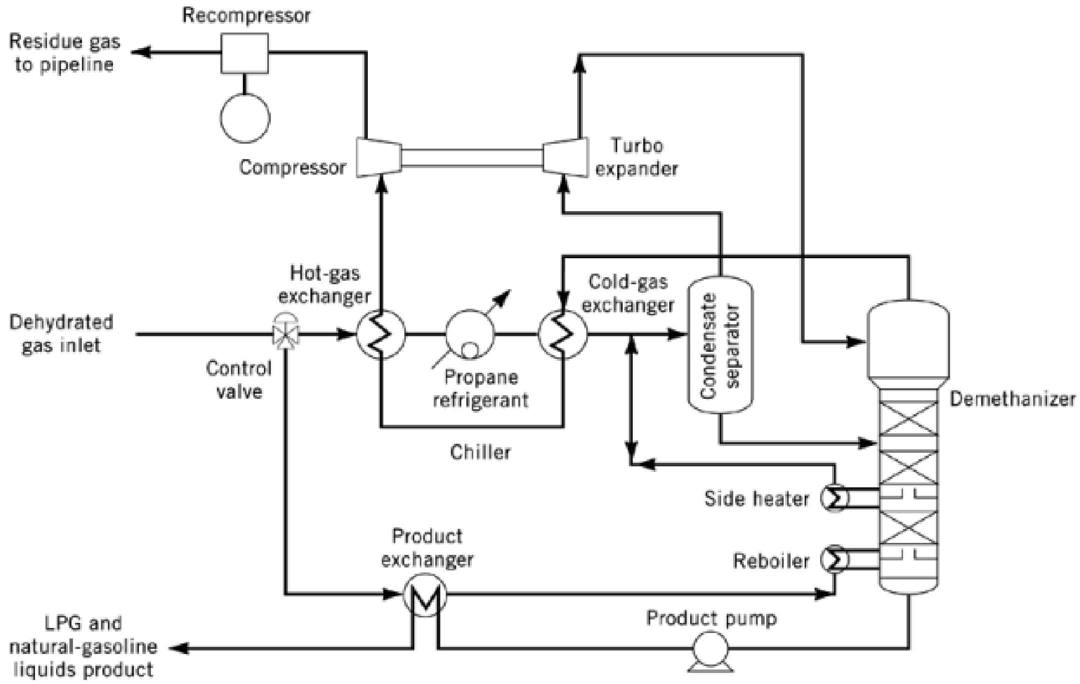

Fig. 1. One-expand cycle with external refrigeration for high ethane recovery in the hydrocarbon liquid product.

desired for high ethane recovery. Figure 1 is a flow diagram of a one-expander cycle having external refrigeration (see Refrigeration).

Dry inlet gas that has been dehydrated by molecular sieves (qv) or alumina beds to less than 0.1 ppm water is split into two streams by a three-way control valve. Approximately 60% of the inlet gas is cooled by heat exchange with the low pressure residue gas from the demethanizer and by external refrigeration. The remainder of the inlet gas is cooled by heat exchange with the demethanized bottoms product, the reboiler, and the side heater. A significant amount of low level refrigeration from the demethanizer liquids and the cold residue gas stream is recovered in the inlet gas stream.

The two portions of the feed stream recombine and flow into the high pressure separator where the liquid is separated from the vapor and is fed into an intermediate section of the demethanizer with liquid level control. The decrease in pressure across the level-control valve causes some of the liquid to flash which results in a decrease in the stream temperature. The pressure of the vapor stream is decreased by the way of a turboexpander to recover power, thus to achieve more cooling than would be possible by Joule-Thompson expansion. The outlet of the turboexpander then is fed into the top of the demethanizer where the separation of liquid and vapor occurs. The vapor is passed as cold residue to the heat exchanger and the liquid is distributed to the demethanizer top tray as reflux.

Essentially all of the methane [74-82-8] is removed in the demethanizer overhead gas product. High recovery of ethane and heavier components as demethanizer bottoms products is commonplace. The work that is generated by expanding the gas in the turboexpander is utilized to compress the residue gas from the demethanizer after it is warmed by heat exchange with the inlet gas. Recompression and delivery to a natural gas pipeline is performed downstream of the plant. A propane recovery of 99% can be expected when ethane recoveries are in excess of 65%.

Recoveries of 90–95% ethane have been achieved using the expander processes. The liquid product from the demethanizer may contain 50 liquid vol % ethane and usually is delivered by a pipeline to a central fractionation facility for separation into LPG products, chemical feedstocks, and gasoline-blending stocks.

2.2. Absorption

Oil absorption is another process used for recovery of LPG and natural gas liquids from natural gas. Recovery is enhanced by lowering the absorption temperature to –45°C and by keeping the molecular weight of the absorption oil down to 100. Heat used to separate the product from the absorption oil contributes to the cost of recovery. Therefore, this process has become less competitive as the cost of energy has increased. A simplified flow diagram of a typical oil-absorption process is shown in Figure 2.

The natural gas feed exchanges heat with the residue gas from the absorber overhead. Ethylene glycol [107-21-1] is injected as an antifreeze and the stream is cooled further by refrigeration to –37°C. The gas from the cooler enters a glycol separator where the glycol that contains water is separated from the natural gas as a liquid phase. A dry glycol is recovered for recycling to the injection point by distilling the water. The gas and any higher boiling hydrocarbons that are present pass to the base of the absorber where the gas comes into contact with absorption oil which enters at the top of the absorber at $-37^{\circ}\mathrm{C}$. Approximately 85% of the propane and essentially all of the higher boiling hydrocarbons are absorbed in the oil. The overhead residue gas from the absorber ($34^{\circ}\mathrm{C}$) is heat exchanged with the inlet gas; at 4°C and 4.24 MPa (600 psig), the gas flows to the booster where the pressure is increased to that of the natural gas line.

The rich oil from the absorber is expanded through a hydraulic turbine for power recovery. The fluid from the turbine is flashed in the rich-oil flash tank to 2.1 MPa (300 psi) and –32°C. The flash vapor is compressed until it equals the inlet pressure before it is recycled to the inlet. The oil phase from the flash passes through another heat exchanger and to the rich-oil deethanizer. The ethane-rich overhead gas produced from the deethanizer is compressed and used for producing petrochemicals or is added to the residue-gas stream.

The bottoms, consisting of absorption oil, absorbed propane, and higher boiling hydrocarbons, are fed to the lean-oil fractionator. The LPG and the natural gas liquids are removed as the overhead product from the absorption oil which is removed as a kettle-bottom product.

The lean oil from the lean-oil fractionator passes through several heat exchangers and then through a refrigerator where the temperature is lowered to $-37^{\circ}\mathrm{C}$. Part of the lean oil is used as a reflux to the lower section of the rich-oil deethanizer. Most of the lean oil is presaturated in the top section of the deethanizer, is cooled again to $-37^{\circ}\mathrm{C}$, and is returned to the top of the absorber, thus completing the oil cycle.

The overhead product from the lean-oil fractionator, consisting of propane and heavier hydrocarbons, enters the depropanizer. The depropanizer overhead product is treated to remove sulfur and water to provide

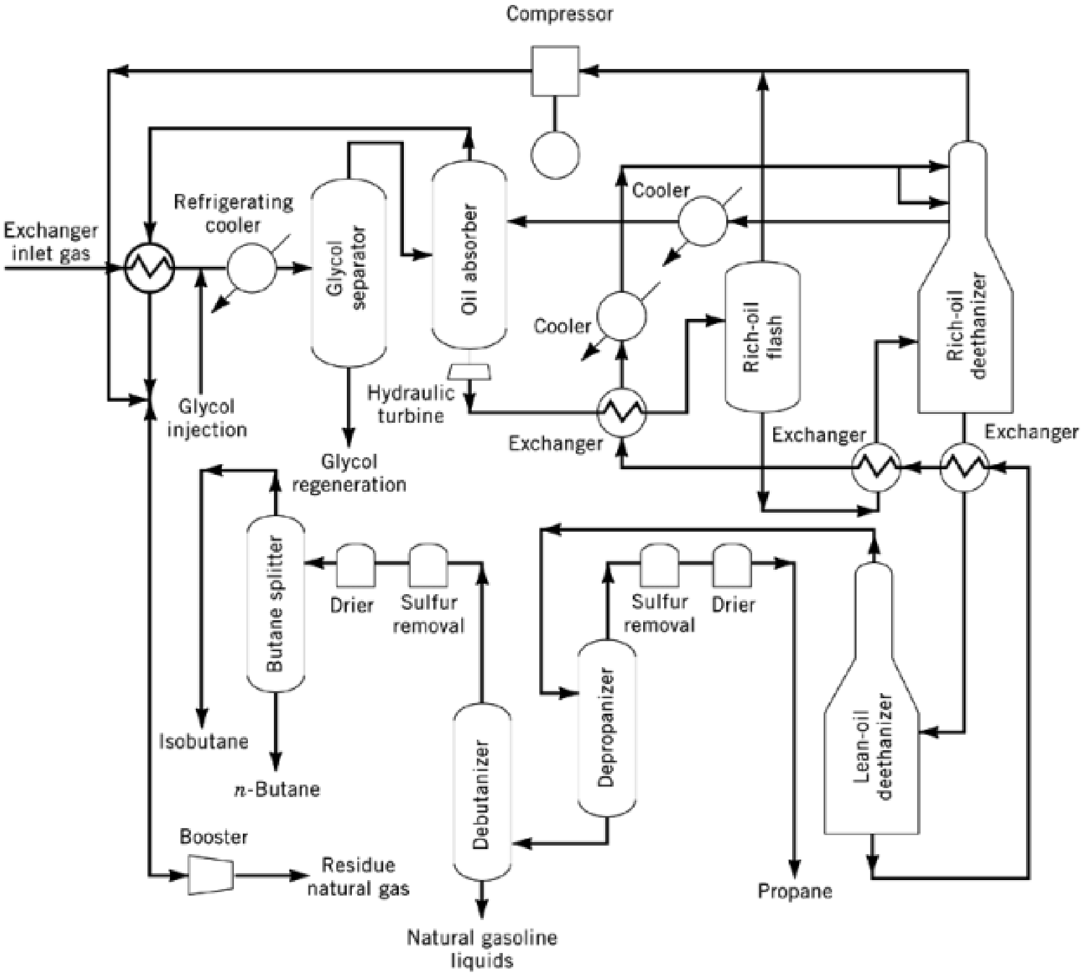

Fig. 2. Refrigerated absorption process for the production of LPG and natural gasoline liquids.

specification propane. The depropanizer bottoms, containing butane and higher boiling hydrocarbons, enters the debutanizer. Natural gasoline is produced as a bottom product from the debutanizer. The debutanizer overhead product is mixed butanes, which are treated for removal of sulfur and water, then fed into the butane splitter. Isobutane is produced as an overhead product from the splitter and *n*-butane is produced as a bottoms product.

2.3. Compression

Compression is the simplest and the least effective of the four recovery methods. It was the first process used for the recovery of hydrocarbon liquids from natural gas but is used only in isolated cases. The most significant application of the compression process is for gas-cycling plants where the natural gas liquids are removed and the remaining gas is returned to the production formation. Figure 3 is a schematic of a typical gas-cycle plant.

The pressure used in producing gas wells often ranges from 690– 10,300 kPa (100–1500 psi). The temperature of the inlet gas is reduced by heat-exchange cooling with the gas after the expansion. As a result of the

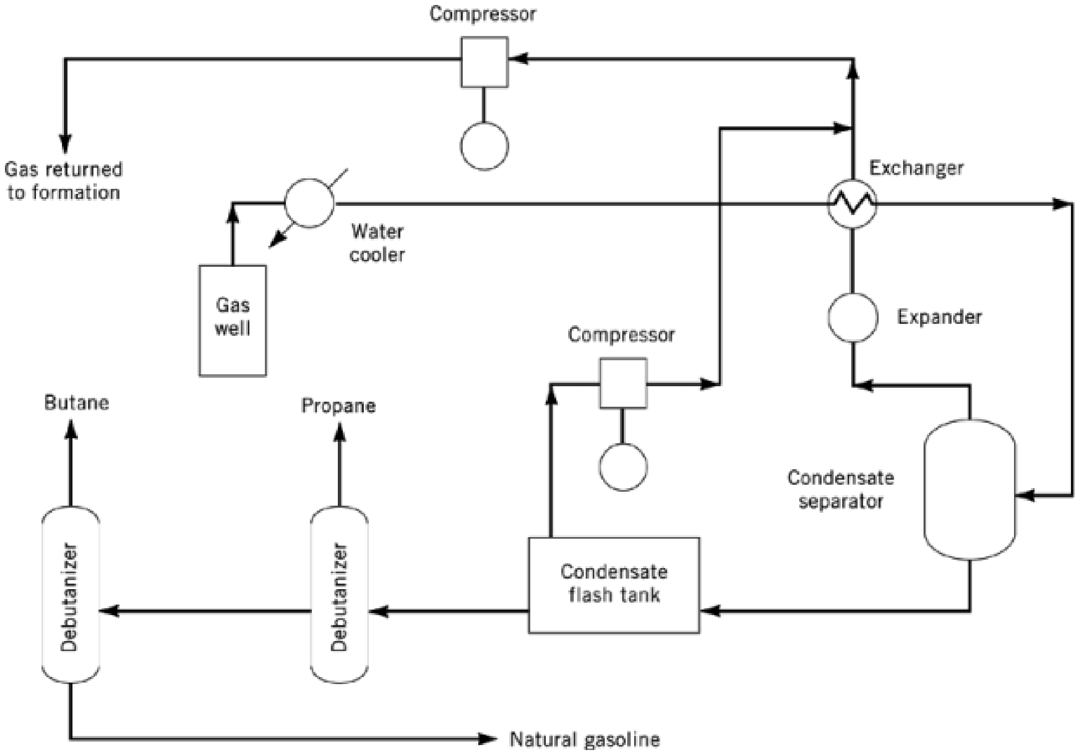

Fig. 3. High pressure gas-cycling plant with compression.

cooling, a liquid phase of natural gas liquids that contains some of the LPG components is formed. The liquid is passed to a set of simple distillation columns in which the most volatile components are removed overhead and the residue is natural gasoline. The gas phase from the condensate flash tank is compressed and recycled to the gas producing formation.

Condensable liquids also are recovered from high pressure gas reservoirs by retrograde condensation. In this process, the high pressure fluid from the reservoir produces a liquid phase on isothermal expansion. As the pressure decreases isothermally the quantity of the liquid phase increases to a maximum and then decreases to disappearance. In the production of natural gas liquids from these high pressure wells, the well fluids are expanded to produce the optimum amount of liquid. The liquid phase then is separated from the gas for further processing. The gas phase is used as a raw material for one of the other recovery processes, as fuel, or is recompressed and returned to the formation.

2.4. Adsorption

Adsorption processes have been used to recover hydrocarbons that are heavier than ethane from natural gas. Although the adsorption process has applications for the recovery of pentane and heavier hydrocarbons from lean gas, the percentage recovery of LPG components in these plants usually is low compared to the normal recovery of LPG in modern turboexpander or oil-absorption plants.

A simplified flow diagram for the fast-cycle adsorption plant is shown in Figure 4. Activated carbon, alumina gel, and silica gel are used as adsorbents. Use of internal insulation in the adsorption towers affords less cycle time. The complete process cycle consists of three phases: regeneration, cooling, and adsorption.

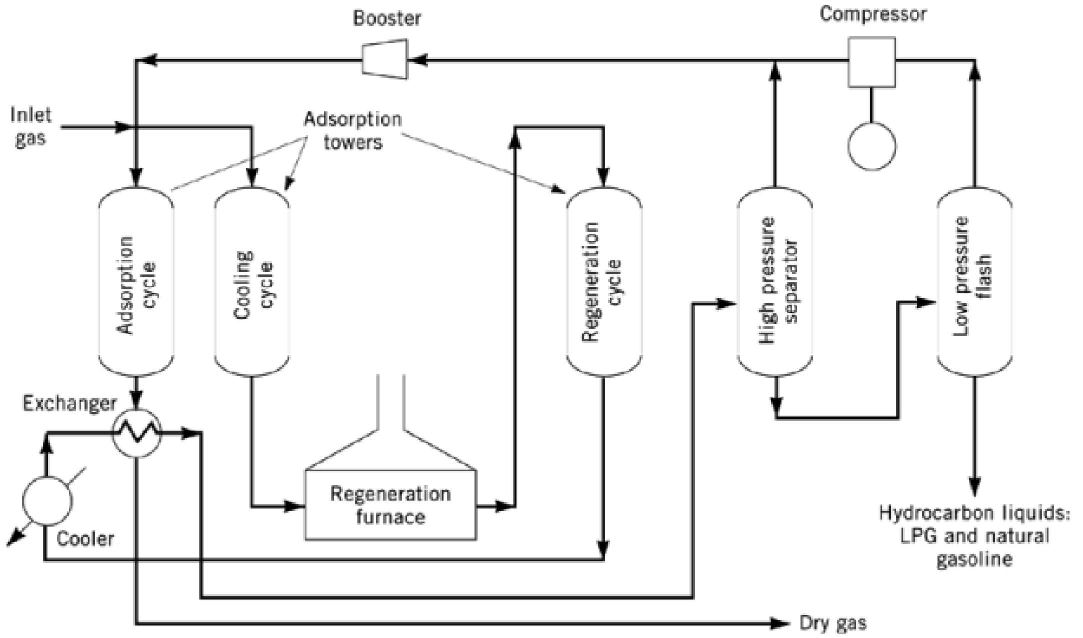

Fig. 4. A fast-cycle absorption unit for recovery of hydrocarbon liquids from natural gas.

In Figure 4, the inlet gas is divided. The larger portion flows directly to the bed in the adsorption cycle; the smaller portion flows first through the bed in the cooling phase of the cycle, then through the regeneration cycle. The effluent gas from the cooling cycle is heated further to 260–315°C in a separate fired furnace before it is injected into the bed in the regeneration cycle. The effluent from the regeneration cycle contains the condensable hydrocarbons that have been stripped from the adsorbent and that are removed in the high pressure separator after the gas that is leaving the regeneration cycle is cooled. The liquid from the high pressure separator is flashed through to remove light hydrocarbons. The flash vapor is compressed and is mixed with the vapor from the high pressure separator. The pressure on the compressed gas stream is boosted to the inlet gas pressure and the gas is recycled to the feed to the adsorption cycle. The liquid from the low pressure flash, which contains LPG and natural gas liquid components, is processed further in a series of distillation towers to produce propane, butanes, and natural gasoline.

Less propane and butanes are produced compared to natural gas liquids by the adsorption process than are obtained normally for the same gas by the oil-absorption process. Because adsorption efficiency increases with a decrease in temperature, the adsorption cycle should operate at the lowest temperature that is economically feasible.

2.5. Purification

The LPG generally requires treatment for removal of hydrogen sulfide [7783-06-4], H_2S, organic sulfur compounds, and water in order to meet specifications. Several methods are used.

2.5.1. Amine Treatment

The LPG is brought into contact with a 15–20 wt % solution of ethanolamine [141-43-5] in water, which removes H_2S to specification levels. The foul amine solution that contains the H_2S is regenerated in a stripper at low pressure using indirect stream stripping, and the stripped amine solution is returned to the LPG contactor. The amine solution generally has little affect on organic sulfur compounds, although diethanolamine [111-42-2] (DEA) and diglycolamine [929-06-6] (DGA) have been used to obtain acceptable carbonyl sulfide [463-58-1], COS, levels in propane.

2.5.2. Caustic Treatment

Amine treatment may be followed by a caustic treatment step in which the LPG is brought into contact with 10–20 wt % caustic solution to remove any residual H_2S and to remove mercaptans. The mercaptans may be stream-stripped from the caustic solution, after which the stripped caustic is recycled to the LPG caustic contactor. Caustics that contain H_2S must be discarded. Various promoters may be added to the caustic to improve the efficiency of the mercaptan removal.

2.5.3. Coalescing

Sand towers or cartridge-type coalescers may be used to separate any undissolved water from the LPG. Removal of the undissolved water meets the specification moisture limit for butanes. However, this step does not produce specification propane.

2.5.4. Solid-Bed Dehydration

Silica gel, bauxite, activated alumina, or molecular sieves can be used for removing dissolved water to meet propane specifications. The solid-bed dehydrators are used in a cyclic adsorption process. After an adsorption cycle has completed, the bed is heated with a purge gas or a vaporized liquid-product stream for regeneration. If the latter is used, the liquid product is condensed, separated from the free water, and returned to the process. After the beds are regenerated, they are cooled and returned to the adsorption cycle.

2.5.5. Molecular Sieve Treatment

Molecular sieve treaters can be designed to remove H_2S, organic sulfur compounds (including carbonyl sulfide), and water in one step. Solid-bed units are utilized and regeneration occurs in the same manner as simple, solid-bed dehydrators.

2.5.6. Solid-Bed Caustic Treatment

Solid-bed caustic units utilizing methanol [67-56-1] injection into the LPG feed stream can be used for carbonyl sulfide removal. The methanol–caustic solution must be drained periodically from the beds and discarded. When the solid bed is exhausted, the spent caustic must be discarded and replaced. The LPG from the treater has a low enough water content to meet the propane specification.

2.5.7. Fractionation

Direct fractionation also can be used to remove dissolved water from LPG. The water-rich overhead vapor from the dryer fractionator is returned to the fractionator as reflux and the water phase is discarded. A dry LPG product that meets either propane or butane water specifications is produced as a kettle product from the fractionator.

3. Production and Shipment

Historically, about two-thirds of the LPG produced in the United States came from natural gas processing and one-third was produced from refinery operations (2). In 1991, this ratio was 61% from natural gas processing and 39% from refinery operations. Total production of LPG in 1991 was 76.85×10^6 m^3 (294.19×10^6 bbl) from natural gas processing and 30.08×10^6 m^3 (189.23×10^6 bbl) produced from refinery operations.

Table 2. 1991 U.S. NGL Supply and Demand, 10^6 m^3[a]

Places	Ethane	Propane	n-Butane	i-Butane	Pentanes[b]	Total
Supply						
gas plants	30.74	28.28	8.72	9.78	18.79	96.30
refineries	1.49	25.27	4.56	0.25		31.57
imports	0.48	5.29	2.04	0.73	1.31	9.85
from (to) stocks[c]	0.35	−0.27	0.06	0.28	−0.18	0.23
Total	*33.05*	*58.57*	*15.37*	*11.04*	*19.92*	*137.95*
Demand						
chemicals	32.09	24.36	5.23		4.81	66.49
RES and COM[d]		20.16	0.00			20.16
engine fuel		2.05	0.00			2.06
industrial	0.05	4.61	1.70			6.36
utility	0.61	0.26	0.02			0.89
gasoline			5.53	11.04	12.93	29.50
farm		4.29	0.00			4.29
export	0.01	1.61	0.80		0.03	2.45
other	0.30	1.23	2.08		2.15	5.76
Total	*33.06*	*58.57*	*15.37*	*11.04*	*19.91*	*137.95*

[a] To convert m^3 to bbl, divide by 0.159.
[b] Includes C_x compounds where $x \geq 5$.
[c] Negative sign indicates supply to stocks (inventory) in order to balance supply/demand.
[d] RES and COM = research and commercial.

Ethane production for 1991 was 30.74×10^6 m^3 (193.32×10^6 bbl) from natural gas operations and 1.49×10^6 m^3 (9.34×10^6 bbl) from refinery operations for a total production of 32.21×10^6 m^3 (202.66×10^6 bbl). A summary of total natural gas liquids (NGL) including LPG supply and demand in the United States is shown in Table 2.

The progress of LPG utilization has been closely related to progress in transportation (qv) and storage of this fuel. Large volumes of LPG must be transported from the producing plants to centers of consumption, and transportation costs are a principal factor in the cost of LPG to the consumer. Large volumes of LPG usually are transported by high pressure pipelines (qv). As of this writing, this use of pipelines is increasing rapidly. Large quantities of LPG are transported in railroad tank cars which have an average capacity of 113.5 m^3/car (3×10^4 gal/car), although this use is decreasing. Tank-truck transports having capacities of about 17.8 m^3 (10^4 gal) also move large quantities of product from producing plants to distribution centers and from the pipeline terminals to points of distribution or consumption. Delivery trucks having capacities from 3.8–11.4 m^3 (1000–3000 gal) generally are used for the final delivery from the distribution bulk plant to the storage plants of the larger consumers. Smaller quantities are shipped in metal cylinders having capacities from 9–45 kg LPG. These cylinders are filled at the distribution bulk plant and are delivered by truck to the consumer or to the cylinder dealer or cylinders may be filled for the customer at small distribution stations.

Tankers and barges are also used for transporting LPG. Tankers and barges are designed for both high pressure ambient temperature and for low pressure refrigerated transportation. For larger volumes and long distances, low pressure refrigerated tankers almost always are used (2).

Ethane usually is transported in high pressure pipelines from the point of production to the point of consumption. However, for small quantities of ethane, multitube trunk trailers are used.

Table 3. Sales of LPG 1984–1991, 10^6 m^3[a, b]

Year	Chemical	Residential and commercial heating	Engine fuel	Industrial	Utility gas	Motor fuel blending	Farm	Export	Other	Total
1984	47.49	15.05	2.80	10.06	1.45	35.71	3.77	2.93	11.82	2,115.08
1985	51.57	16.97	2.78	7.66	0.91	37.09	4.38	3.72	8.90	2,118.98
1986	48.72	16.62	2.48	8.23	2.73	36.36	4.29	2.59	2.90	2,110.92
1987	54.45	18.39	2.39	8.35	0.54	37.69	4.08	2.33	2.96	2,118.17
1988	60.24	18.25	2.21	8.19	1.23	35.15	4.03	2.96	3.97	2,124.23
1989	55.52	20.41	2.20	9.90	1.14	33.26	4.44	2.37	6.74	2,123.99
1990	57.57	18.84	2.01	5.80	1.06	32.71	4.30	2.16	3.78	2,118.23
1991	66.49	20.16	2.06	6.36	0.89	29.50	4.29	2.45	5.76	2,128.95

[a] Ref. 2.
[b] To convert m^3 to bbl, divide by 0.159.

4. Economic Aspects

The production and consumption of LPG in the United States increased dramatically from its early beginnings in the 1930s until the international energy crises of the 1970s when rising prices and regulatory restraints resulted in reduced domestic production. However, total consumption, including imports, resumed a modest growth characteristic after that time. In 1984, total LPG consumption in the United States was 76.58×10^6 m (481.71×10^6 bbl); by 1991, total consumption was 84.98×10^6 m^3 (534.50×10^6 bbl). The principal growth segment has been the increasing use of LPG for petrochemical feedstocks as can be seen from Table 3.

5. Specifications and Standards

Specifications for the principal LPG products are summarized in Table 4. Detailed specifications and test methods for LPG are published by the Gas Processor's Association (GPA) (3) and ASTM (4). The ASTM specification for special-duty propane and GPA specification for propane HD-5 apply to propane that is intended primarily for engine fuel. Because most domestic U.S. LPG is handled through copper tubing, which could fail if corroded, all products must pass the copper strip corrosion test. A test value of No. 1 represents a LPG noncorrosive to the copper.

6. Storage

Large volumes of LPG are stored to meet peak demand during cold seasons. LPGs are both volatile and flammable and must be stored and handled in special equipment. Standards for storing and handling LPG are published by the National Fire Protection Association (5) and API (6).

Four main types of storage are used: high pressure storage above ground, low pressure refrigerated storage above ground, frozen earth storage, and underground cavern storage. The capacities of the storage unit vary from 500 mL pressure cylinders to 1.9×10^6 m^3 (500×10^6 gal) underground storage caverns. Economic factors determine the proper storage for any given requirement.

Above ground pressure-storage tanks usually are designed for a 1720 kPa (250 psi) working pressure for propane and 860 kPa (125 psi) for butane. Refrigerated, aboveground storage tanks usually are designed for a

Table 4. Commercial Liquefied Petroleum Gas Specifications[a, b]

Parameter	Propane	Butane	Butane–propane mixture	Propane HD-5
composition	predominantly propane and/or propylene	predominantly butane and/or butylenes	butanes and/or butylenes plus propane and/or propylene	$\geq$90 liquid vol % propane, $\leq$5 liquid vol % propylene
vapor pressure at 37.78°C, kPa[c]	1434	482.6	1434	1434
temperature at 95% evaporation,[d] °C	−38.3	2.2	2.2	−38.3
total sulfur, ppmw	185	140	140	123
moisture	pass			pass
free water		none	none	

[a] Refs. 3 and 4.

[b] Corrosion copper strip test, maximum = No.1·

[c] To convert kPa to psi, multiply by 0.145.

[d] Values are maximum.

few kilopascals of pressure. These tanks must be coupled with refrigeration systems to cool the product that is to be stored to a temperature equal to the product's boiling point at the operating pressure of the tanks. Vapors generally are recondensed by refrigeration and returned to the tanks.

In frozen earth storage of propane, the walls and bottom of a pit in the ground are frozen and a dome is constructed over the pit. The pressure in the storage cavern is maintained at nearly atmospheric pressure by refrigeration systems that cool the product to its boiling point at storage pressure. Heat leaks into the cavity and vaporizes some of the propane. The vapor that is formed is compressed, cooled, and returned to the pit as a liquid by the refrigeration system. Because this storage must operate at temperatures considerably below the freezing point of wet earth and at atmospheric pressure, it cannot be used for butane storage.

Underground storage caverns, which operate at approximately formation temperatures and at the corresponding LPG vapor pressure, may be either mined underground storage caverns or cavities that have been produced in a salt formation by solution mining. The underground caverns must be of sufficient depth to develop an overburden pressure greater than the vapor pressure of the stored liquid. Mined storage caverns are 60–152 m deep, whereas salt formation caverns may be from 106–1524 m deep. Underground as compared to aboveground storage is much more economical for storage of large volumes, ie, more than 2785 m^3 (10^7 gal) of LPG. A washed-out salt cavern costs only 10–50% as much to develop as typical mined cavern storage. In 1987, underground storage capacity for LPG in the United States was 79×10^6 m^3 (493×10^6 bbl) (2) and 90% of this was in salt formations.

7. Uses

About 35% of total U.S. LPG consumption is as chemical feedstock for petrochemicals and polymer intermediates. The manufacture of polyethylene, polypropylene, and poly(vinyl chloride) requires huge volumes of ethylene (qv) and propylene which, in the United States, are produced by thermal cracking/dehydrogenation of propane, butane, and ethane (see Olefin polymers; Vinyl polymers).

Residential and commercial fuel demands represent about 24% of total U.S. LPG consumption. Although this market demand is weather dependent, it has assumed the characteristics of a mature market. Growth is related to the general economic trends.

Nearly two-thirds of total butane supply, about 20% of total LPG, is consumed in the manufacture of motor gasoline. However, the environmental mandates of the early 1990s have had a negative impact on this market segment. These mandates have reduced gasoline volatility requirements, effectively reducing the value of the butanes as blending stocks. However, normal butane can be used as feedstock for production of isobutylene, a key ingredient of ether blendstocks, such as methyl *tert*-butyl ether [1634-04-4] (MTBE) for motor gasoline. Shifts in U.S. use patterns can be seen in Table 3.

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Fuels, synthetic; Gas, natural; Vinyl polymers; Refrigeration